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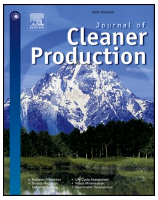

Dairy processing sludge and co-products: A review of present and future re-use pathways in agriculture

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ABSTRACT

The dairy industry is one of the largest global producers of wastewater and generates huge volumes of dairy processing sludge (DPS). There are two main types of DPS, lime-treated dissolved air floatation sludge and biochemically-treated activated sludge. These sludge types may also be converted to STRUBIAS (STRUvite, BIochar, AShes) products which have potential as fertilizers, secondary feedstocks for phosphate fertiliser granules, and soil amendments. A small number of studies indicate that these products have variable nutrient and metal contents, which differ across sludge and STRUBIAS product types. This is due to many factors such as the type of dairy plants, wastewater treatment process and production technologies. Although such products are land applied, the phosphorus (P) and nitrogen (N) fertilizer equivalency value (FEV) are often unknown and not factored into application rates, and therefore need study under field conditions (across soil and crop types). This review identifies a need to quantify antimicrobial drugs, hormones, pesticides, disinfectants, persistent organic pollutants (POPs), microplastics and nano-particles in all DPS and STRUBIAS types. Where detected, testing should follow the transfer of these contaminants to the soil, crop and water continuum. Further knowledge in the areas identified would enable both agronomic and environmental goals to be met and promote higher uptake of DPS and STRUBIAS re-use in agriculture.

1. Introduction

In the aftermath of the COVID-19 pandemic and the associated economic downturn, the world's food system will be under threat and must become more sustainable and resilient (EC, 2020). The recently published Farm to Fork Strategy of the European Union (EU) aims to accelerate the transition to a sustainable food and agriculture system (EC, 2020). One of the recommended practices is to reduce excess fertilisation and to foster the recycling of nutrients from different kinds of organic waste as fertilisers. This will contribute to the delivery of the "zero pollution ambition" of the EU Green Deal (EC, 2020). The European Commission (EC) has recently revised the EU Fertiliser Regulation (EC, 2019), expanding its scope to include secondary-raw-material-based fertilising products to support the shift to sustainable agriculture and a "circular economy" (Huygens et al., 2018). In particular, the EU needs safe recycling sources of phosphorus (P), as Europe lacks natural phosphate rock deposits and mainly depends on imported P. Exploring alternatives to mineral P fertilisers and increased recycling of P may substantially contribute to the reduction of demand for fossil P resources and the dependency on the importation of P from other countries (Arenas-Montaño et al., 2021).

The reuse of raw materials that are now disposed of as waste is one of the key principles of sustainable agriculture and the circular economy. As one of the largest agricultural sectors in the EU (Augère-Granier, 2018), the dairy industry is now considered to be the largest global industrial food wastewater source and one of the main sources of P-rich industrial effluents (Kolev Slavov, 2017; Erkan et al., 2018). To meet discharge limits, dairy wastewater must be treated before discharge. It can be either discharged along with other wastewaters into municipal wastewater treatment plants (WWTPs) or treated on site if dairy plants have their own WWTP. As conventional wastewater treatment systems are used, a large volume of solid organic wastes is generated. These are referred to as dairy processing sludge (DPS) when the dairy wastewater is treated on site (Ashekuzzaman et al., 2019a).

According to current practices in the EU, DPS is categorised as a biosolid (Pankakoski et al., 2000), and therefore can be spread on

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agricultural lands (arable and grassland) as it is rich in both the macro- and micro-nutrients required for healthy plant and animal growth (Ryan and Walsh, 2016). It also has potential to be used as an additive in compost, animal feed, biofuel, or it may be dried and incinerated (Korsström and Lampi, 2001; Ryan and Walsh, 2016). However, very few studies focus on DPS specifically. The fertiliser value and the possible environmental risk of DPS have not been studied in any great detail, and such knowledge gaps prevent such products from being recognised as sustainable marketable products. For example, the fertiliser value of DPS, which is an important parameter for farmers and agricultural advisors to know before land application, is rarely reported. It is significantly affected by the type of dairy plants, e.g. cheese factories generally have 50% more P than fresh milk dairies (Kwapinska et al., 2019). Therefore, more studies and tests should be conducted across the DPS from different factory to avoid improper landspreading. Moreover, although the heavy metal concentration of DPS has been reported to be low (Kwapinska et al., 2018; Pankakoski et al., 2000), some emerging organic pollutants may be present in DPS due to their lipophilic properties. The contamination of the soil with these emerging compounds, as a result of the DPS application, could be transferred to the plants via the roots into different plant tissues (Navarro et al., 2017). This would discourage many food companies from using crops or products (e.g. grazing of animals) originating from land amended with DPS (Perkins, 5 2019). There are also other concerns related to the use of DPS for land spreading. DPS decomposes quickly and releases strong odours due to high fat, oil and grease (FOG) and total suspended solids (TSS) content (Atallah et al., 2020; Bharati and Shinkar, 2013). Therefore, it cannot be stored for long periods and as the transport costs are high, it is commonly spread on lands in the vicinity of the dairy factories. Since the land bank of the nearby lands that can receive DPS is limited, it is easy to cause local oversupply of DPS, potentially leading to the accumulation of nutrients in soil, which may ultimately damage the aquatic ecosystem (Healy et al., 2016; Peyton et al., 2016). Weather conditions also constrain land spreading. For example, the land application of DPS is prohibited during the closed period over winter (i.e. hydrologically active period) in Ireland (S.I. No 378/2006). For these reasons, DPS cannot be fully utilized for land spreading. In the long term, there is a need to find alternative treatment and disposal methods of DPS. Secondary-raw-material-based fertilising products, which are referred to as STRUBIAS (STRUvite, BIochar, or incineration AShes), have already been recognised as fertilisers by EU to address this issue (EC, 2019; Huygens et al., 2018). STRUBIAS materials derived from wastewater and sludge are expected to be on the EU fertiliser market by 2030 and to be safe and effective alternatives for mined rock phosphate and processed P fertilisers (Huygens et al., 2018).

Knowledge gaps pertaining to present and future re-use of DPS and STRUBIAS products in agriculture still remain. Before these products can be deemed sustainable and safely used in agriculture, these aspects need to be reviewed and recommendations presented. Therefore, this paper aims to review present and future re-use pathways and potential challenges for these products in agriculture. Identification of such knowledge gaps will give the dairy processing and agricultural industries guidance on future research that is needed and may add value to the supply chain of the dairy production process.

2. Methodology

The review was carried out using scientific literature from databases and search engines including Google Scholar, American Chemical Society (ACS), Science Direct, Scopus, Springer Nature, Wiley and Web of Science. A detailed search of DPS and co-products reuse in agriculture in relevant literature was completed using the following keywords: dairy waste, dairy processing sludge, dairy wastewater treatment, STRUBIAS, struvite, sludge ash, biochar, fertiliser, fertiliser replacement/equivalent value, phosphorus, recovery, recycling, reuse, and emerging contaminants. Various combinations and derivations of the keywords were used.

