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Evaluating land application of pulp and paper mill sludge: A review

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ABSTRACT

It is estimated that >400 Mt of board and paper are produced globally per year, and that 4.3–40 kg (dw) of sludge like material, pulp and paper mill sludge (PPMS), is generated for every tonne of product. PPMS are now more widely reused in agriculture as a soil amendment due to their high organic content of 40–50% by weight, perceived low toxicity and possible liming capabilities. Within this review article historic and recent literature on PPMS land spreading are combined with knowledge of European and UK regulation to explore the benefits, potential impacts and viability of land spreading PPMS. The review reveals that risks relating to potential N immobilisation in soils post-application can be readily mitigated, if desired, by coapplication of an N source, or even pre-treatment of sludge via composting. The benefits to crops have been demonstrated emphatically, while negative ecological impacts under typical field application rates have not been observed to date. The case is therefore strong for continued land application of the material as an environmentally responsible and sustainable use option. However, there are currently gaps in the literature regarding longer-term implications of PPMS applications in agriculture and in regards to the possible presence of emerging contaminants in some PPMS materials, both of which have been identified as areas that merit further research.

1. Introduction

During the production of paper and board, virgin or recovered timber and possibly a portion of recycled paper and board go through a series of processes at pulp mills to separate out the cellulose fibres and so produce a cellulose rich product known as pulp. Although mechanical pulping is still used in some areas, most large-scale pulping operations now use a chemical pulping process based on heat and pressure plus either an alkali treatment (the most prevalent is known as the Kraft process) or an acidic treatment (known as the sulphite process) (Demuner et al., 2021; Monte et al., 2009). This pulp can then be used at an onsite paper mill, dried and bailed for transportation to offsite paper mills, or used in other industries such as a binding agent in pharmaceuticals and food products. At the paper mill this pulp is mixed with water and refined to the end product's specification before fillers such as clays, talc and calcium, as well as colouring agents, are added. This material is then processed to the desired specification of the end product (Fig. 1). Globally, it is estimated that these processes produce 184.4 Mt of pulp and 402.790 Mt of paper and board annually (CEPI, 2018; Magnaghi, 2015). This leads to the production of approximately 400 million wet tonnes of pulp and paper mill sludges (PPMS) globally every year (Faubert et al., 2016).

The complete process of production of both pulp and paper leads to the creation of multiple waste- and by-products, with around 87% of these materials being classified as PPMS, whilst the other 13% is accounted for by impurities, waste chemicals and gaseous emissions (Norrie and Fierro, 2020). For example, during pulping, the Kraft process produces approximately 100 kg of waste per air dried tonne (ADt) of pulp, whilst other less common semichemical or physical methods produce around 60 kg ADt^{-1} (IPCC, 2001). The waste output can increase to 200–400 kg per tonne of product at mills where recycled paper is used as a feed stock (Balwaik and Raut, 2011; de Alda, 2008). This is important as 71.6% of paper is recycled in Confederation of European Paper Industries (CEPI) member states, and around 60% in the U.S.A, both of which are major producers of paper and board, while comparable figures are reported in other parts of the world (CEPI, 2018; Scott, 2019).

This review focuses on the end of life disposal or reuse of PPMS, particularly spreading PPMS on agricultural land and the impacts that it has on soil chemistry, physical properties, and terrestrial ecology based upon past studies and available grey literature. The regulations on PPMS reuse within the UK will be discussed as a case study of the effective

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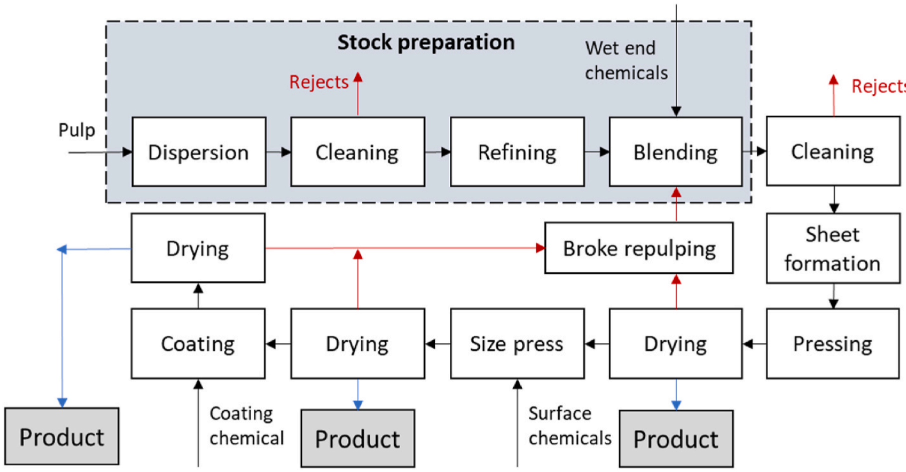

Fig. 1. The typical processes employed at a paper mill, red lines indicate rejects and blue lines indicate products, based upon Webb (2003). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

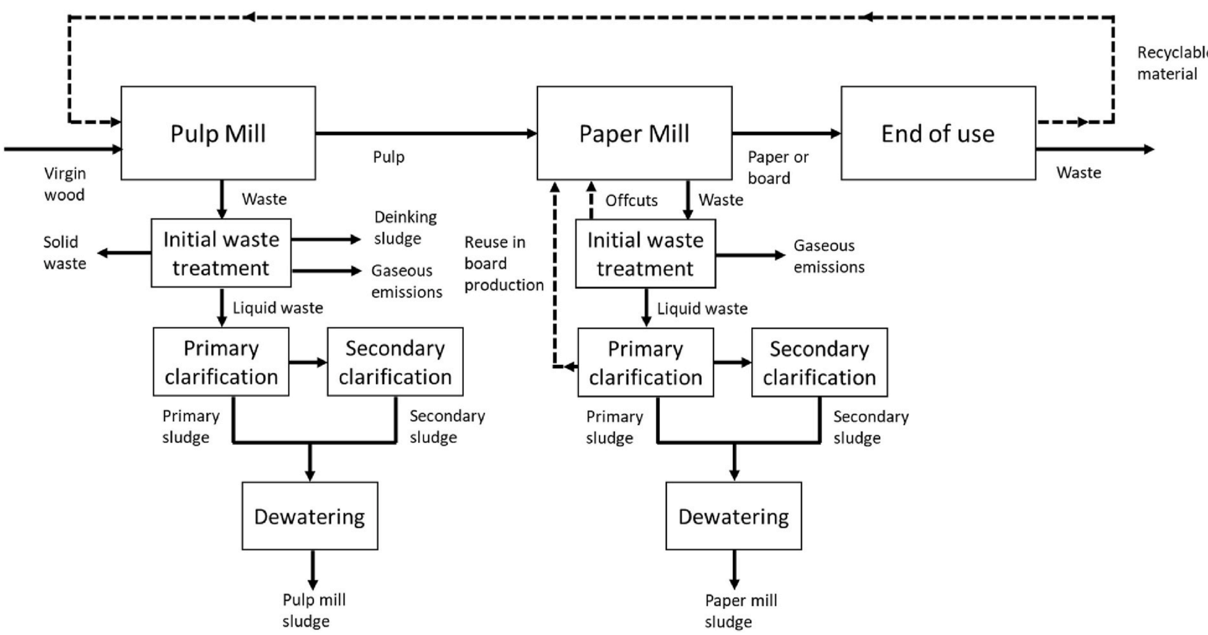

Fig. 2. Waste streams produced during the production of pulp and paper.

implementation of land spreading PPMS. The type of PPMS (i.e. pulp/paper or primary/secondary) will be emphasised as to distinguish the effects of each of these factors. Advantages and limitations to further adaption of this current recycling route will be highlighted.

2. Paper and pulp mill sludge production

Sludge is produced at both pulp and paper mills from the clarification of the liquid waste stream, although these wastes may be combined when both mills are integrated. As the main aim of pulping is to liberate the cellulose fibres, which are the foundation of paper products, from the lignin and other components found within wood, the pulp mill liquid waste stream is comprised of predominantly lignin and short chain cellulose fibres which are not suited for use in pulp production. Whereas, paper mill liquid waste is mainly composed of fines, added fillers and coatings, particularly kaolinite and calcium carbonate ($CaCO_3$), that are used in varying quantities depending on the end product of the paper mill.

The PPMS generated at pulp and paper mills can be categorised into Primary and Secondary sludges (Fig. 2). Primary sludge refers to the material generated by the initial clarification of raw paper/pulp mill effluent via flotation or sedimentation. A proportion of this material can be reincorporated at paper mills for the production of lower quality end products such as board but is typically less suitable for reincorporation into higher end products. In order to reduce the volume, chemical oxygen demand and biochemical oxygen demand, the primary sludge may undergo further treatment. This commonly involves biological decomposition through aerobic activated sludge systems, aeration and mixing to oxidise, or a successive combination of these or other methods to generate a more processed waste material known as secondary sludge. Secondary sludge is more difficult to dewater due to the high biologically active content and thus is often combined with primary sludge before dewatering. These combined sludges generally contains approximately 70% primary sludge and 30% secondary sludge in developed countries, but will vary from mill to mill (Bajpai, 2015). Furthermore, some mills do not employ biological treatment leading to no secondary sludge being produced (Bajpai, 2015). Primary and secondary sludges are sometimes combined and at operations where both pulp and paper

Table 1
The reported physicochemical properties of primary, secondary and mixed PPMS (Faubert et al. (2016) and references therein; Simão et al. (2018); Faubert et al. (2016); Negi and Suthar (2013); Ganguly and Chakraborty (2018); (Coimbra et al., 2015); (Ribeiro et al., 2010)).

