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Paper mill biosolids application to agricultural lands: benefits and environmental concerns with special reference to situation in Canada

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Abstract

The pulp and paper industry plays an integral role in the global economy. The primary concern, however, is the use of chlorine-based bleaches and resultant toxic emissions to air, water, and soil after dumping of biosolids (sludge) that are produced as a by-product. Large amounts of this sludge are being accumulated in the neighborhood of the paper mills, causing serious handling and pollution problems, especially under an increasingly tight economic situation and strong environmental protection policies. Best approach from the economic and ecologic standpoints over the long run is to dispose paper mill biosolids on the agricultural land. We present a review on the application of paper mill biosolids to agricultural lands and its effects on crops, soil physical, chemical and biological properties.
Keywords: Paper mill biosolids, land application, crop growth, environmental effects

Introduction

The pulp and paper industry plays an integral role in the global economy. The sector's global annual revenue from its over 300 million tons of products, exceeds 500 billion dollars (CPBIS, 2003). There are about 500 kraft mills and thousands of other forms of pulp and paper mills in the world. Primary concerns include the use of chlorine-based bleaches and resultant toxic emissions to air, water, and soil. With global annual growth forecast at 2.5%, the production of paper mill sludge is projected to rise between 48 and 86% over 2000 levels. This rise means that current paper mill biosolids production levels of 3 million tons y^{-1} will increase to somewhere between 4 and 5 million tons y^{-1}, creating up to 2.5 million tons y^{-1} of extra paper mill biosolids to be disposed of in 2050.

The total pulp production of Canada was 31.2 million tons per year and exported 10.2 million tons worth $23 billion in 2004 (PPPC, 2005). The Canadian industry defends itself by citing its investment of six billion dollars towards environmental upgrades since 1990 (FPAC). While use of elemental chlorine – one of the worst toxic offenders of the industry – has been all but eliminated, and water consumption has decreased, there remains ample evidence of the industry's ongoing damage to the environment especially fresh water fish (McNair *et al.*, 2003). The pulp and paper industry remains the third largest industrial polluter in Canada. On a national scale, the benthic invertebrate surveys most commonly revealed a eutrophication response pattern as a result of exposure to pulp mill effluent. Effects were also observed on fish downstream effluent, with the overall response pattern being one of combined metabolic disruption and nutrient enrichment. Though effluent quality has vastly improved since the 1992 Pulp and Paper Effluent Regulations mills continue to have an impact on fish and their habitats (Environment Canada, 2003).

Paper mill biosolids are produced as the by-product of the pulp and paper industry. The characteristics of biosolids are variable and directly related to the technology used to pulp, the wood and manufacture the paper and to the type of effluent treatment that is employed. The pulp and paper mill industry in Canada produces 1.5 million tons of paper mill sludge annually, originating from primary and secondary waste water treatment processes (Marche *et al.*, 2003) which, goes to landfills (41%), burned (54%) or applied to land (5%). Because of the different disposal methods, sludge pollutes soil, air, and water. Traditionally, most of the paper mill biosolids were incinerated or land filled. This has considerably changed ever since, with stronger restriction in landfill, mechanization of paper production process and wastewater treatment systems. The best approach from the economic and ecologic standpoints over the long run is disposal of paper mill biosolids on the agricultural land (Dolar *et al.*, 1972). The application of paper mill biosolids to agricultural lands is gaining momentum as the amount applied

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to agricultural lands in Ontario and Quebec was 35% and 38%, respectively of the total volume of biosolids generated in these two provinces (Curnoe, 1998; Gagnon and Ziadi, 2004).

Large amounts of this sludge are being accumulated in the neighborhood of the paper mills, causing serious handling and pollution problems, especially under an increasingly tight economic situation and strong environmental protection policies. We present a review on the application of paper mill biosolids to agricultural lands and its effects on crops and soil properties.