As a result of these search criteria, 136 scientific papers were selected, from which about 45% were published in the last 5 years and 70% in the last 10 years. A deeper analysis was conducted on these papers and relevant information was extracted such as: dairy wastewater treatment methods, properties of DPS and current practices, fertiliser efficiency of DPS, potential environmental risk of DPS application, potential co-products derived from DPS and potential use in agriculture.

3. Dairy processing sludge characterisation

3.1. Current knowledge of dairy effluent nutrient and metal content

The dairy industry produces various products such as sterilised and pasteurised milk, yogurt, ice cream, butter, cheese, and milk powder, with different processes taking place such as pasteurization, coagulation, filtration, centrifugation and chilling (Carvalho et al., 2013). Dairy effluents vary significantly both in quantity and quality based on dairy factory characteristics (Janczukowicz et al., 2008) (Tables 1 and 2). The flow rates of dairy effluents vary due to scale, products, techniques, processes and equipment (Gutiérrez et al., 1991), and may also vary diurnally (Danalewich et al., 1998). Milk processing rates are typically higher in summer and lower in winter, and result in high seasonal variations in wastewater volume and properties (Janczukowicz et al., 2008). Moreover, the composition of these effluents varies greatly depending on the different types of products, system and operation methods (Carvalho et al., 2013). The effluent generally comprises dilutions of milk (or milk constituents including lactose, minerals, fat, whey and protein) lost in the technological cycles, starter cultures used in manufacturing, by-products (whey, milk and whey permeates), residues and contaminants from washing milk containers, equipment and floors, disinfectant applied in clean-in-place (CIP) processes, and other additives that may be used (Ahmad et al., 2019; Kolev Slavov, 2017). Dairy processing effluent is distinguished by high concentrations of organics and nutrients, and a pH varying from 4 to 12. Such a large variation of the pH is attributed to the use of acid and alkaline detergents and sanitizers for washing (Britz et al., 2006). The residues of milk and milk by-products in the waste stream result in higher nutrient and organic contents than those normally present in domestic wastewater (Booker et al., 1999). Suspended solids are derived from coagulated milk, cheese curd, or flavouring ingredients (Demirel et al., 2005). High concentrations of sodium (Na), potassium (K), calcium (Ca) and magnesium (Mg) have been measured in the wastewater, while heavy metals may be also present in low concentrations (Table 3).

3.2. Current knowledge of DPS nutrient and metal content

Dairy wastewater must be treated to meet licensed discharge limits before discharge to surface water bodies. Normally, there are three main stages of wastewater treatment (Fig. 1). Primary treatment consists of sedimentation/physical screening to remove large particles or debris, flow and composition balancing to stabilize effluent, chemical addition to control pH, and dissolved air floatation (DAF) to remove FOG (Ryan and Walsh, 2016). Two types of biological degradation systems, aerobic and anaerobic systems, can be used in secondary treatment to remove organic materials. Large quantities of DPS are produced during this stage and pollutants can be absorbed into it. Aerobic biological techniques, including activated sludge process, sequencing batch reactors, bio-towers or membrane bioreactors, are carried out using dissolved oxygen (Ryan and Walsh, 2016). This is a reliable and cost-effective treatment in producing a high-quality effluent, but results in high DPS generation (0.6 kg dry DPS per kg of biochemical oxygen demand (BOD_5) removed) and costly disposal problems (Britz et al., 2006). Frequently used anaerobic biological technologies involve anaerobic lagoons, up-flow anaerobic sludge blankets, membrane anaerobic reactor systems, and completely stirred tank reactors (Britz et al., 2006). Less DPS is generated during anaerobic digestion than during aerobic

Table 1
DPS generation (per unit volume/mass of processed milk) and disposal pathways in different countries.

Region	Water consumption	Effluents loads	DPS volume	Method of Disposal	Reference
EU	0.2–11 L/L processed milk	0.3×10^6–3×10^6 L (in a factory with capacity:10^6L milk/day)	1-3t dry matter sludge (in a factory with capacity: 10^6L milk/day)	Wastewater: drained to rivers sludge: land spread	Daufin et al. (2001)
EU	0.8–60 m^3/t processed milk	0.9–60 $m3^3$/t processed milk	0.2–30 kg sludge/t processed milk	–	EC (2006)
Sweden	0.96–4.0 L/L processed milk	0.86–4.3 L/L processed milk	–	Landfill, compost, irrigation, biogas production. In Denmark, 2/3 sludge from dairies is irrigated on cultivated land and the rest is utilized in biogas production.	Korsström and Lampi (2001)
Denmark	0.60–1.9 L/L processed milk	0.75–1.5 L/L processed milk	–		
Finland	1.2–4.6 L/L processed milk	1.2–3.9 L/L processed milk	–		
Norway	2.5–6.3 L/L processed milk	2.0–3.3 L/L processed milk	–		
Ireland	2.3 m^3/m^3 processed milk	2.71 ± 0.9 L/L processed milk	15–19.7 kg sludge/m^3 milk processed	Sludge: land spread (63%), compost (13.6%), or removed by licensed contractors (23.4%)	Ashekuzzaman et al., (2019a); Ryan and Walsh (2016)
Australia	0.07–2.90 L/L milk	–	31 kg organic waste/t product	Compost, fertiliser, stockfeed and recovery of marketable products.	Prasad et al. (2004)
United States	–	0.10–12.4 kg/kg milk	–213	Effluent: discharge into municipal sewage treatment system or irrigate on the land	Durham and Hourigan (2007)
United States	–	170–2081 m^3/d	–		Danalewich et al. (1998)
UK	1.8 L/kg product	1–5 L/L processed milk	–	Sludge: landfilling	Klemes et al. (2008)

processes (Britz et al., 2006). Phosphorus is removed in tertiary treatment through the use of chemicals like aluminium (Al) and/or iron (Fe) salts, before final discharge (Britz et al., 2006; Ryan and Walsh, 2016). Recently, the enhanced biological phosphorus removal (EBPR) process, without the need for chemical precipitants, has received increased attention. EBPR is achieved through the activated sludge process by recirculating sludge through anaerobic and aerobic conditions (Oehmen et al., 2007).

The wastewater treatment processes within a dairy processing plant generates a specific DPS type, which can be predominantly categorised into (1) lime-treated DAF sludge and (2) bio-chemically-treated activated sludge (Ashekuzzaman et al., 2019a). The former is produced after chemical and DAF treatment of raw wastewater during primary treatment. The latter is stabilized sludge from secondary biological degradation treatment, which can be either aerobic or anaerobic, or a combination of the two.