Parameter	Paper mill		Pulp mill		Mixed paper and pulp mill		Deinking sludge
	Primary	Secondary	Primary	Secondary	Primary	Secondary	
Dry matter (% w/w)			22.9–33.0	47.3	15–57	1–47	19–60
Ash content (% solids)			33.10	24.39	10–15	10–20	20
Nitrogen (ppm)	2390–5400	4680	38	2560	450–2800	11000-77000	7000–36000
Phosphorous (ppm)	31400		167	370	100–600	2500–28000	2200–7400
Potassium (ppm)	3170				200–900	780–7000	300–3300
C:N ratio	138.92–289.47	86.21			111:1–943:1	8:1–50:1	13:1–31:1
pH	7.58		6.6–8.0	6.8–8.2	5.0–11.0	6.0–8.5	3.8–8.5
Organic matter (% w/w)	75.1	40.2	36–47.8	11–76.1			

mills are integrated at the same site sludge from both mills is sometimes combined (Hooda et al., 2018). These combined sludges are generally referred to as mixed PPMS. Any of these sludges can be commonly referred to as PPMS.

2.1. The influence of recycled paper incorporation

Repulping of recycled paper at pulp and paper mills can vastly change the characteristics and quantity of sludge produced. As mentioned in section 1, the quantity of waste sludge produced at paper and pulp mills using virgin materials only is relatively low, but this quantity increases (typically 2–4 fold; (Balwaik and Raut, 2011; de Alda, 2008)) where recycled paper is used in the production process. This increase in sludge production is due to the increased number of impurities. For example, when using recycled paper in pulping, components such as ink residue, coatings and fillers (some high grades of paper contain up to 40% filler (w/w) (de Alda, 2008)) must be removed and so these components end up in the sludge residue and therefore the sludge produced contains a lower proportion of organic content (CPI, 2015). The reuse of paper waste in pulp production is common, not only because it is environmentally beneficial but also because the cellulose fibres are already separated from the lignin present in wood and so reducing the need for that stage of processing. However, the recycling of paper cannot be repeated indefinitely as cellulose fibres are broken down and shortened with use, therefore eventually becoming unsuitable for paper making (García et al., 2008).

A separate, third kind of sludge, deinking sludge, can also be generated during the paper making process when using recycled paper as a feedstock. However, deinking sludges have different constituents and properties to the primary and secondary sludges (i.e. lower nutrient and organic matter content), which makes their potential land applications different to those of the main PPMS, and therefore they will not be discussed in this review. For a review of deinking sludge, see Camberato et al. (2006), while for a review of the wider suite of wastes from paper production, see that by (Simão et al., 2018). Additionally, for an example of governmental guidance on spreading of PPMS (UK example) see Gibbs et al. (2005).

2.2. Production figures

While pulp and paper mill companies do often report their waste production figures in annual reports, drawing comparisons is complicated because there is no common system for measurement or quantification of these wastes. That is, wide variations are observed such as to whether wet or dry masses are determined, the water content of wet materials, which other waste streams are included in the measurements, and at what point during the treatment process quantities are measured.

3. Physicochemical properties of PPMS

The physicochemical properties of PPMS produced at different mills can vary (Table 1) depending on the raw materials used, the treatment processes employed at the mill, and the nature of the end product (i.e. grade of product produced influences the level of treatment and nature of additives). Previous studies have also highlighted that variations in sludge properties can arise even when comparing sludges from different mills that employ similar processes and/or produce similar products (Scott and Smith, 1995). Therefore, individual characterisation of sludges is crucial if informed decisions are to be made about their suitability for land application and are currently already conducted in many countries. Bulk density and water content are important characteristics of PPMS that are dependent upon the pulping mechanisms utilised at the mill, the waste treatment processes employed, and the level of dewatering (which is achieved through various means, most commonly via vacuum filtration, centrifugation or mechanical press using a screw or belt press system: (Amberg, 1984; Meyer et al., 2018)). Dewatering is particularly influential, with PPMS in its initial state often having only 0.5–2% solids content (Bajpai, 2015), but the end result of dewatering can be variable. For example, a study by de Alda (2008), which analysed 20 sludges from various paper and pulp mills, found the final water content to be 65 ± 17% (w/w), while Meyer et al. (2018) and Bajpai (2015) reported values across the range of 60–75% water content, however the European Commission reports that up to 50% solids can be achieved by employing a screw press (Suhr et al., 2015).

Previous studies have reported the bulk density of PPMS to vary between 0.419 and 0.598 g cm^{-3} (Jackson and Line, 1997c, 1998; Jain et al., 2018; Rios et al., 2012), while the cation exchange capacity (CEC) of PPMS can also vary widely (e.g. 5.3 cmol (+) kg^{-1} to 297 cmol (+) kg^{-1}) depending on clay and organic matter content (Camberato et al., 2006). The work by de Alda (2008) also reported a typical pH near neutral at 7.6 ± 1.3, which was similar to the values later reported by Veluchamy and Kalamdhad (2017) who found the pH of PPMS samples to be 7.39 ± 0.004. These values, in turn, are within the range of pH values (6.6–8.2) noted by Simão et al. (2018) for primary and secondary PPMS, which indicates that PPMS are typically in the pH neutral to mildly alkaline range. This neutral to alkaline pH is derived from their high $CaCO_3$ content that originates from paper coating materials or from causticizing which occurs during pulping when sodium carbonate reacts with calcium hydroxide to form $CaCO_3$ (Camberato et al., 2006; Norris and Titshall, 2011; Nunes et al., 2008; Vasconcelos and Cabral, 1993). This in turn imparts PPMS with a liming capacity, with the $CaCO_3$ equivalence of PPMS having been reported to be between 12.7% and 50% (Camberato et al., 2006). In keeping with this, they are also known to have a high buffering capacity (5 mol H^+ kg^{-1} sludge/pH) (Calace et al., 2003). In fact, PPMS are often applied as a liming agent when their $CaCO_3$ equivalence in above 30%, with organic and nutrient additions considered to be a secondary benefit.

Jackson and Line (1997b) determined the proportions of lignin, holocellulose, cellulose, and hemicellulose in primary pulp and paper mill sludge (via a modified Klason method) to be: isoluable lignin) 27.42 soluble lignin 2.87 holocellulose 72.65 cellulose 57.08 hemicellulose 11.23 (dry w/w %). Typically, paper mill sludges have high mineral contents, with primary sludge has a 40:60 ratio of organic material: mineral matter, while secondary sludge has a 50:50 ratio of the two

(Bajpai, 2015). Primary sludge has lower N and P content than secondary sludge (0.045–0.28% N and 0.01–0.06% P versus 1.1–7.7% N and 0.25–2.8% P respectively) due to the addition of nutrients used to stimulate microbial activity during secondary treatment and the microbial biomass itself (Faubert et al., 2016). And while the C:N ratios of PPMS may vary, the C:N ratios of primary sludges are much higher than those of natural soils, up to 943:1, while those of secondary sludges may be similar to soils (~70:1), if not lower, with those reported in the literature ranging from 8:1 to 50:1 (Faubert et al., 2016).

4. Historic disposal methods for paper and pulp mill wastes

Although the focus of this article is on sludges from pulp and paper production, historic disposal of liquid wastes does warrant a mention also. Discharging of liquid effluents into waterways without primary or secondary treatment was historically common practice (Dolar et al., 1972). However, as regulations and environmental policy have become more stringent this practice has all but been abandoned mainly due to the high chemical oxygen demand and biochemical oxygen demand of the effluents (as high as 1100 and 550 mg l^{-1} in chemical treatment mills and 1160 and 500 mg l^{-1} in mechanical treatment mills respectively) (IPCC, 2001; Möbius, 2006). Disposal alongside municipal waste (sewage) treatment was also commonplace for smaller mills as the material is compatible with the infrastructure available at wastewater treatment plants, although this was not feasible for larger scale operations due to the quantities of pulp wastes produced (Scott and Smith, 1995). Therefore, treatment of waste to generate the semi-solids material we now refer to as PPMS became common practice to reduce the overall quantities of waste and recover water. Presently in the UK, any remaining effluent or liquid waste produced is removed from site under a consent from the regulator. Either via a discharge consent, if the waste meets the specified parameters, treated on site prior to discharge or treated in an appropriately permitted independent facility.