Biosolids produced by paper mills

The sludges can be divided into several categories including i) the waste paper coming from the production of virgin wood fiber or primary biosolids, ii) the waste paper produced by removing inks from post-consumer fiber or de-inking paper biosolids, iii) the activated sludge from the secondary treatment systems or secondary biosolids, and iv) combined wastepaper and activated sludge or combined biosolids. Paper mill biosolids contain several plant nutrients including N, P, K, Ca and Mg. However, nutrient concentrations of paper mill biosolids vary according to the pulping method used and the level of microbial decomposition that has occurred during secondary treatment (Vance, 2000). Nitrogen contents and C:N ratio of paper mill biosolids vary widely depending on the type of paper being produced, the paper producing process, and the type of raw material used. Primary biosolids consist of organic matter mainly in the form of cellulose (Jackson and Line, 1997) or wood fiber settled out in a primary treatment and usually contain 0.3% or less nitrogen by dry weight, with a C:N ratio >100:1 (Bellamy *et al.*, 1995).

De-inked paper mill biosollids is mainly composed of short wood fibers and kaolin clays used during paper making processes, and inks and chemicals used for paper recycling purposes. De-inking paper mill biosolids is very similar to primary biosolids because it is composed mainly of wood fiber but also contains fillers, ink and chemicals used to dissociate these materials from the fiber (National Council of the Paper Industry for Air and Stream Improvement (NCASI), 1991). Composition of de-inking sludge varies considerably due to the origin of the recycled paper and to the de-inking process employed. However, C content is consistently high while N and P contents are consistently low (NCASI, 1991; Trépanier *et al.*, 1996) which is similar to most wood residues like bark, sawdust, wood chips, etc.

Secondary biosolids contain fibers and other fine materials that were not removed in the primary treatment. Bacteria decomposed the remaining organic matter that is contained in the water, i.e., the sugars and other constituents such as cellulose, and then they are collected through decantation of the treated water. Nitrogen, phosphorus and potassium fertilizers are added in the secondary treatment to allow microbial growth and activity. Therefore the secondary biosolids are very rich in N (3.0-4.0%) and moderate in P (0.1-0.3%) content (Bellamy *et al.*, 1995; Zibliske, 1997).

Combined paper mill biosolids are prepared by mixing primary and secondary biosolids in different proportions (50:50; 40:60 or 67:33). The chemical composition of these paper mill biosolids vary according to the processes used and the relative amounts of primary and secondary paper mill biosolids included (Vance, 2000). Combined paper mill biosolids usually contain 1.0-2.5% nitrogen by dry weight. The C:N and N:P ratios for these materials ranges from 100:1 to 20:1 and 4:1 to 8:1, respectively (Curnoe, 1998). Nitrogen fertilizer is usually added to combined paper mill biosolids at the mill, before its delivery to farm sites and has elevated amounts of NH_4-N and NO_3-N contents (Bellamy *et al.*, 1995).

Chemical composition of sludge

About 150 chemicals can be detected in de-inked paper mill biosolids (Beauchamp *et al.*, 2002). In general the C, N, P and K contents of de-inking paper mill biosolids are similar to those of primary paper mill biosolids. The contents of arsenic, boron, cadmium, cobalt, chromium, manganese, mercury, molybdenum, nickel, lead, selenium, and zinc are also low and showed low variability. However, the copper contents were above the Canadian compost regulation for unrestricted use and required a follow-up. The fatty- and resin acids and polycyclic aromatic hydrocarbons were the organic chemicals measured at the highest concentrations. They further concluded that raw de-inking paper mill biosolids and its young compost do not represent a major threat for the environment but can require an environmental follow-up. Heavy metals in the sludge are known to pose potential health risks to plants and animals if present in too high concentrations. They are strongly retained by soils and therefore can persist for long periods in the environment. Heavy metals are present as contaminants in pulp mill sludge either as a result of chemicals added during the pulping process or they originate in the wood itself, having been adsorbed from soil by trees.

Land application

Early research on paper sludge utilization in agriculture (Dolar *et al.*, 1972; Aspitarte *et al.*, 1973; Hermann, 1982; NCASI, 1984; Thiel, 1984) have shown numerous benefits like increased plant growth and yield, improved soil moisture and nutrient retention, and increased CEC etc. Adverse effects have been limited to high salt levels (ammonium, sodium and sulfate) and deficiencies of available N in some types of sludge. Effects of high salts and N deficiency have been minimized by controlling sludge application rates and by delaying planting for some period after treatment, and/or by applying fertilizer to soil or sludge (Simpson et., 1983). The benefits to land application of pulp mill biosolids are many and have recently been demonstrated in numerous studies (Bellamy *et al.*, 1995; Phillips *et al.*, 1997; Atiken *et al.*, 1998; Arfoui *et al.*, 2001 Simard, 1998, 2001; N'Dayegamiye *et al.*, 2003; Gagnon and Ziadi, 2004). Paper mill biosolids are somewhat limited by the high C:N ratio in primary sludge. Mixes or composts of primary and secondary sludge to create a C:N of 20:1 create a high quality soil amendment. This material provides the soil with a slow release of minerals, a source of replenishment of base cations, and a source of C for microorganisms in soils that may be starved for C following years of cultivation. The increased C content stimulates microbial activity and results in an increased humus content which relates directly to increased water holding capacity, increased CEC, increased metal sorption, and increased aggregation of soils. Lastly an important benefit of this type of application is diversion of a useable organic material from the waste stream into a beneficial use.