As DPS is categorised as a biosolid, it is commonly landspread in agricultural areas (Ryan and Walsh, 2016). DPS is a relatively new waste type and it is a much cleaner and valuable fertilising product than biosolids derived from sewage sludge, but it is rarely studied specifically. So far, very few studies have investigated the properties and fertilising effect of DPS. López-Mosquera et al. (2000) used DPS as a fertiliser for grassland and found that the heavy metal content didn't lead to harmful accumulation of metals in the short- or medium-term (4 years) (Table 4). Ashekuzzaman et al. (2019a) collected and characterised 63 DPS samples covering 9 major dairy processing companies of Ireland and found that the nutrient content varied across different sludge types (Table 4). The reported values of heavy metals in DPS (Table 4) were found to be lower than the EU upper limit thresholds recommended for bio-based fertiliser (EC, 2019), which indicates their relatively low metal bioaccumulation risk if used in agriculture.

4. Fertiliser equivalent value (FEV) of DPS

The efficiency of most bio-based fertiliers is normally unstable and lower than chemical fertilisers because of their relatively low nutrient content, slow nutrient release rate and highly variable nutrient composition (Chen, 2006). Therefore, the agronomic value of DPS should be determined before they are used in agriculture, which will make farmers more confident to use them. The FEV is defined as the application rate of mineral fertiliser to which the fertilisation effect of bio-based fertilisers on crop yield or nutrient uptake is equivalent (Brod et al., 2012).

The FEV can both provide a quantitative estimate of the amount of efficient nutrients in bio-based fertiliser and a theoretical estimate of its actual price in comparison to a mineral fertiliser. This can give farmers information about how to use bio-based fertilisers and the economic impacts associated with their use (Ashekuzzaman et al., 2019a). However, the results of FEV may vary widely, as FEV is not only affected by the assessment method, but also by factors like type of bio-based fertilisers, crop type, fertiliser application time (Delin, 2011), rates (Hijbeek et al., 2018), and method (Lalor et al., 2011).

To date, studies of FEV have mainly focused on the fertiliser equivalent value of nitrogen (N) (FEV-N) of manure and slurry. Research on the FEV (both FEV-N and fertiliser replacement value of P (FEV-P)) of DPS is scarce. Ashekuzzaman et al. (2021) applied an agronomic trial in grassland with four representative DPS to determine both the FEV-N and FEV-P of DPS. The FEV-N of DPS samples was observed to be between 8 and 54%, but the FEV-P was not derived as the experimental site was non-responsive to increasing mineral P rate. Ashekuzzaman et al. (2019a) provided a theoretical estimation of the FEV for the four types of Irish DPS from the total nutrient concentration (N, P, K), which showed a wide variation due to the considerable variation of DPS properties. In addition, the crop available fraction of N and P is still not well understood, which would play a vital role on the fertiliser value of DPS. The wastewater treatment process may have a significant effect on the plant available N and P. The ammonium-N (NH_4–N) concentration, which is easily plant available N, would decrease significantly with the use of lime (Libhaber and Orozco-Jaramillo, 2012), but may increase after an anaerobic digestion process (Mtshali et al., 2014). This effect on the plant availability of P is more complicated. Krogstad et al. (2005) found that the P fertilising effect of sludge with biological purification without chemical additives and lime treatment could be comparable to mineral P fertiliser, whereas P fertiliser value of sludges precipitated by use of Fe and Al salts without liming treatment was very low. Kahiluoto et al. (2015) found P was more available in sludge with a moderate Fe/P ratio (1.6), but had an adverse effect on the plant-availability of soil P with a surplus Fe coagulant (Fe/P of 9.8). Some studies have indicated that liming increases the plant-available P in sludge produced from the wastewater treated by Al and/or Fe salts (Bøen and Haraldsen, 2013; Krogstad et al., 2005; Montgomery et al., 2005). However, Kahiluoto et al. (2015) found that P was not available to plants in the sludge

Table 2
Characteristics of dairy waste effluent.

Effluent type	pH	BOD_5 (g/L)	COD (g/L)	TS (g/L)	TSS (g/L)	VS (g/L)	VSS (g/L)	FOG (g/L)	TN (mg/L)	TP (mg/L)	DOM (mg/L)	Reference
Milk factory	5.5–6.9	0.092–0.116	0.160–0.208	0.094–0.110							76.4–86.4	Mishra et al. (2000)
Dairy plants (produce cheese)	6.2–11.3	0.565–5.72	0.785–7.62	1.84–14.21	0.326–3.56	0.562–11.03	0.225–1.94		14.0–40.0	29–181		Danalewich et al. (1998)
Mixed dairy	4–11	0.24–5.9	0.5–10.4	0.71–7	0.06–5.8			0.02–1.92	10–660	0–600		Kolev Slavov, 2017
Milk reception	7.18	0.798	2.54		0.654			1.06				Janczukowicz et al. (2008)
Butter	12.08	2.42	8.93		5.07			2.88				Janczukowicz et al. (2008)
Cheese	7.90	3.46	11.75		0.940			0.331				Janczukowicz et al. (2008)
Cottage cheese	7.83	2.60	17.65		3.38			0.950				Janczukowicz et al. (2008)
Cheese whey	4.46	40	60	59	1.5							Gannoun et al. (2008)
Cheese whey	4.0–4.6	10–12.5	8.8–25.6	7.0–8.3	1.6–4.8			1.83–3.76	310–356	6.6–7.2		Rivas et al. (2010)
Hard cheese whey	5.80	29.48	73.45		7.15			0.994				Janczukowicz et al. (2008)
Cottage cheese whey	5.35	26.77	58.55		8.13			0.492				Janczukowicz et al. (2008)
Ice cream	5.2	2.45	5.2	3.9		2.6			60	14		Karadag et al. (2015)
Creamery	8–11	1.2–4	2–6		0.35–1		0.33–0.94		50–60			Demirel et al. (2005)
Cleaning water	10.37	3.47	14.64		3.82			3.11				Janczukowicz et al. (2008)

BOD_5 = biological oxygen demand for 5 days, COD = chemical oxygen demand, TS = total solids, TSS = total suspended solids, VS = volatile solids, VSS = volatile suspended solids, FOG = fat, oil and grease, TN = total nitrogen, TP = total phosphorus, DOM = dissolved organic matter.

Table 3
Concentrations (mg/L) of trace elements in dairy waste effluent.