For the semi-solid sludge materials produced currently, a popular method of disposal is incineration; this is more feasible in pulp and paper mills where other waste materials such as debarking material are already incinerated for energy production. The high organic content of PPMS makes them potentially combustible and so suitable for incineration, with primary and secondary PPMS having modest energy contents of 2690 and 4000–5000 MJ per wet tonne of material respectively (Bajpai, 2015). Initially, PPMS contain around 0.5–2% solids content but are generally dewatered (often by belt pressing) to 25–40% solid content on site (Bajpai, 2015). However, their high-water content typically necessitates further dewatering before incineration, decreasing the net energy yield. A further complication is that paper sludges have high ash contents which require more specialised equipment to incinerate and produce more ash which requires disposal, and therefore greater initial capital investment. Eikelboom et al. (2018) estimated the costs related to incineration, including labour, transport and quality control, at US$332–441 t^{-1}, while the products produced (energy and ash) are worth US$ 91.83. Therefore, incineration as a disposal option can only offer partial cost recovery. The impacts of incineration on the environment must also be considered. Incineration of PPMS can release NO_x and SO_2 (which are definitively linked to acid rain generation), as well as particulates, with one estimate of total emissions from paper and pulp mill waste incineration for the USA alone in the year 2005 comprising 40000 t SO_2 and 59000 t NO_x (Pinkerton, 2007). It is also possible that incineration can release potentially harmful chlorinated compounds from any plastic contaminated PPMS or those including residual cleaning agents (Simão et al., 2018). Therefore, infrastructure to treat emissions, such as gas scrubbers, is required, with the associated expense. However, on the other hand, if the mill relies on a mainly fossil fuel derived power supply, sludge incineration could be a way to offset its carbon footprint. Furthermore, paper sludge ash can also be spread to land for its liming benefits, although this will not be further discussed (Tony Marsland, 2015).

Landfilling has been a common disposal method (Scott and Smith, 1995; VEN, 1997) and remains dominant today in some countries (Simão et al., 2018), however, the cost incurred by landfilling is increasing in most countries and new legislation is constantly being developed to reduce landfill waste. An example of such legislation from within the European Union is the landfill limitations Directive 99/31/EC, which states that "biodegradable municipal waste going to landfills must be reduced to 35% of the total amount (by weight) of biodegradable municipal waste produced in 1995" by the year of 2016 and this was followed by the promotion of the waste hierarchy through Directive 2008/98/EC (waste framework directive) which emphasised that disposal of waste, e.g. via landfilling, should be considered a final resort only. Nevertheless, landfilling remains a final alternative after reuse and recycling options are exhausted.

5. Land application

Driven by the financial burden and potentially negative environmental impacts of landfilling or incinerating PPMS, they are increasingly being disposed of in more environmentally positive ways while landfilling itself has decreased (Camberato et al., 2006; Gibbs et al., 2005; Scott, 2019; VEN, 1997). As the chemical composition of paper sludges shares some properties with livestock manure (Bellamy et al., 1995), landspreading of the material emerged as a clear solution to the disposal problem several years ago. In fact, the spreading of paper mill sludge to land has been recorded since the 1950s and has, for example, been considered a common practice for more than 30 years in the UK (CPI, 2015; Norrie and Fierro, 1998). Although transport costs must be taken into account, similar transport costs are incurred by any offsite movement (e.g. for landfilling or incineration). Land spreading consists of either simply spreading a layer on top of the soil (mulching), or application followed by incorporation into the soil through ploughing or other means. The aims of mulching also differ from those of incorporation; mulching is employed to help manage the temperature of soil, reduce evaporation, prevent weed germination and reduce nutrient loss by runoff while incorporation aims to increase the nutrient availability of soils, alter the physical properties of the soils (i.e. bulk density and hydraulic conductivity) or to add organic matter back into the soil.

The use of paper based mulches can also play a key part in the replacing of plastic mulches which can contribute to microplastic pollution of soils (Bandopadhyay et al., 2018). However, a practical consideration is that, as it is only beneficial for land managers to spread PPMS during drier seasons of the year and before or after crop production on arable land, sufficient storage is required during wetter periods if land spreading is to be undertaken as a major recycling route. A third option when applying PPMS in an agricultural setting is to initially use them as animal bedding followed by spreading the used bedding material to land (Faubert et al., 2016). However, this method requires further drying beforehand. Nevertheless, this method is likely to be favoured in some regions, for example in England and Wales where it allows for the material to be used on farm and subsequently spread to land without deployment of a mobile plant permit (i.e. there would be no need to wait for approval from the environmental regulator, as registration of the exemption is completed immediately online).

Possible hazards of spreading PPMS to land can be mitigated by proper regulation which includes taking into consideration factors such as site-specific conditions, soil characteristics, and crop requirements on the site. These precautions and regulatory measures will vary from country to country, for example some members of the EU do not permit paper sludge spreading to agricultural land at all while in others there are specific legal requirements for land spreading (Suhr et al., 2015). In Europe (within the European Union countries) the use of PPMS in land spreading is regulated by two directives, Directives 86/278/EEC (the sewage sludge directive) and 91/692/EEC (standardizing and rationalizing reports on the implementation of certain Directives relating to the environment). The spreading of primary, secondary or deinking sludge

Table 2
A selection of pulp and paper mill sludge composting experiments and their impacts on the materials C:N ratio.

Sludge origin	Duration	Additional materials	C:N before	C:N after	Citation
Paper mill	2 years	Ramial wood, urea and fly ash	109:1	42:1	Gagnon et al. (2001)
Pulp and paper	18 weeks	Tailings, wood ash and cattle paunch	270:1	14–67:1	Campbell et al. (1995)
Pulp and paper	121–169 days	Mineral nitrogen, phosphorus and potassium	218:1	35–54:1	Jackson and Line (1997a)
Paper mill	129 days	None	41.5:1	21.1:1	Evanylo and Daniels (1999)
Paper mill	17 days	Hardwood sawdust	21.5:1	21.9:1	Marche et al. (2003)
Pulp and paper	34 weeks	Fly ash	70.1:1	40–46:1	Hackett et al. (1999)
Paper mill and deinking	28 days	Pinewood bark	42.7:1	28.1:1	Jokela et al. (1997)
Paper mill primary	60 days	Vermicomposting with Eisenia fetida	138.92:1	13.35:1	Ganguly and Chakraborty (2018)
Paper mill secondary			86.21:1	6.61:1	
Paper mill mixed	150 days	Vermicomposting Eisenia fetida	85:1	44:1	Kaur et al. (2010)
Paper mill mixed	56 days	Vermicomposting Eisenia fetida	257:1	72:1	Negi and Suthar (2013)

from paper making is covered by European Waste Catalogue (EWC) Codes 03 03 05, 03 03 10 & 03 03 11 (CPI, 2015).

European Directives are regulated differently in each of the Member States, and still underpin a lot of the UK based regulations at time of writing despite the UK leaving the EU. In England and Wales those who landspread PPMS are regulated via the Environmental Permitting Regulations (specifically Standard rules SR2010No4 Mobile plant for landspreading, (Environment Agency, 2012)), whilst in Scotland and Northern Ireland, applications of wastes for agricultural benefit are still regulated under the Waste Management Licensing Regulations. On farm, additional regulations and codes of practice also cover landspreading and waste storage activities, such as the Reduction and Prevention of Agricultural Diffuse Pollution Regulations (Agriculture England, 2018) which underpins the Farming Rules for Water (Department for Environment Food and Rural Affairs, 2018). Many countries have further governmental advice and best practise codes (e.g. Gibbs et al., 2005).

While research has been conducted into the use of PPMS in construction (Andreola et al., 2005; Balwaik and Raut, 2011; Naik et al., 2004; Thomas et al., 1987), combustion (Coimbra et al., 2015) and ceramics (Asquini et al., 2008), application to land remains the most common beneficial recycling pathway. Land spreading is briefly touched upon in a review by Monte et al. (2009), in which a short overview is presented on some of the logistics, advantages and limitations, while Simão et al. (2018) also included information about land spreading of PPMS within a wider review of paper mill related wastes. Camberato et al. (2006) reviewed the subject of paper mill sludges including land application in North America, however deinking sludges were also discussed interchangeably and, as mentioned in section 2.2., these have very different characteristics leading to differing conclusions being derived than to those that would come about from only studying primary and secondary PPMS. Faubert et al. (2016) reported the greenhouse gas (GHG) impacts of PPMS management, concluding that further research is required into the GHG emissions of PPMS used in silviculture, land reclamation and composting.

6. Composting of sludges

The composting of PPMS before land application can help improve chemical characteristics, reduce pathogenic organism content and reduce the overall volume of waste while increasing its bulk density (Hazarika et al., 2017; Jackson and Line, 1998). Particular interest has been placed on decreasing sludge C:N ratios through composting, with multiple studies having explored this (Table 2). It is especially effective when co-composting with N rich wastes (Camberato et al., 2006; Campbell et al., 1995; Gagnon et al., 2001; Jackson and Line, 1997a), as this not only alters the ratio by virtue of the N additions but also through stimulating enhanced microbial activity that reduces the C content. Most studies agree that composting directly leads to a decrease in C:N ratio (Table 2) because the organic matter present is consumed by microorganisms which release carbon to the atmosphere as carbon dioxide. An additional benefit of composting is that soluble N and P can be converted into organic forms by microbial processes which can reduce leaching and therefore extend their availability to crops (Bajpai, 2015). It can also lead to a threefold increase in CEC (Camberato et al., 2006) and an increase in the nutrient content, including P, K, Ca, Mg, Na, Fe and Mn, as organic matter is reduced (Campbell et al., 1995; Gagnon et al., 2001). In terms of organic matter composition, composting was found to lead to an increase in the concentrations of carbohydrates (from 54.4% relative abundance of total carbon to 62.9% at the beginning of the experiment and after 17 days respectively) and lignins, whereas those of sterols, lipids and proteinaceous compounds decreased (Marche et al., 2003).

The composting conditions have a significant effect on the end product. For example, the composting period must be sufficiently long to enable this process to lead to a notable change, as highlighted by a short term (17 day) trial by Marche et al. (2003) which found the overall C:N ratio was maintained in the PPMS + woodshavings compost investigated. Two other condition which are essential for effective composting include substrate moisture content and temperature. Hubbe et al. (2010) reviewed the effect of compost condition on the composting of different lignocellulosic materials, and found that relative low temperatures (35–50 °C) were required to effectively decompose pulp mill fibers and suggested compost moisture content varied from 40 to 65% (Hubbe et al., 2010).

Composting of PPMS can be conducted *in situ* at the paper/pulp mill or can be conducted by the party who is responsible for land spreading the PPMS however in many countries the operator would require additional permitting to compost on site. Commercial scale composting of PPMS by paper mills dates back to the 1990s although the process has not been adopted by many mills globally (Bajpai, 2015), most likely because of space limitations at mills and additional permitting requirements. In the UK composting is conducted at composting facilities rather than on site, mainly due to space and permit requirements.

Vermicomposting (earthworm enhanced composting) has also been investigated as a method of pre-treatment before land spreading and was found to reduce the overall volume of material, increase the concentration of nutrients and lower the C:N ratio (Table 2), similar to other composting techniques (Butt, 1993; Elvira et al., 1996, 1997, 1998; Ganguly and Chakraborty, 2018; Kaur et al., 2010; Negi and Suthar, 2013; Sonowal et al., 2014). Ganguly and Chakraborty (2018) explored the roles played by microbes during vermicomposting. The results indicate that the activity microbial enzymes arylamidase and β-glucosidase play significant roles in altering C:N ratios.

7. Effects of land spreading on the physical and chemical properties of soils and plant growth

Land application of sludges benefits soils in multiple ways, most of which are derived from their high organic content (this can be as high as 94% in primary sludges) and their liming effect (CPI, 2015; Méndez et al., 2009). These benefits include improving aeration and drainage, improved nutrient cycling, the stimulation of microbial activity, increasing of microbial and fauna populations and immobilising

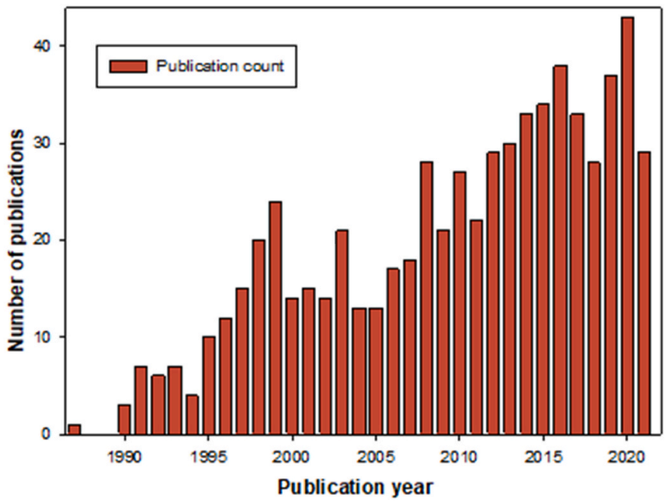

Fig. 3. Number of publications, by year, that feature 'Paper pulp' AND 'Soil' in their topic. Data compiled from Web of Science™ (https://www.webofscience.com).

potentially toxic elements (PTEs) (Battaglia et al., 2007; CPI, 2015; Levy and Taylor, 2003; Norris and Titshall, 2011). There are also known potential risks of N immobilisation in the soil due to the high C:N ratio of the materials, particularly primary sludge, which sometimes requires management to prevent impact on the growing crop. The level of interest in the effects that land spreading of PPMS has on soils and the plants and agro-ecosystems they support has been steadily growing over recent decades, with a major upswing in research publications on the topic since the 1980s (Fig. 3). The principal observations from this research and the remaining knowledge gaps are discussed in the following sections.

7.1. Effects on the physicochemical properties of soils

Nunes et al. (2008) applied 40–120 g kg^{-1} of PPMS that led to an increase in cation exchange capacity (CEC) from 4.26 to 5.00 cmol$_c$/kg and 7.32 to 8.53 in control and 120 g kg^{-1} PPMS application plots of a Cambic arenosol and a Cromic Cambisol respectively after 188 days. Camberato et al. (2006) also reported that PPMS addition typically raises soil CEC substantially, citing examples where heavy applications increased values from 4 to 12 cmol (+) kg^{-1}. Zibilske et al. (2000) found that a single application, biennial applications and annual application of 135 and 225 Mg ha^{-1} PPMS to field plots in Maine (USA) all led to general decrease in soil bulk density over a 4-year period (from ~1 Mg m^{-3} in the first year to 0.8–0.9 Mg m^{-3} in the fourth year). More pronounced effects on bulk density were observed (as low as 0.6 with 225 Mg ha^{-1}) after 5 years, but only under annual application. Chow et al. (2003) found that additions of 20, 40, 80 and 160 Mg ha^{-1} of air-dried pulp mill sludge led to increased hydraulic conductivity and macroaggregate formation (and therefore increasing the ratio of macropores to micropores), thus improving infiltration and water storage in soil with up to 2.1 fold increase in delay in runoff initiation and 23% reduction in total runoff and 71% less soil loss via erosion. Improved soil aggregation and soil stability in a sandy loam following PPMS applications were also reported by Gagnon et al. (2001). Similarly, Zibilske et al. (2000) also reported an increase in soil aggregation after 3 years of PPMS application. However, due to potentially high Na concentrations in some PPMS (e.g. 25000 mg/kg), it is possible that repeated application of such materials could lead to increased sodicity and salinity in soils of warm and dry climates that could potentially cause aggregate instability and slaking and so also inhibit plant development (Abdullah et al., 2015; Cabral and Vasconcelos, 1993). More research on the possibility of this should be conducted, particularly in areas of Africa, Australia, and South America that have sodic soils and low annual rainfall.

Application of PPMS to land is also recognised as having a substantial liming potential, thanks to its neutral to alkaline pH and carbonate. Nunes et al. (2008) for example, found that 80 Mg ha^{-1} of secondary paper mill sludge raised soil (Cromic Cambisol) pH from 6.1 to 7.2. Other studies have reported similar capacity of PPMS addition to raise soil pH (Environment Agency, 2015; Méndez et al., 2009; Shipitalo and Bonta, 2008). Such a liming effect can be beneficial to agricultural landowners and managers as it offsets the cost of purchasing liming agents, thus providing a further incentive to facilitate land-based use of PPMS.

7.2. Organic matter

PPMS are rich in organic matter in the form of lignin and short cellulose fibres, while secondary sludge is further enriched with organic matter in the form of dead microbial biomass. Their applications have repeatedly been proven to increase soil C stores (Gallardo et al., 2012) which is of particular interest in the current agricultural and sustainability markets. At a field scale, Zibilske et al. (2000) showed that single applications of 180 and 225 Mg ha^{-1} of paper mill sludge (dry weight basis) on land used for corn growth led to a significant increase in soil C, and this increase was still significant after five years without further application, although initial effects were not seen during the first year following application, likely due to the time taken for soil PPMS decomposition. However, lower single application rates of 45, 90 and 135 Mg ha^{-1} had no significant impact on soil organic C% over three years. In contrast, annual or biannual applications of this paper mill sludge at 45 Mg ha^{-1} maintained C stores, which might otherwise have been lost through decomposition, within the soil while similar repeat applications of ≥90 Mg ha^{-1} increased soil C over the five years. Nunes et al. (2008) found application rates of greater than 120 Mg ha^{-1} increased soil C significantly by 1.3 g and 1.8 g kg^{-1}. The benefits of a single application of composted paper mill sludge (tested at rates of 45 and 90 Mg ha^{-1}) on soil organic matter content, macroaggregation, and microbial growth and activity have been found to persist even after 3 years of continuous cropping (Gagnon et al., 2001).

7.3. Greenhouse gas emissions

Due to their high organic content, the release of greenhouse gasses (GHG) during the decomposition of PPMS has been explored (Baggs et al., 2002; Faubert et al., 2017, 2019). Land applied PPMS tend to release a peak of N_2O soon after incorporation which is related to the C additions stimulating microbial activity. This N_2O peak was found to last for 3 weeks by Baggs et al. (2002). However, over longer periods Faubert et al. (2017) found that the area-based N_2O emissions produced from PPMS application were similar or only slightly higher than those from commonly used urea fertilizer (fertilizer-induced N_2O emission factors −0.3 and 4.5% vs 0.8–3.1% for urea and PPMS) over two snow-free seasons (Jun–Nov 2013 and 2014). Faubert et al. (2019) compared the GHG emissions of landfilling and land spreading PPMS (equally mixed PPMS and a predominantly primary PPMS mix) and combined it with longer-term (0–100 years) modelling. Their findings indicated that land spreading could reduce GHG emissions by two thirds when compared to landfilling over the longer term. Therefore, while the GHG emissions of land spreading PPMS deserves further exploration, current evidence suggests that it offers a greener alternative to landfilling in terms of emissions.

7.4. Nutrient availability and plant growth

Numerous studies have investigated land spreading of primary and secondary PPMS on agricultural soils and the subsequent effects on crops and plant health. A summary of the reported observations is presented in Table 3 and indicates that, while there are differing results,

Table 3
A summary of studies examining the impacts of incorporating paper and pulp mill sludge on plant growth.