Effluent type	Cd	Fe	Cu	Pb	Zn	Ni	Na	K	Ca	Mg	Al	Co	Mn	Reference
Dairy plants (mainly produce yogurt)	0.090	1.181	0.350	1.095	0.234	0.166								Afolabi et al., 2016
Creamery		2–5				0.5–1.0	170–200	35–40	35–40	5–8		0.05–0.15	0.02–0.10	Demirel et al. (2005)
Cheese		0.039–4.33	0–0.03			0.012–0.071	263–1265	8.6–155.5	1.4–58.5	6.5–46.3	0.063–0.257	1–0.007	1–0.835	Danalewich et al. (1998)
Mixed dairy		0.5–6.7				0–0.13	123–2324	8–160	11–120	2–97		0	0.03–0.43	Demirel et al. (2005)

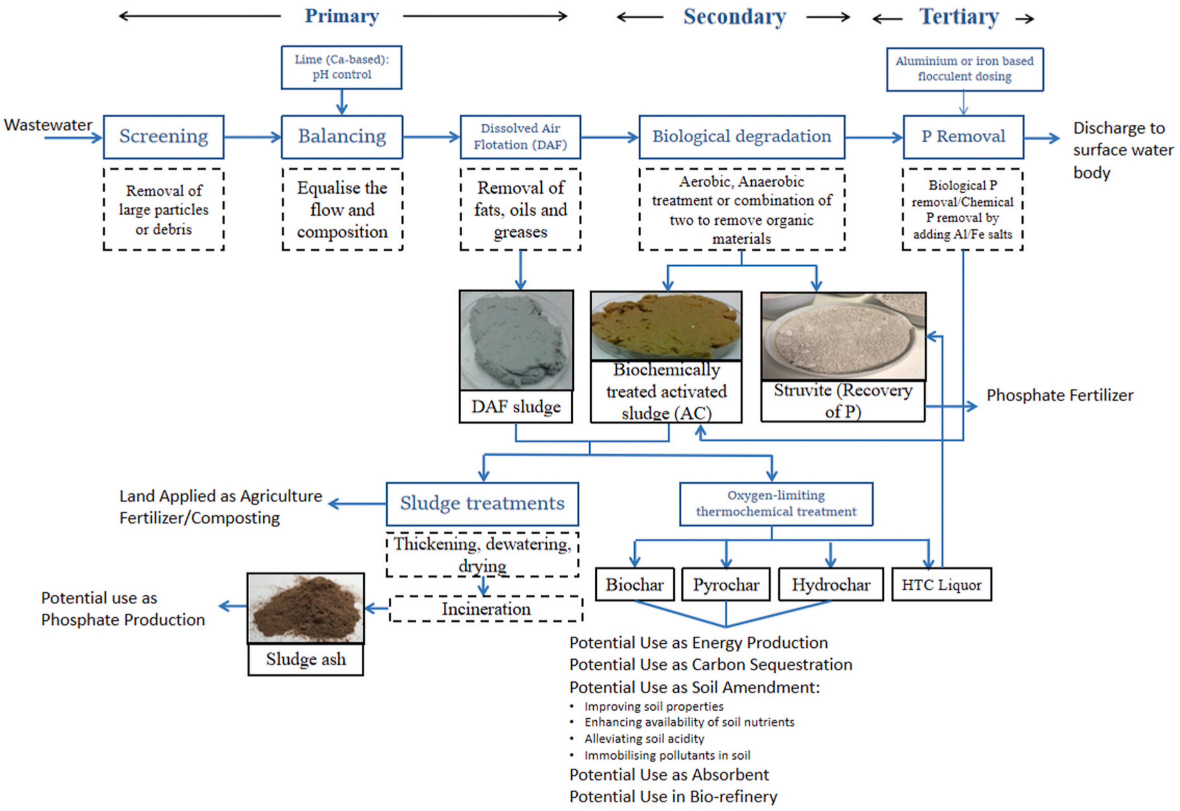

Fig. 1. Flow chart of dairy wastewater treatment process and sludge, struvite, and char generation. DAF sludge = Lime treated dissolved air flotation processing sludge; AC sludge = Bio-chemically treated activated sludge (adapted from Ashekuzzaman et al., 2019a).

Table 4
Characteristics of DPS. Adapted from Ashekuzzaman et al. (2019a) and López-Mosquera et al. (2000).

Parameters	Bio-chemically treated activated sludge "AC"*	Lime treated DAF sludge "DAF"*	Combined treated sludge "CM"[a]	Anaerobically digested sludge "AD"	Dairy-plant sludge	EU requirements of bio-based fertiliser[b]
DM (% of wt.)	13.3	25.9	16.1	3.5 ± 1.1		
OM (% of DM)	62.9	46.9	73.9	72.5 ± 1.3		
pH	7.3	7.2	6.8	7.5 ± 0.1		
TN (g/kg)	57.2	19.5	46.0	70.4 ± 1.2		>10
TP (g/kg)	36.8	65.9	20.0	14.6 ± 1.2		>10
TC (g/kg)	29.4	24.3	42.2	35.6 ± 1.2		
K (g/kg)	7.2	3.9	2.9	6.1 ± 1.1		>10
Mg (g/kg)	3.2	4.3	1.4	1.9 ± 0.1		
S (g/kg)	4.8	2.1	7.6	5.3 ± 0.7		
Na (g/kg)	5.3	3.5	3.6	19.9 ± 3.0		
Ca (g/kg)	44.8	152.9	21.0	59.7 ± 12.0		
Cr (mg/kg)	9.8	5.4	8.8	13.4 ± 3.5	15.99 ± 0.04	
Cu (mg/kg)	12.6	5.3	17.3	38.2 ± 6.7	58.55 ± 0.08	<300
Ni (mg/kg)	4.6	4.0	7.9	9.3 ± 2.4	11.04 ± 0.04	<50
Pb (mg/kg)	<2.0	<2.0	<2.0	6.3 ± 2.9	10.05 ± 0.12	<120
Zn (mg/kg)	75.2	54.7	109.8	217 ± 46	289.74 ± 0.67	<7800
Al (g/kg)	27.7	0.6	37.2	1.5 ± 0.5		
Fe (g/kg)	1.5	1.1	1.8	0.7 ± 0.1		
Co (mg/kg)	0.8	0.3	0.7	0.9 ± 0.2		
Mo (mg/kg)	2.2	0.5	2.1	18.4 ± 3.6		
Mn (mg/kg)	55.1	28.2	80.7	28.2 ± 6.8		
Cd (mg/kg)					0.11 ± 0.001	<1.5
Hg (mg/kg)					0.08 ± 0.02	<1

DM = dry matter, OM = organic matter, TN = total nitrogen, TP = total phosphorus, TC = total carbon, n.a. = not available.

[a] Median values are presented.

[b] The requirements of EU solid bio-based fertiliser with more than one macronutrients (EC, 2019).

hygienized with a high Ca/P ratio. Therefore, more agronomic trials are needed on the fertilising effect of N and P of different DPS relative to mineral FEV to optimise DPS utilization.

5. Potential contaminants in DPS

A number of potentially harmful compounds may enter the milk processing chain through various routes and ultimately accumulate in DPS (Fig. 2). Lactating animals are exposed to various chemicals, directly or indirectly, via the agricultural and veterinary practices on a farm (Fischer et al., 2011a). The active ingredient may be absorbed by animals, subsequently excrete into the milk, and eventually enter the waste stream through residual milk in the factory. In addition, some common contaminants such as dioxins and heavy metals are likely to be found in milk and dairy products, as they may enter and form incidentally during the production process (Fischer et al., 2011a). At present, there is limited information available on emerging contaminants in dairy processes. In this section, we list potential contaminants and their sources and fate in DPS.

5.1. Antimicrobial drugs

Antibiotics, including the *β*-lactams (penicillins, cephalosporins), tetracyclines, macrolides, aminoglycosides, quinolones and polymyxins, are the most frequently and commonly used antimicrobial drugs in dairy cattle management (Fischer et al., 2011a). They are widely administered to treat, control and prevent spread of diseases of dairy cows such as mastitis, laminitis, respiratory diseases, and metritis, and to enhance animal growth and feed efficiency (IDF (International Dairy Federation), 1997). All the administered antibiotics could enter the milk and subsequently transfer to other dairy products to some extent, depending on their physicochemical properties and ability to intact with the fat and protein (Giraldo et al., 2017). Adetunji (2011) found streptomycin, penicillin and tetracycline residues in soft cheese and yoghurt. Rama et al. (2017) indicated that amoxicillin, penicillin G and cloxacillin were the most frequently detected residues in the raw milk collected from six different major regions of Kosovo. Sniegocki et al. (2015) observed that chloramphenicol can be easily transferred from raw milk to commercial butter, white cheese, sour cream and whey, as this antibiotic accumulates in dairy products with high fat content. The antibiotic residues in the dairy products may eventually enter the waste stream, but current wastewater treatment technologies are unable to remove traces of antibiotics from wastewater (Phoon et al., 2020). Once added to soil, antibiotics affect the structure and function of soil microbial communities and induce phytotoxic effects on plant growth (Jechalke et al., 2014). Current antibiotic wastewater technologies including advanced oxidation processes (AOP), advanced treatment (adsorption and membrane) and biological treatment, have advantages (AOP can destroy the chemical structure of pollutants) and disadvantages (the pollutants were degraded in after AOP, but the toxicity remained) (Phoon et al., 2020). Hybrid technologies, involving several combinations of several technologies, are capable of removing antibiotics (Phoon et al., 2020).

5.2. Hormones

Endogenous hormones occur naturally in food of animal origin because animals can excrete steroid hormones. The amount excreted depends on age, state of health, diet, or pregnancy (Silva et al., 2012). Hormones are also used to promote growth, increase food production, medical treatment and improve reproductive performance, but the use of anabolic hormones in animal production is prohibited in the EU (EC, 1996; EC, 2003; IDF , 1997). Seventy-five percent of milk is produced predominantly by pregnant cows, which means that milk represents an important source of steroid hormones (Goyon et al., 2016). The natural hormone content of milk is typically between 40 and 500 μg/kg for the steroids (IDF , 1997). During the processing in the dairy plants, the residual hormones will enter the effluent through residual milk. In a WWTP, some hormones are removed through sorption to TSS and degradation, followed by removal of the excess sludge (Silva et al., 2012), which means that hormones may accumulate in the DPS.

5.3. Pesticides

Pesticides, including insecticides, herbicides, rodenticides and fungicides, applied in agriculture, have been shown to transfer to dairy animal bodies through feed and fodder (Rather et al., 2017). In addition, to protect the animals against disease from mites, ticks and insects, some

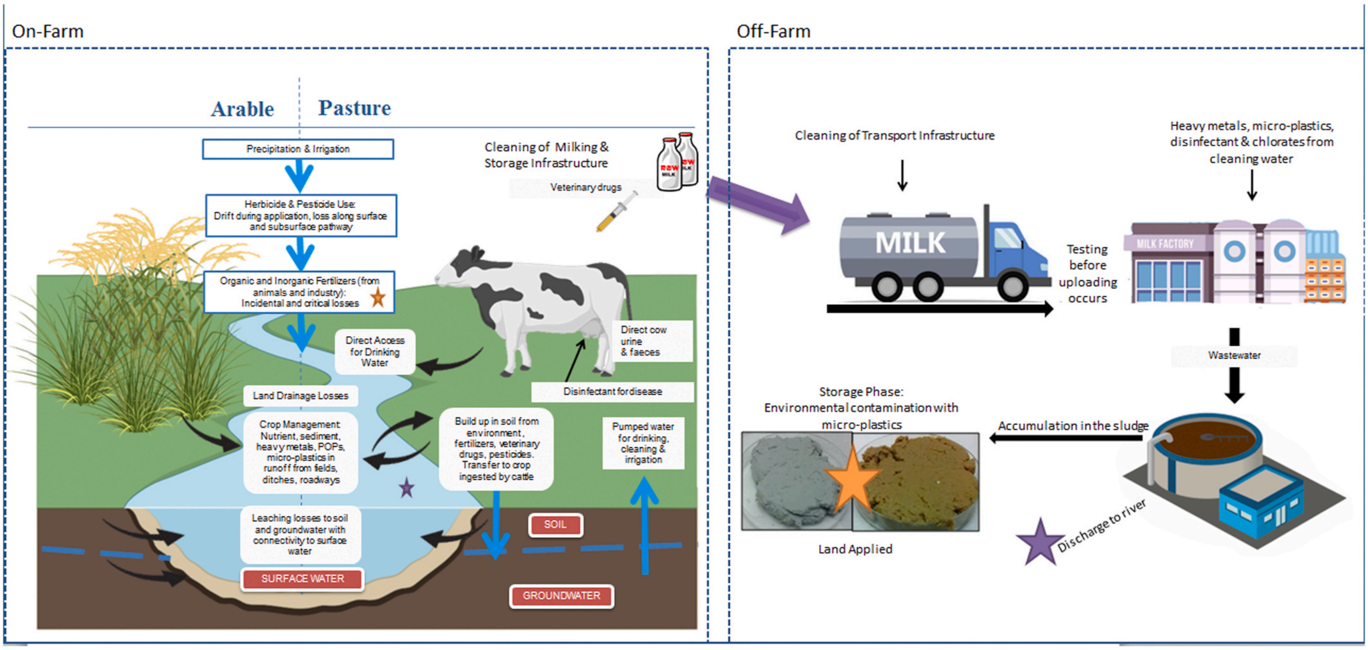

Fig. 2. The sources and fates of emerging contaminants in DPS.

pesticides are directly sprayed to the animals when they are housed. Animals will absorb pesticides orally, cutaneously, or via inhalation in such closed environments (Fischer et al., 2011a). Currently, common pesticides, including organophosphate, pyrethoids and carbamates, can be used on both routes and lead to the bioaccumulation in the dairy products (Akhtar and Ahad, 2017). The pesticides used in the cropping system and their metabolites will be lost to the environment via volatilization, aerial drift, runoff to surface water bodies, and leaching into groundwater basins (Wang et al., 2019), which can accumulate in the dairy animals or directly compromise drinking water used in the dairy factory. The residues of organochlorines and their metabolites also need to be considered. Although banned in many countries since the 1970s, residues still can be found in the environment due to their persistence and prolonged efficacy (Fischer et al., 2011a; Akhtar and Ahad, 2017). There is a vast list of pesticides used currently or in the past in agriculture with various levels of persistence in the soil, bedrock and water phases (McManus et al., 2017). This could have implications for grazing animals especially on heavy drained soils where, for example, 2-methyl-4-chlorophenoxyacetic acid (MCPA), which has a high solubility and low adsorption to soil matrix, is used to clear vegetation and has been found to have a much longer residence time in anaerobic waterlogged conditions (Morton et al., 2020).