Sludge type	Application rate (s) & method	Co-applications	Crop	Soil type	Main outcomes	Reference
Paper mill sludge	8.5, 17, 34 Mg ha^{-1}	None	Blueberry (*Vaccinium angustifolium*)	Sandy soil (l'Afrique sand); pH 5.0	8.5 and 17 Mg ha^{-1} treatments increased fruit yield by more than double, while 34 Mg ha^{-1} treatment was equivalent to control.	Gagnon et al. (2003)
Paper mill sludge (70% primary: 30% secondary)	0, 20, 100 and 200 Mg ha^{-1} applied as mulch	None	*Brassica rapa* (mustard plant)	Commercial topsoil OM 10%; clay + silt fraction = 55%	20 Mg ha^{-1} enhanced plant survival (142%), flower (116%) and seedpod (125%) development, and a root-length index (111%). Extreme application rates (100 and 200 Mg ha^{-1}) adversely affected plant growth and development.	Bostan et al. (2005)
Paper mill sludge (with ~2% w/w sewage sludge)	0, 50, 100, 150, 200, 250 g kg^{-1} (simulating 0, 112, 224, 336, 448, 560 Mg ha^{1})	0 or 200 kg N ha^{-1}	Corn (*Zea mays* L.)	Sandy loam; pH 7.8	• Delaying sowing until 21 days after sludge incorporation increased mean germination from 88% to 100% in treated soils. • With co-applied N, sludge additions at 112 Mg ha^{1} equivalent increased crop growth (plant dry mass), while higher application rates decreased it. • At 336 Mg ha^{1} and above, sludge applications increased plant P levels.	O'Brien et al. (2002)
Primary pulp mill sludge	0, 10, 30, 50, 70, 90, 110 and 130 Mg ha^{-1}	0.44 g P, 0.22 g K and 0.05 g Mg per 6 kg soil	Yellow lupin plants (*Lupinus luteus* L.),	Cambic Arenosol	• Applications above 50 Mg ha^{-1} depressed yield in the first year (linked to nutrient immobilisation) but not in the second year.	Vasconcelos and Cabral (1993)
Pulp and paper mill sludge	0, 225, 450 Mg ha^{-1}	50 kg P ha^{-1} and 100 kg of K ha^{-1} or 100 kg P ha^{-1} and 200 kg K ha^{-1}, depending on sludge application	Black cottonwood (*Populus trichocarpa* Torr. and Gray) and red alder (Alnus mbra Bong.)	Silty clay loam of alluvial origin	• Increased the yield of black cottonwood • Decreased the yields of Alder • Irrigation was highly significant in its effects on yield of both species	Harrington and DeBell (1984)
Secondary pulp mill sludge	0, 40, 80 and 120 Mg ha^{-1} (equivalent 0–40 g kg^{-1})	140 mg N kg^{-1}, 93 mg K kg^{-1} and 3 mg P kg^{-1}	Wheat (*Triticum aestivum* L.)	Cambic Arenosol (pH 6.4) and a sandy loam Cromic Cambisol (pH 5.0)	• Applications linearly increased wheat grain nitrogen levels (both soils) • Wheat grain P levels unaffected • Marginal effect on grain K on one soil • Grain Ca increased. • Variable effects on grain Cu, Mn and Zn • 40 and 80 Mg ha^{-1} rates increased grain yield on Cromic Cambisol soil led to mild decreases on other soil (more notable decreases at 120 Mg ha^{-1}). • Fertiliser supplements recommended when applying sludge	Nunes et al. (2008)
Primary and secondary pulp sludge	5.1, 8.8, 9.6, 10.9, or 13.8% dry weight	0, 8.4, or 20.7% flume grit on a dry weight basis; with varying combinations of fertiliser comprising triple superphosphate + potash, and N as either 728 kg $^{-1}$ urea or 8.6 Mg ha^{-1} chicken manure.	Grass mix (34% *Lolium perenne* L., 34% *Festuca rubra* L. subsp, rubra, 13% *Poa pratensis* L.,9% *Loliurn rnultiflorum* Lam., and 6% *Trifoliurn repens* L.), or Hybrid poplars (*Populus* spp.).	Artificial soil comprising quarry pit bank sand and the blended additives. Compared with a natural sandy soil control.	• Grass yield (all species) and tree growth were enhanced by sludge applications • Plant tissue P and N concentrations significantly enhanced by sludge treatment	Carpenter and Fernandez (2000)
Primary and secondary paper mill sludge	9:1 soil-sludge weight ratio	None	Barley (*Hordeum distichum*)	Pb and Zn polluted Sandy loam; pH 7.6	• Sludge application improved plant root development and reduced chlorosis symptoms • Decreased plant uptake of toxic levels of metals (Zn and	Battaglia et al. (2007)

(*continued on next page*)

Table 3 (*continued*)

Sludge type	Application rate (s) & method	Co-applications	Crop	Soil type	Main outcomes	Reference
					Pb) by >50% • Reduced peroxidase activity (stress response) in plants by ~50%	
Paper mill sludge	10, 20 and 40 Mg ha^{-1} dry basis	Basal fertiliser added according to unspecified 'fertility recommendations for each soil'	Perennial ryegrass (*Lolium perenne*)	Clay, silty clay and a sandy loam; pH 5.3–6.0	• 10 Mg ha^{-1} rate increased yield in two out of three soils, while higher rates decreased yields • 40 Mg ha^{-1} rate decreased plant N to below critical levels in all soils • Plant P remained in the adequate range at all application rates and was enhanced in plants from two of the soils	Norris and Titshall (2011)
Pulp mill sludge	0, 12, and 24 dry Mg ha^{-1}	NH_4NO_3 at rates of 0, 100, and 200 kg-N ha^{-1}.	Corn (*Zea mays* L.)	Clay-silt loam; pH 5.5–6.6	• Sludge applications with and without added N increased plant emergence • Grain yield increased with sludge application when N was co-applied (yield decreases were observed without N supplement) • Recommended paper sludge application rate of 12 dry Mg ha^{-1} plus 100 kg N ha^{-1}	Bellamy et al. (1995)
Primary sludge	Blends of sludge: organic soil (terre noir) in ratios 0:50, 10:40, 30:20, and 50:0 were mixed with sand (50%) and applied at rates equivalent to 0, 23, 68, and 113 Mg ha^{-1} sludge	N at 4.5–5.5 Mg ha^{-1}, P at 1.18–1.26 Mg ha^{-1}, and K at 1.34–1.46 Mg ha^{-1} · Increasing with sludge content	Kentucky bluegrass (*Poa pratensis* L. 'Georgetown') and perennial ryegrass (*Lolium perenne* L. 'Prelude')	Schist-Loam; pH 6.1	• With nutrient addition, ground cover, turf colour, and stand quality were maintained across all application rates • Without nutrient addition, ground cover and stand quality decreased for some grass species at 68 or 113 Mg ha^{-1}.	Norrie and Gosselin (1996)
Mixed pulp mill sludge-Composted or lime-stabilised	17.7 to 25.3 dry Mg ha^{-1}	250 kg ha−1 additional mineral fertiliser (50 kg N ha-1) in 2016.400 kg ha−1 in spring 2017 (80 kg N ha-1)	Wheat (Triticum aestivum) and oat (Avena sativa)	Sandy Clay; pH 6.2	• Higher yields than an untreated control plot. • Yields were comparable to those of a mineral fertiliser only plot. • Three out of four sludge treatments did not reduce nitrogen uptake compared to the mineral fertiliser treatment.	Kinnula et al. (2020)

immobilisation of N or other mechanism leading to N availability limitation is a common occurrence that requires monitoring and/or management. This can occur due to the large C pool provided by PPMS that can stimulate microbial activity and growth which in turn requires the consumption of N, and if there is not enough readily available from the PPMS itself the microbes will utilise (i.e. immobilise) N from the soil (Chen et al., 2014). Such effects were likely involved in a study by Norris and Titshall (2011) where applying as little as 10 Mg ha^{-1} of paper mill sludge reduced N uptake by ryegrass (*Lolium perenne*) significantly. Other studies similarly found that reduced nutrient uptake by common crops can often be a side effect of applying PPMS to soil above certain rates (Bellamy et al., 1995; Camberato et al., 2006; Carpenter and Fernandez, 2000; Norris and Titshall, 2011; Nunes et al., 2008) and this is managed through controlled applications in the UK that take account of site-specific factors. To counteract or mitigate against this, the benefits of co-application of nutrients have been widely reported in the literature (Bellamy et al., 1995; Camberato et al., 2006; Nunes et al., 2008; O'Brien et al., 2002), while it would also be possible to positively exploit N immobilising effects by applying PPMS in situations where there may be excessively high levels of available N (i.e. as a positive land management approach in nitrate vulnerable zones).