From the US Food and Drug Administration data, dichlorodiphenyltrichloroethane (DDT) and its metabolites dichlorodiphenyldichloroethylene (DDE) and dieldrin, are the most commonly detected pesticides in foodstuff, including baked goods, fruit, vegetables, meat, poultry and dairy products (Schafer and Kegley, 2002). The OC pesticide, chlordane, has been found at a concentration of 1 ng/mL in raw milk samples (Fernandez-Alvarez et al., 2008). Golge et al. (2018) analysed 92 real dairy samples including raw milk, whole UHT (ultra-high-temperature) milk, Feta cheese and cream obtained from retail markets in Turkey, but none of the 167 pesticide residues were detected.

5.4. Disinfectants

Each procedure of the milk and dairy products process requires cleaning and disinfection to ensure removal of the bacteria and milk residues from all contact surfaces, including all processing equipment, transfer lines, tanks, trays, bins, blenders and conveyors (Cardador and Gallego, 2015). The most commonly used disinfectants are iodine-liberating agents, chlorine-containing substances, quaternary ammonium compounds, and hydrogen peroxide (Fischer et al., 2011a). A large amount of cleaning and disinfection agents enter dairy wastewater during the rinse-and-wash cycle of CIP system. Furthermore, using inadequately treated water to rinse and wash can be another source of contamination (McCarthy et al., 2018). Disinfectants are directly applied in the dairy wastewater to kill pathogens (e.g. faecal coliform and total coliform) during wastewater treatment (Akhlaghi et al., 2018). The residual of disinfectants could be either in their original state or as disinfection by-products (DBPs). Iodine sanitizers, usually as iodophors, are widely used in teat and skin disinfectants, filling/packaging machines, culture processing equipment, drop hoses, and hand dipping stations (Hladik et al., 2016). Iodinated DBPs are considered to be one of the most toxic DBPs, but have been tested less frequently than chlorine DBPs (Postigo and Zonja, 2019). Hladik et al. (2016) found trihalomethanes (THMs), including iodinated THMs, in the dairy wastewater and surface waters that receive dairy effluents (either directly from the dairy or through a WWTP).

Sanitation of water and equipment with chlorine-containing substances such as chlorine gas (Cl_2), dioxide (ClO_2), chlorhexidine and hypochlorite (ClO^-), remains common practice due to chlorine's bactericidal and oxidative properties (McCarthy et al., 2018). Chlorine reacts with any natural organic matter present in milk to form chlorine DBPs (Cardador and Gallego, 2015). Cardador and Gallego (2015) tested 84 milk and dairy products samples and found that 17 of them contained haloacetic acids (HAAs), the major class of non-volatile DBPs. The HAAs found in commercial samples can be attributed to contamination within the industrial processes like the washing of packages and equipment.

5.5. Persistent organic pollutants (POPs)

There are thousands of persistent organic pollutants (POPs) widespread in the environment. POPs tend to accumulate in the food chain because of their lipophilicity and low biodegradability (Jones and De Voogt, 1999). Since POPs occur ubiquitously, dairy animals are at danger from various sources of POPs, and these contaminants may transfer to the milk. In addition, some POPs such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), dioxins and furans, are common by-products or formed incidentally in industrial processes, and may subsequently enter the wastewater and sludge (Fischer et al., 2011b). PAHs are generally formed through a series of combustion processes occurring in industrial units. Boruszko (2017) detected 16 PAHs contents in three types of DPS and found 689 μg/kg dry matter (DM) in excess sludge, 95 μg/kg in post-flotation sludge, and 497.7 μg/kg DM in a mixture of excess and flotation sludge, which are considerably lower than the maximum permissible content of PAHs in biosolids (6 mg/kg DM) defined by EC (EC, 2000). A survey on 239 raw milk samples in France found that the average polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and PCBs concentrations were 0.33 pg toxic equivalent (TEQ)/g fat and 0.57 pg TEQ/g fat, respectively (Durand et al., 2008). Mamontova et al. (2007) found PCBs residues in milk and obtained a good correlation between PCB concentrations in milk and soil. Furans can be formed from the dehydration of sugars and would be expected to be found in dairy products that have been heated. Heaven et al. (2014) found three analogues of furan in the milk sample.

5.6. Microplastics

Plastic particles with diameters ranging from 0.1 μm to 5 mm are defined as "microplastics" and are a widespread anthropogenic pollutant in the environment with the extensive use of plastic (Phuong et al., 2016). Microplastics are mainly derived from synthetic fibres in clothing, industrial processes and personal care products, such as face cleaning soaps (Åström, 2016; Fendall and Sewell, 2009; Mahon et al., 2017). As an important food processing industry, the fate and sources of microplastics during the production process of dairy industry are largely unknown. The possible risks of milk contamination for microplastics may occur from cleaning equipment, the surrounding environment, as well as water supply conditions and inadequate handling of milk (Kutralam-Muniasamy et al., 2020). In addition, plastic-based packaging materials may lead to the microplastic contamination of milk. Kutralam-Muniasamy et al. (2020) collected 23 milk samples in Mexico and measured microplastics in the samples with an average of 6.5 ± 2.3 particles/L.

5.7. Nano particles

Nanotechnology, the designing and manufacturing of nano-scale (<100 nm) materials with specific chemical and physical properties (Kaegi et al., 2011), has been widely used in such applications as medicines, alternative energy, catalysts, and consumer products (Wang et al., 2017). Nanoparticles (NPs) primarily include silver, gold, copper, copper oxide, zinc oxide, titanium dioxide, manganese oxide, carbon nanotubes and magnetic matter (Wang et al., 2017). WWTPs are one of the most important pathways for NPs to enter the environment. The presence of NPs may have an effect on P removal and recovery (Chen et al., 2013).

6. STRUBIAS materials derived from DPS

Dairy factories produce large amount of DPS, which, on occasion, cannot be applied to land due to the limited nearby land bank for its application. This suitability may be driven by many factors such as soil type, crop type, weather conditions for trafficability, or farmer perception due to a lack on crop and sample specific FEV. Local oversupply of DPS leads to environmental issues including nutrient runoff, leaching, methane emissions, odour, and the accumulation of certain substances in soil through application over many years (Gascó et al., 2018; Kwapinska et al., 2018). Incidental runoff losses of nutrients and carbon from land application of DPS may also pose a risk to surface water quality deterioration. A recent study showed that edge of field-losses of NH_4–N and carbon from three types of DPS application was highest for Fe-rich DPS, whereas Ca–P-rich DPS showed highest dissolved reactive P losses but lowest losses of NH_4–N and carbon (Ashekuzzaman et al., 2020). Therefore, in the long term, there is a need to find alternative technologies to recover energy and nutrients from DPS. STRUBIAS manufacturing technologies has attracted attention and can potentially add value to DPS. The potential use, current problems and knowledge gaps of STRUBIAS products are investigated in this section (Table 5).

Table 5
The potential of DPS and its by-products application and current knowledge gaps.

Products	Potential Use	Current Problems	Current Scientific Knowledge Gaps
Bio-chemically treated activated sludge "AC"	As a grassland and arable organic fertiliser	Farmers need more fertiliser value to optimise application and maximise yield responses, odour and local oversupply	Full nutrient and emerging contaminant content characterisation, N–P–K fertiliser value for a variety of crops and soil types, gaseous emissions and long-term agronomic trials absent.
Lime treated DAF sludge "DAF"	As a grassland and arable organic fertiliser	Farmers need more fertiliser value to optimise application and maximise yield responses, odour and local oversupply. Decomposes quickly leading to fungus problem	Full nutrient and emerging contaminant content characterisation, N–P–K fertiliser value for a variety of crops and soil types, gaseous emissions and long term agronomic trials absent.
Sludge Ash	Phosphorus resource.	High heavy metal content	Need technology to remove heavy metals. Alternative uses.
Biochar	Energy production, carbon sequestration, organic soil amendment, absorbent for heavy metals	The impacts on soil and crops, the heavy metal and organic contaminants, the cost of production and transportation	The properties of chars and the mechanism of interaction between chars and soil, long term environmental risk
Pyrochar	Carbon sequestration, organic soil amendment, absorbent		Very few studies on pyrochar and hydrochar. More data are needed. What are suitable amendment rates and how often? Fertiliser value, the technology to remove heavy metals from feedstocks (ash and hydrochar) and optimise the P recovery
Hydrochar	Energy production, carbon sequestration, organic soil amendment, absorbent, bio-refinery.		
Struvite	Phosphate fertiliser	The technology of struvite precipitation	More research is needed on the purity of struvite.

6.1. Struvite

Struvite (magnesium ammonium phosphat hexahydrate, $MgNH_4PO_4.6H_2O$) is a P mineral that can be precipitated from aqueous waste streams by increasing the pH of wastewater and maintaining a stoichiometric PO_4^{3-} to Mg^{2+} molar ratio (Hertzberger et al., 2020). Struvite precipitate is normally formed in WWTPs during the anaerobic digestion process when significant levels of Mg occur in the wastewater (Booker et al., 1999). Occasionally, large amounts of struvite may form and deposit on the walls of the digesters and connecting pipes, which results in downtime, loss of hydraulic capacity and increased maintaining costs (Booker et al., 1999). However, struvite precipitation is an effective P recovery method. The pilot and operational facilities that manufacture struvite are commonly installed at municipal WWTPs, but are not frequently installed at food processing plants (Huygens et al., 2018). Struvite is an excellent fertiliser because it has similar fertiliser efficiency to common mineral P fertilisers such as single super phosphate and triple superphosphate (Johnston and Richards, 2003). Compared with traditional fertilisers, struvite has a high P_2O_5 content, and is an excellent slow release fertiliser that does not "burn" roots when over applied (Xu et al., 2012). The fertilising effect of the struvite precipitate on maize was investigated in a pot trial and the results obtained show that struvite can be an effective source of fertiliser (Uysal and Kuru, 2015). Struvite precipitation from different wastes like dairy, urine, swine manure, semiconductor wastes, sludge, and reject water from sludge thickening and dewatering process is also practised (Li et al., 2019; Ren et al., 2016). However, the chemical compositions of waste-recovered struvite are not always consistent with pure struvite (Hall et al., 2020). Furthermore, metal impurities such as Al, Fe, Ca and small amount of heavy metals can also precipitate along with the struvite (Li et al., 2019). Dairy waste including wastewater, DPS and other STRUBIAS co-products show a significant potential for P recovery in the form of struvite. Uysal and Kuru (2015) detected high N, P and Mg contents in struvite precipitate produced from dairy industry wastewater, while heavy metal concentrations were below detection limits. However, if the dairy wastewater is rich in Ca, the struvite crystallization rate and product quality might be affected and may require additional steps (e.g. calcium removal or step-by-step precipitation) as a pre-treatment process (Li et al., 2019). Chelating agents like ethylenediaminetetraacetic acid (EDTA) addition, sodium carbonate addition and CO_2 stripping are the feasible technologies to remove Ca in wastewater to enhance the purity of the obtained struvite (Hu et al., 2020; Zhang et al., 2010). Becker et al. (2019) reclaimed both N and P from hydrochar-derived sewage sludge and its process liquid via struvite precipitation. An acid leaching step removed phosphate from the hydrochar, while the process liquid arising from hydrothermal carbonization (HTC) was used as an NH_4 source for struvite precipitation. Xu et al. (2012) used an acid leaching method to extract P and produce struvite from sludge ash, which recovered more than 97% of P in sludge ashes.

To date, very few studies have investigated struvite precipitation from the dairy industry. The efficiency of P recovery and the precipitation technology needs to be further studied and optimised, as there are multiple factors that could potentially lead to inconsistency in the composition and speciation. In addition, research is needed to assess the toxicological compounds in the struvite because the contaminants in hydrochar and sludge ash might be simultaneously leached during P extraction.

6.2. Char-based materials

The term "char-based materials" is used here to replace 'biochar' in the STRUBIAS acronym, as they have different terms depending on the technology. Char-based materials obtained from the thermochemical conversion of biomass in an oxygen-depleted atmosphere are porous and carbonaceous, and are more stable and C-rich and less toxic than the feedstock (Atallah et al., 2020; Kambo and Dutta, 2015). Different thermochemical pre-treatment processes and conditions result in different final products. Pyrolysis is a prevailing thermal decomposition technology of OM (e.g. agricultural wastes, lignocellulosic biomss and sewage sludge) to convert biomass into valuable products like biochar, bio-oil and gas components at temperatures between 350 and 1000 °C in the absence of oxygen (Nanda et al., 2016; Ashekuzzaman et al., 2019b). Pyrolysed OM with a C content higher than 50% of DM are defined as biochar, otherwise, they are defined as pyrochar (EBC, 2012). HTC is, in contrast to pyrolysis, a wet conversion technique, degrading the OM content of sludge in the presence of water at a temperature range of 180–260 °C (Kambo and Dutta, 2015). Other than in pyrolysis, the HTC process does not require the drying of feedstock before and/or during the reaction (Malghani et al., 2013; Fakkaew et al., 2015). The HTC process produces a solid product, known as hydrochar, and a process liquid with high loads of small-chain organic acids, NH_4 and phosphate (Becker et al., 2019). It may therefore be more energetically efficient to convert wet biomass like DPS to hydrochar (Mau and Gross, 2018).