However, there are conflicting reports in the literature with some having concluded variously that additions of PPMS to agricultural soils can either lead to an increase or decrease in availability of nutrients, including N (Table 3). Both studies reported that N which was initially immobilised became readily available again after two years of cropping, and in the case of Aitken et al. (1998) the overall N availability increased compared to pre-application levels. In a similar outcome, Vasconcelos and Cabral (1993) found that, after producing significant negative impacts on yellow lupin (*Lupinus luteus*) growth during the first year of cropping when PPMS were added at 50 Mg ha^{-1} and above, the negative effects were not observed in the second year. They also noted that Mn, Zn and P uptake by yellow lupin decreased almost linearly under applications of 10–130 Mg ha^{-1}, which contrasts sharply with increases in the availability and/or plant uptake of these and other nutrients reported elsewhere (Table 3). For example, application of secondary PPMS resulted in a linear increase in available P content (Egner-Riehm method) when applied at rates of 40–120 Mg ha^{-1}, rising from 88 to 196 mg P kg^{-1} and from 120 to 206 mg P kg^{-1} at the highest treatment in a cromic arenesol and cromic cambisol soil, respectively (Nunes et al.,

Table 4
Typical concentration (mg/kg) ranges reported for potentially toxic elements (PTEs) in primary and secondary pulp sludge, and deinking sludge on dry matter basis (compiled from[a]Cabral et al. (1998); (Ribeiro et al., 2010)[b]; (Deviatkin et al., 2015)[c].

Element	Primary sludge[a]	Secondary sludge[a]	Primary sludge[b]	Secondary sludge[b]	Deinking sludge[c]
Cu	7–58	25–77	13	2.8	64–345
Zn	30–40	40–130	83	12.9	34–1320
Cd	0.3–3	1–9	1.4	0.34	0.02–1.54
Cr	3.6–10	12–38	19	1.9	4.8–96.6
Ni	5–75	10–26	10.5	1.5	<10–31
Pb	7.4–74	20–100	13.2	1.1	9.5–79.4
Hg	0.8–1.2	0.5–3	–	–	0.1–0.9

2008), with similar increases recorded for available K. Elsewhere, combined primary and secondary PPMS applied at 34 Mg Ha^{-1} significantly increased soil available N, P and Mn in a low nutrient sandy soil (Gagnon et al., 2003).

The method of PPMS application also seems to have an influence on its effects on plant growth, as while the studies above mainly involved PPMS that underwent incorporation into soil, using mixed PPMS (224:12 C:N ratio) as a mulch has been reported to increase the available N, Ca, and Mg (N determined by KCl extraction, and Ca and Mg via soil-saturated paste extract) as well as increase wheat (*Triticum aestivum*) N and K uptake, when applied without any coapplication of fertiliser (Amini et al., 2012). That study compared mulching versus incorporation and found that, when compared to soil incorporation, un-amended controls and separate treatments where only N, P and K fertilisers were applied (at up to 92, 50 and 83 kg ha^{-1} respectively), mulching at 100 Mg ha^{-1} produced the greatest yield of wheat (*Triticum aestivum* Var. Tajan) save for one of the fertiliser treatments while the PPMS incorporated treatments (applied at 50 and 100 Mg ha^{-1}) produced the lowest yields. Relatedly, when PPMS were trialled as a mulch for turfgrass growth, Karcher and Baser (2001) found that PPMS provided a viable, cheap alternative to the commercial mulching product hydromulch (wood fibre based) with no significant drawbacks in terms of grass height, turf cover, and soil water infiltration.

Not all plants are affected in the same manner by PPMS application. For example, Harrington and DeBell (1984) found the same treatments increased the yield of black cottonwood (*Populus trichocarpa*) while decreasing the yield of Alder (*Alnus rubra*). This was observed at application rates of 225 and 450 Mg ha^{-1} of PPMs that had been corrected to a C:N ratio of 100:1 by N fertiliser additions. Yield impacts have also been shown to depend on the soil type involved (Nunes et al., 2008).

The use of PPMS as a media for seeding and germination has also been explored (Bellamy et al., 1995; Levy and Taylor, 2003). Through a series of nursery container studies Bellamy et al. (1995) confirmed that paper sludge amendments were suited to use in nursery container cultures for a range of species (tomato (Lycopersicon esculentum L.), cucumbers (Cucumis sativus L.), and peppers (Capsicum annuum L.)), sufficing that the low N content of sludges is corrected for (to approximately 1.2% dry w/w). When germinating radish in pulp mill sludge alone, Levy and Taylor (2003) found that radish (*Raphanus sativus* L.) and cress (*Lapidium sativum* L.) germination rates were not reduced when compared to control soil, but seedlings were smaller, lighter coloured, misshapen and displayed necrotic patches (Levy and Taylor, 2003), indicating that raw or unamended PPMS is not universally suitable as a germination medium.

7.5. Potentially toxic elements

The concentration of potentially toxic elements (PTEs) in any type of land amendment, be it PPMS, sewage biosolids, or mineral fertiliser, is always an issue that may pose concern. However, while paper sludges may contain traces of PTEs such as cadmium, copper, lead, and nickel, with primary paper mill sludges tending to have higher levels than secondary sludges (IPCC, 2001), the concentrations are typically low and similar to those found in commonly land-spread livestock manure (Bellamy et al., 1995; CPI, 2015) (Table 4). Indeed, concentrations in paper sludges remain lower than those typically found in sewage sludge which is applied to land as common practice (Deviatkin et al., 2015; Epstein, 2002). de Azevedo et al. (2019) found that As, Pb, Cr, Fe, Al, Mn, Zn and Cu in leachate and solubilised extracts (Brazilian standard for leaching (NBR 10005, 2004)) from paper sludge fell within the concentration limits for non-inert non-harmful wastes as set by Brazilian standard and the US EPA – Code of Federal Regulations. PTEs can be a concern linked to deinking sludge which may have higher concentrations of certain PTEs (e.g. Cu, Zn and Cr) due to the processes employed to remove the ink, however these concentrations have reduced in recent years to comply with packaging regulations (Environment Agency, 2013; Scott and Smith, 1995). Therefore, land application of de-inking sludge (alone or in combination with primary and/or secondary sludge) is monitored closely in most developed countries. In general, therefore, it has been concluded that paper sludge application to land, particularly at the rates relevant to agricultural practises, will present little risk of metals accumulation problems.

As opposed to posing a metals risk, retention of PTEs by paper sludges can help reduce their availability to crops and other ecological receptors and thereby decrease or even neutralise toxicity. Most early studies with this perspective focussed on the use of paper mill sludge for removing PTEs from solution, which they proved to be highly capable of even at low pH (Baek et al., 2014; Calace et al., 2002, 2003). Later soil studies demonstrated that the paper sludges were also able to immobilise PTEs in the soil, for example Battaglia et al. (2007) found that the addition of paper mill sludge (1:9 sludge to soil) to Pb and Zn contaminated soils reduced the availability of both elements when measured by a five step sequential extraction with increases in the i) sulphide/bound to organic matter but not soluble in NaOH fraction and ii) the non-extractable metals fraction (overall increase in these non-labile fractions of 11% and 8% for Pb and Zn respectively). These sorption effects are likely caused by the sorption of elements onto clay and organic matter within the sludge in addition to the immobilisation brought about by the increase of pH caused by sludge application. In fact, Calace et al. (2003) found that due to these properties of paper mill sludges (i.e. high lignin and clay content) they have similar metal ion sorption trends to clay and organic matter rich soils.

Battaglia et al. (2003) showed that paper mill sludge amended soil (9:1 9:2, 9:4 and 9:8 soil:PPMS w/w) has a greater Pb sorption than the original soil (Dystric xerocrept), but the Cd sorption differences were negligible during batch sorption testing. In a column leaching test by Calace et al. (2005) it was also found that Cd leaching was decreased to a lesser extent than that of Pb (viz. Cd leaching was reduced by ~30%) when applying sludge at a 1:9 PPMS:soil w/w ratio. In another study, up to 115 μg l^{-1} of Zn was leached from a 10 g sample of primary paper mill sludge during column leaching experiments when placed on top of 20 g of soil and leached with 60 mL of water (1.7 pore volume), while this concentration gradually decreased in subsequent leachings until it was below the limits of detection after six leachings (Xiao et al., 1999). However, leaching of Zn from the soil could have also contributed to this figure. In the case of Cd and Pb, no appreciable amounts of Pb or Cd were found to leach (Xiao et al., 1999). Analysis of the column soils after the experiment revealed that the PPMS additions had enriched the soil total Ni, Pb and Zn contents by < 0.04 mg/kg.

Battaglia et al. (2003) modelled sorption isotherms of PPMS for Pb and Cd and found them to be best modelled using Langmuir isotherms. This was in agreement with the findings of Calace et al. (2002) who also found that Ag(I) and Cr(VI) were best fit by Langmuir isotherms while Cu (II) sorption was best fit by Freundlich. However, Yoon et al. (2017) found that As and Cd sorption by PPMS were better fit by the Redlich-Peterson isotherm, which combines elements of the Langmuir and Freundlich isotherms.

7.6. Organic pollutants

Industrial plants that employ chlorine dioxide during the bleaching process can lead to the presence of organochlorine compounds in pulp and paper sludge (Simão et al., 2018). For example, Koistinen et al. (1994) found that sludge from pulp mill discharge where chlorine is employed as a bleaching agent contains trace amounts of methylfluorenes and dimethylfluorenes (0.5 and 0.06 ng l^{-1} respectively). Similarly, 2,3,7,8-tetrachlorodibenzo-p-dioxin was found at trace levels in effluents from pulp and paper mills in India (Thacker et al., 2007). However, these issues are not prevalent in developed countries today because legislation limiting the use of elemental chlorine in paper making has led to its replacement, mainly by hydrogen peroxide or ozone (Wolf et al., 2011). Nevertheless, other organic contaminants can still be present. For example (Rigby et al., 2021) determined estimated toxic equivalents (TEQs) for dioxins/furans, dioxin-like biphenyls and polychlorinated naphthalenes approximating 10 ng/kg (dry mass) in three paper sludges from the UK, but these concentrations in the paper sludges were much lower than those determined in sewage biosolids and some poultry litter ash.