There are many functions of char-based materials including, but not limited to, energy production, agriculture, carbon sequestration, wastewater treatment and bio-refinery (Kambo and Dutta, 2015). The utility of a specific char-based material for any particular application depends on its inherent properties, which are mainly affected by their feedstock, pre-treatment method, and temperature (Amoah-Antwi et al., 2020). For energy production, hydrochar is a very suitable candidate as hydrochar shows considerable reduction in the ash content compared to that of raw feedstock and biochar produced via slow pyrolysis (Kambo and Dutta, 2015). In agriculture, the use of char-based material as a soil amendment is anticipated to improve chemical, physical and biological properties of soil and thereby crop productivity (Laird et al., 2010). Those rich in available nutrients and minerals and/or showing high water holding capacity could be better used as soil amendments to improve fertility (Graber et al., 2010). If char-based materials are used for C sequestration, it is necessary for them to have high environmental stability (Mašek et al., 2013). The stability of biochar in soil depends on several factors, especially the production method (Lehmann et al., 2009). Studies have rejected the potential of using hydrochar for carbon sequestration due to the low stability of hydrochar in soil (Berge et al., 2013; Eibisch et al., 2013). Biochar usually has a high specific surface area (SSA, $>400\ m^2/g$) and more condensed polyaramatic structures, and hence is a good adsorbent for various contaminants (Amoah-Antwi et al., 2020; Kambo and Dutta, 2015). Hydrochar usually has very low SSA and porosity compared to biochar; however, due to the presence of oxygen-rich functional groups on its surface, the adsorption capacity of hydrochar is also high (Liu et al., 2010). The HTC process is promising in the field of pyrolysis of biomass for bioenergy production. The intermediate products includes 2,5-HMF, aldehydes (acetic, lactic, propenoic, levulinic, and formic acids), and other phenolic compounds generated during HTC can potentially be used for the manufacture of chemicals in the bio-refinery industry (Kambo and Dutta, 2015). DPS could be potential candidate for thermochemical treatment due to its low heavy metal content. Sadeghi et al. (2018) spread biochar derived from air-dried DPS over the surface of small-scale boxes filled with an erosion-prone soil and found that the biochar increased C, N, OM and C/N of the soil. In addition, they detected that biochar production significantly decreases the heavy metal, N, P and K contents, and increased the C and C/N ratio. Their study showed the potential of DPS-derived biochar to be an eco-friendly soil amendment and carbonaceous adsorbent. Ashekuzzaman et al. (2019b) studied pyrochars originating from two DPS types, i.e. activated sludge and DAF sludge, and used them as a carbonaceous adsorbent for P removal from wastewater. They found that the type, composition and the mineral composition (i.e. availability of Ca, Mg and Si) of DPS-derived pyrochar samples were associated with P removal process. Atallah et al. (2020) carried out batch HTC experiments using DAF sludge to investigate the effects of changing temperature, residence time and water-sludge ratio on the yield and quality of the hydrochar. They found that the production of hydrochar improved the characteristics of DPS, and an increase in reaction temperature, residence time and water-sludge ratio increased the hydrochar yield along with their energy and carbon content, and decreased the oxygen and volatile matter content.

Despite the benefits of char-based materials, there are several knowledge gaps with respect to the application of char-based materials derived from DPS. First of all, thermochemical treatments increase the risk of producing chars with other highly toxic compounds produced from high-temperature reactions such as PAHs, PCBs, dioxins, furans, and PCDD/Fs (Amoah-Antwi et al., 2020; Kambo and Dutta, 2015). Heavy metals present in the feedstock are most likely to remain and concentrate in the chars (Shackley et al., 2010). Therefore, careful analysis of feedstock and final products is necessary to avoid contamination in the soil. Second, char-based materials are complex, multi-functional materials that require improved mechanistic knowledge and understanding of its production, properties, impacts and interactions. The knowledge of char-based materials, especially hydrochar, derived from DPS is still in its early stages of development and all the aspects mentioned require additional research. Their benefits should be maximized through the mechanistic process understanding. Third, the cost of collecting of feedstocks, transportation, production and storage need to be properly assessed and managed.

6.3. *Ashes*

Ashes are characterized as fly ash or bottom ash, or a combination formed through the incineration of bio-based materials by oxidation (Huygens et al., 2018). They can be obtained from incineration plants which produce ash-based materials specifically for further agricultural use, or can be a production residue resulting from incineration of wastes or other production process (e.g. energy). Ash normally contains valuable plant macronutrients such as K, P, S, Ca and Mg (Brod et al., 2012; Haraldsen et al., 2011; Insam and Knapp, 2011), especially the amounts of P (13.7%–25.7% P_2O_5), which can be comparable to commercial superphosphate (Xu et al., 2012). However, the potential utilization of ashes as fertiliser is limited, since it is also inevitably enriched in heavy metals (Franz, 2008; Herzel et al., 2016). Sludge ash could be a secondary feedstock in the production of marketable phosphate fertiliser. So far, there have been a number of studies on the technologies to extract and recover P from sewage sludge ash. Nakagawa and Ohta (2019) used alkaline leaching technology to recover P as calcium hydroxyapatite from sewage sludge ash. Acid solutions like H_2SO_4, HCl, HNO_3, and H_3PO_4 are usually used for ash leaching to extract P (Biswas et al., 2009; Tan and Lagerkvist, 2011). Franz (2008) recovered P as fertiliser by adding lime water to precipitate calcium phosphates and other calcium compounds. Herzel et al. (2016) used a new thermochemical process for sewage sludge ash treatment, which transformed the phosphate-bearing mineral phases into plant available phosphates.

7. Conclusion and future research

Based on the EU's Farm to Fork Strategy, sustainable agriculture and organic farming will be encouraged in the future. DPS is recognised as a new organic fertiliser and a potential feedstock of STRUBIAS products. STRUBIAS products have potential commercial applications as both fertilisers (e.g. struvite), fertiliser components and soil amendments (e.g. chars). An important outcome of this review is that testing and publication of nutrient and metal data pertaining to DPS and DPS-derived STRUBIAS characteristics is not common. This is exasperated by the lack of testing and publication of data for other constituents such as heavy metals, pathogens, antimicrobial drugs, hormones, pesticides, disinfectants, POPs, microplastics and nano particles. These constituents, introduced during processing or treatment of the products, may be present at the land application stage. This is of particular concern for bioaccumulation in the soil and crops, with associated incidental losses in surface or near surface runoff to the aquatic environment. In addition the nutrient content and availability to plants differs across sludge and STRUBIAS product types due to many factors such as the type of dairy processing plant, wastewater treatment process and production technologies. Equally, the fertilizer equivalency value for both P and N is not known for all products and is not factored into application rates. This means that at farm scale neither agronomic nor environmental needs are being optimised. Future work should focus on these outcomes in long term field trials.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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