Per- and polyfluoroalkyl substances (PFAS) are a group of contaminants receiving increasing attention. They have been used in the coating of paper food packaging, and therefore can be incorporated into PPMS where recycled paper products are used as a feedstock (Wiegand, 2021). PFAS are a family of >4700 persistent chemicals which are a growing human health concern because of their accumulation in plant and human tissue and evidence of their possible links with multiple health effects including hypercholesterolemia, hyperuricemia, decreased glomerular filtration rate (GFR) in kidneys, increased chronic kidney disease, kidney cancer, and testicular cancer (Costello and Lee, 2020; Kirk et al., 2018; Pelch et al., 2019). Sewage biosolids are known to be a potential source of PFAS, with concentrations up to 2615 μg/kg having been reported for PFOS in digested sludge (Jensen et al., 2012). Thus, sewage biosolids treated soil can accumulate PFAS compounds; for example, in Alabama soil concentrations of up to 990 μg/kg for perfluorodecanoic acid, 530 μg/kg for perfluorododecanoic acid and 410 μg/kg for perfluorooctane sulfonate (PFOS) were reported (Washington et al., 2010), while a study from Arizona found lower PFAS concentrations (8.6 μg/kg) in agricultural soils that received >67 t/ha biosolids (Pepper et al., 2021). PPMS application could thus also be a potential source of PFAS in soils; indeed Rigby et al. (2021) examined bioresource materials from the UK and found the sum of 9 PFAS compounds to have concentrations of up to 231 μg/kg in sewage biosolids and up to 35 μg/kg in dried paper sludge. However, in one case in Germany PFAS contaminated compost (including paper sludge) was believed to be the source of soil contamination in an agricultural site in which the PFOS + PFOA soil concentration was reported to be as high as 6300 μg/kg (cited in Röhler et al. (2021)). Further work in Germany on those agricultural soils where contaminated PPMS had been applied identified, either fully or tentatively, 61 PFAS compounds and their transformation products (Bugsel and Zwiener, 2020); that study estimated soil concentrations of PFOS to be up to 100 μg/kg and those of PFOA to be up to 250 μg/kg at the study site. Regular monitoring of PFAS levels and setting appropriate regulatory threshold limits in PPMS and other land amendments, such as that now established in the regulations under the German Fertilizer Ordinance which limits the sum of PFOA + PFOS to 100 μg/kg dry weight (Röhler et al., 2021), would prevent any similar pollution incidents occurring in future. Monitoring of PFAS in soils amended with sewage biosolids, urban composts and PPMS should also be undertaken and assessed against the best available estimated no observed effect level (e.g. 1000 μg/kg soil; (Jensen et al., 2012)). As is the case with sewage biosolids and animal manures applied to land, PPMS are also known to potentially contain the endocrine disruptors bisphenol A, nonylphenol ethoxycarboxylates and 17α-Ethinylestradiol (Dsikowitzky and Schwarzbauer, 2014; Fernandez et al., 2007). Research is therefore warranted to determine the extent to which these chemicals persist in soils treated with all such amendments and also into whether treatment steps can be introduced in their production lines to remove or reduce them.

7.7. Odour

Similar to other land spread materials such as anaerobic digestate, odour can be a concern when spreading PPMS near residential areas, particularly during the initial 30 days following spreading during which the material still has a high water content and thus can create anaerobic conditions and associated smells (Amberg, 1984; CPI, 2015; Frechen and Köster, 1998). Historically, when PPMS were discharged to river systems, there were observed impacts on the odour of water downriver of the discharging point (Kenefick et al., 1995). Odour in pulp mill treatment ponds was attributed mainly to the release of Geosmin ($C_{12}H_{22}O$) at 2000–9000 times the odour threshold (i.e. the concentration at which it is perceivable by the human sense of smell), likely released due to the breakdown of organic matter by microbial activity (Watson et al., 2003). Indeed, odour associated with PPMS is typically associated with material that has been in prolonged storage. The spreading of PPMS may be limited based on their odour emissions, as is the case in Quebec where all landspread materials are categorised by human-perceivable odour, and PPMS are considered to be amongst the most odorous materials and hence classified as "strongly malodorous" as measured by olfactometery (Camberato et al., 2006; Environnement Québec, 2004). This classification categorises the smell as being stronger than that of solid dairy cattle manure, but not as strong as hog slurry. The odour released by PPMS can reportedly be reduced by composting, although no study has quantified this (Bajpai, 2015). In the UK, PPMS are not typically malodorous, although risks from odour can occur through poor management of materials. Furthermore, risks associated with odour are managed through incorporation into soil promptly after spreading, and risk assessment prior to spreading to take account sensitive receptors and prevailing winds.

8. The impacts on soil ecology

8.1. Effects on fauna

Prior to the 1990s the dominant method for bleaching paper pulp employed elemental chlorine, resulting in the formation of dioxins and related chemicals in the pulp waste including 2,3,7,8-Tetrachlorodibonzo-p-dioxin (TCDD), which is a known carcinogen (Axegård, 2019). Keenan et al. (1990) explored the potential risks of surface applying PPMS in woodland in the USA to American woodcock (*Philohela* or *Scolopax* minor) and to humans that consume them due to the presence of TCDD within PPMS. The highest reported concentrations of TCDD in PPMS treated woodland soils was 50 ng/kg (ppt) and considered not high enough to cause any risk to woodcock health or reproduction, nor to humans who consume them. Since the mid 1990s the majority of pulp bleaching worldwide has been conducted using non-elemental chlorine methods (most commonly via gaseous chlorine dioxide and oxygen, so called Elemental Chlorine Free (ECF) bleaching; (Axegård, 2019)), which has vastly reduced the potential for dioxin production during pulping and thus its presence in PPMS.

There are very few studies in the literature investigating the potential effects of PPMS land application on earthworms, or on any other soil invertebrates vital to soil ecosystems. Nevertheless, a few do exist. Piearce and Boone (1998) conducted a series of investigations and experiments on this topic, in which the first examined the earthworm population in a sandy soil in north west England (pH 5.77) that had been treated with PPMS 4 years earlier at a rate of 200 Mg ha^{-1} and had been used to grow flax (*Linum usitatissimum*) but received no further nutrient addition. Across ten x 25 cm soil cubes excavated from the treated area they found 38 earthworms from two species (*Aporrectodea caliginosa* and *Octolasion cyaneum*), compared with just one individual *A. caliginosa* from across the equivalent untreated control samples, indicating enhanced earthworm presence in PPMS treated areas. In subsequent laboratory tests on various soils the authors observed no behavioural avoidance by *Aporrectodea rosea* earthworms of soil amended with 20% (w/w) PPMS, and indeed they even noted a selection preference by the earthworms for treated soil in one case where the unamended soil had a pH of 3.9 (the treated soil had a pH > 6.6). Similar behaviour was recorded in tests with the garden snail (*Helix aspersa/Cornu aspersum*), indicating no adverse effects (Piearce and Boone, 1998). When 20 Mg ha^{-1} PPMS were applied to a commercial topsoil in a laboratory test, no adverse effects on survival or cocoon production of earthworms (*Lumbricus terrestris*) were observed, while the number of juveniles present was higher in treated soils. In another study, at a restored landfill site which had been amended with a 50:50 PPMS:soil mixture to 40 cm depth and then surfaced mulched with PPMS and inoculated with earthworms, a successful earthworm community spanning 12 species across a range of ecology types (anecic, endogeic and epigeic) had established over a six year period following PPMS addition (Piearce et al., 2003). Moreover, $\delta^{13}C$ isotopic analysis revealed that the earthworms were consuming the PPMS as a food source.

Butt (1993), examined the growth of earthworms (*Lumbricus terrestris* and *Octolasion cyaneum*) when allowed to mature in loamy soil (steam-sterilised) with surface applied (i.e. mulching) paper mill sludge (50 g:150 g sludge:soil w/w) in a pot trial. Three different yeast extracts (0.75 g) were added as an additional N source to selected pots (corrected to a C:N ratio of 25:1 from 93:1). After 120 days the development of earthworms was severely hindered by the PPMS only treatment (i.e. without supplemental yeast), to the extent that none of the earthworms had sexually matured and mean masses were lower, viz 0.7 g and 0.4 g were achieved versus >3 g and >1 g in all supplemented treatments for *L. terrestris* and *O. Cyaneum* respectively. However, mortality of *L. terrestris* was lower in the PPMS only (no yeast) treatment after 120 days (20% vs 40–50%), while for *O. Cyaneum* the mortality was ~20% in three treatments but 80% for one of the PPMS plus yeast treatments. The results highlight that PPMS addition, and that of any supplementary N sources, can have varying influence on growth and development of different earthworm species and so warrants further research. However, it should also be noted that the application rate in that particular study was very high, beyond what would be used in a typical setting of application to agricultural land and was more in keeping with land restoration application scenarios in which the existing substrate at the sites is likely to be very hostile to soil biota and thus in need of improvement that could be brought about by PPMS addition. Indeed, a Department of Environment, Food and Rural Affairs (DEFRA, UK) funded study (Environment Agency, 2015) found that when applied to an agricultural plot at 75 t/ha paper mill sludge had no negative effects on soil mesofauna and actually increased soil biomass N and potentially mineralisable N.

Although not specifically or directly focused on effects in a land-spreading scenario, there have been studies that investigated the relationship of PPMS and earthworms during vermicomposting of PPMS and/or the use of PPMS as an earthworm growth medium that do give some further insight into earthworm related issues. Elvira et al. (1996) explored the conversion efficiency of pulp mill sludge into vermicompost and the impacts on earthworm (*Eisenia andrei*) growth and reproduction. When mixed in a 3:1 ratio of pulp mill sludge:sewage sludge, the mixture was found to be an ideal medium for composting earthworm growth and reproduction. In a follow-up growth and mortality study using *Eisenia andrei,* Elvira et al. (1997) confirmed that the 3:1 ratio of pulp mill sludge:sewage sludge performed the best as a habitat for earthworm development when compared with mixtures of 1:1, 2:1 and 3:1 of paper pulp mill sludge with sewage sludge, pig slurry or poultry slurry. In a later study, (Elvira et al., 1998) compared earthworm growth medium potential of paper mill sludge to that of cattle manure, dairy sludge and combinations of the three and found that while PPMS led to the greatest increase in mean earthworm mass after 70 days, it also led to the greatest inhibition of cocoon production. However, beneficial effects on both earthworm growth and cocoon production can be achieved by combining it with cattle manure and dairy manure (1:4 PPMS: cattle manure or 1:1:3 PPMS:dairy manure:cattle manure). Similarly Kaur et al. (2010) found that co-composting with cattle dung increased the materials acceptability for earthworm (*E. fetida*) reproduction. Therefore, having been found to be suitable for vermicomposting and as an earthworm growth medium (with minor amendments), as well as having no notable negative impacts reported in the few land application studies that have examined earthworms, it seems unlikely that PPMS would pose an ecotoxicological threat to earthworms when applied to soils. Nevertheless, the longer-term influence of PPMS application on earthworms is a potential future research topic.

8.2. *The soil microbiome*

Gagnon et al. (2001) explored the microbial biomass (chloroform fumigation-extraction method) and enzyme activity of soils treated with raw and composted PPMS (combined with ramial wood, urea and fly ash). Both treatments led to increased microbial biomass relative to a control plot in the two years following application, however the magnitude of increased microbial biomass decreased with time. However, a longer term study in which PPMS had been applied to agricultural soil annually for 6–9 years showed a 1.5–2 fold increase in topsoil microbial biomass (Environment Agency, 2015).

Gallardo et al. (2012) found that soil incorporation of up to 30 Mg ha^{-1} of PPMS had very limited effects on overall structure of fungi and bacterial communities (as determined by denaturing gradient gel electrophoresis), but that a small number of new strains of fungi were introduced by the sludge and some of the bacterial strains native to the soil were reduced in abundance or disappeared from the soil. An aragose gel electrophoresis study into *E. coli* and other possible pathogens in PPMS found that the E. coli isolates found in PPMS were not pathogenic and were likely of environmental origin, while no other potential pathogens were identified (Croteau et al., 2007).

Flemming et al. (2017) conducted a series of plate experiments to assess the microbiological quality of PPMS in Ontario. It was found that pathogens, *Salmonella, Cryptosporidium* and *Shigella*, appeared in 6–8% of samples (n = 93) and at low concentration (2 MPN g^{-1} dry wt., 9 oocysts g^{-1} dry wt., and 7 cells g^{-1} dry wt. respectively). However, *E. Coli* exceeded limits set by the regional branch of the Canadian Fertilizers Regulations which uses a different unit of measurement (1000 colony forming units g^{-1} dry wt.) in a third of the samples, most of which were fresh samples as opposed to lagoon or stored samples. Additionally, *Giardia,* a microscopic parasite that causes diarrhoea, was present in 19% of samples at a mean concentration of 30 cysts g^{-1} dry wt. Overall, mills fed by recycled material contained more Bacterial contaminants, while those fed by virgin fibre were more commonly found to contain *Giardia* which is likely to persist until land spreading (Flemming et al., 2017).

8.3. *Run-off ecotoxicological impacts*

To assess any impacts on waters receiving run-off from treated fields, a series of aquatic ecotoxicological bioassays were conducted by Bostan et al. (2005) using runoff from a commercial topsoil treated with surface applications of 70% primary: 30% secondary PPMS at 20 Mg ha^{-1} (equivalent dry weight) collected after a one day 20 mm rainfall event equivalent (the run-off was generated in a laboratory rain simulator with a soil bed of angle 15% slope). Runoff was diluted with dechlorinated tap water to 0% (control), 10%, 25%, 50% and 100% of the original concentration. *Daphnia magna* (waterflea), *Hyalella Azteca* (an amphipod crustacean), *Selenastrum capricornutum* (algae), *Lemna* minor (duckweed) and *Gambusia affinis* (mosquitofish) were chosen as bioassay species. No effects were observed in mosquitofish at any concentration, while for the other species there were generally no negative effects at the

Table 5
The benefits and limitations of land spreading primary and secondary paper and pulp mill sludges.

	Benefits	Limitations/usage notes
PPMS in general	• Increased soil organic matter • Increased soil cation exchange capacity • Improved soil water holding capacity • Liming effect (soil pH enhancement) • Increased soil stability/ macro-aggregation • Enhanced soil microbiological activity • Weed suppression • Improved plant health and growth possible	• Nutrient addition (particularly N) along with PPMS application is sometimes recommended because PPMS can temporarily decrease nutrient availability to some crop or pasture types. Alternatively, composting of PPMS first may also address this issue. • Odour potential needs consideration
Primary PPMS	• More widely available than secondary PPMS	• High C:N ratio, may lead to N immobilisation initially (increasing the need for nutrient supplements) • Lower micro-nutrient content than secondary PPMS • Higher PTE content
Secondary PPMS	• More desirable C:N ratio • Can contain appreciable amounts of P	• More difficult to dewater • High biological activity can make handling more difficult • Not produced at all mills

concentrations of run-off considered environmentally relevant (i.e. 25% and below). At higher, less environmentally relevant, concentrations, negative effects were observed in all species except for the mosquitofish. These effects at high concentrations may have been linked to increased chemical oxygen demand in paper mill sludge run-off (Shipitalo and Bonta, 2008) but, as the concentration of runoff in receiving waters would be extremely unlikely to ever reach such levels, these results indicate that run-off from well managed land spreading of PPMS is unlikely to have detrimental effects on the aquatic environment. Indeed, PPMS application typically increases water retention in soil (Environment Agency, 2015) and was found to reduce run-off fourfold to sixfold and to decrease soil erosion (e.g. from 47 Mg ha^{-1} to $<$1 Mg/ha in a restored coal mine trial; Shipitalo and Bonta (2008)).

9. Summary and conclusions

As the economic costs of PPMS disposal are increasing, and with the growing desire to re-cycle and re-use resources within a more circular economy, land spreading continues to offer a suitable and potentially environmentally positive alternative to landfilling or incineration and should be encouraged where possible. While the physicochemical characteristics of PPMS can be partly predicted by a number of factors, namely the feedstock of the mill, the treatment methods and the end product of the processes employed, the variable nature of PPMS leads to the conclusion that the most robust basis upon which to verify suitability for land application is via a case-by-case chemical and physical analysis (or at least via a routine, periodic analysis of PPMS generated at a facility); such systems are already in place in many countries. Application methods appropriate for a site, i.e. mulching or incorporation, can be considered based on the land's requirements and application goals (e.g. soil enhancement, weed suppression or full scale land remediation). Both mulching and incorporation have a financial incentive as it decreases or removes the need to purchase commercial mulching products or liming/organic matter amendments, although coapplication at higher application rates may be required. While all PPMS are comparable in many ways, the content of primary and secondary sludge is highly important and so must be borne in mind when considering land application, i.e. primary PPMS tends to have a less favourable C:N ratio and often lower nutrient levels overall compared with secondary PPMS while secondary PPMS may be more difficult to dewater and handle.

Secondary PPMS is often more in demand for agriculture due to its increased liming capabilities and lower C:N ratio, whereas co-application of a nitrogen source is beneficial when mixed or exclusively primary PPMS are land spread. Alternatively, composting can be employed to produce a material with a more ideal C:N ratio for land application. Therefore, where composting opportunities may be available it should be considered, especially since land spreading is limited by seasonality and storage is part of typical practise. While this study did not consider deinking sludge, Camberato et al. (2006) came to similar conclusions as to land application of that material.

Long term studies into the impacts and benefits of PPMS on agricultural fields are rare, however those that do exist suggest that any benefits to soil C are long lasting, while N immobilisation issues are alleviated by nutrient co-application and/or by natural processes occurring in the year after spreading. Leaching of pollutants has been shown to be an unlikely problem under most conditions, while sorption of pollutants is an added benefit of PPMS application. Furthermore, when applied at typical field concentrations the ecological impacts on terrestrial ecology and from runoff are likely to be insignificant, especially when coapplied along with other materials. The benefits and limitations of land application of PPMS, including points specific to primary and secondary types, are summarised in Table 5.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: The second author, Rebecca Wheeler is employed by a consultancy engaged in land-spreading operations.

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