Decomposition in soil

Paper mill biosolids are mostly (about 90% of dry matter) composed of short wood fibers, and thus contains cellulose, hemicellulose and lignin, as major components. There is a general agreement that the initial phase of fresh organic matter decomposition in soil is rapid and mostly determined by free cellulose (holocellulose) disappearance, whereas the second phase of slow decomposition is driven by the degradation of cellulose encrusted with lignin (ligno-cellulose) (Berg *et al.*, 1984; Melillo *et al.*, 1989). The hemicellulosic fraction of plant residues is much smaller than the cellulosic one, and is often included with cellulose in the holocellulose fraction (Ryan *et al.*, 1990). The proportion of remaining material accounted for by lignin like compounds increases with time during organic matter humification Melillo *et al.*, 1989). Herman *et al.* (1977) also suggested that the rate of lignin loss during decomposition of various organic residues is directly linked to the initial lignin content of the residues. Lignin is considered as the most recalcitrant fraction of plant tissues (Aber *et al.*, 1990; Ryan *et al.*, 1990) and as a precursor of soil humic substances. Complex organic materials such as crop residues (Voroney *et al.*, 1989) and forest litters (Melillo *et al.*, 1989; Aber *et al.*, 1990) often decompose according to a two-compartment decay model.

Addition of a high C:N ratio material to soil, in general, will limit decomposition due to N limitation and N availability can be a limiting factor for soil microorganisms responsible for decomposition of organic materials (Cheshire and Chapman, 1996; Mary *et al.*, 1996). When organic materials having a wide C:N ratio undergo microbial decomposition, the microorganisms can become N limited (Kay and Hart, 1997). Paper mill biosolids appear to display decomposition characteristics similar to those of other organic materials commonly added to the soil (Chantigny *et al.*, 2000a, b). Application of primary paper mill biosolids caused significant reduction in extractable N in laboratory incubations and the decomposition was slow (Zibliske, 1987). However, after a time, enough C was apparently mineralized during the incubation to allow the onset of N mineralization. Zibliske (1997) reported that decomposition of combined biosolids (0.8% N) and secondary paper mill biosolids (3.3 % N) depended partly on the quality of carbon substrates contained in the waste material and partly on the availability of N. Nitrogen immobilization was more pronounced in combined biosolids treated soil compared to secondary biosolids amended soil and lasted for 82 days, followed by a high rate of N re-mineralization.

Fierro *et al.* (1999) reported that paper mill biosolids presented a particularly large recalcitrant pool decomposing slowly. Fifty-one percent of the initial material decomposed with a half life of 0.4 year, whereas the remaining material had a much slower rate of decay with a half life of 13 year. The large size and slow decomposition rate of the recalcitrant pool of this material were attributed to the high lignin content and the presence of clay in the sludge. After 27 months in the soil, the remaining mass of sludge was still 43% of its initial amount. The pattern of paper mill biosolids decomposition in soil follow two-compartment decay model and was fairly comparable to crop residues with an initial phase of rapid C losses followed by a second phase of slow C losses (Chantigny *et al.*, 2000a,b; Fierro *et al.*, 1999). The initial phase of decomposition was mostly explained by the decomposition of holocellulose and lignocellulose. One-third of de-inked paper mill biosolids-C was decomposing at a rapid rate (Chantigny *et al.*, 1999) whereas the major part was decomposing more slowly. The second slow-decay phase was mostly driven by lignocellulose decomposition, which was expected to have a mean residence time of at least 8.5 years. After 2 years, 40% of initially added sludge remained in soil, mainly as acid-resistant C, whereas holocellulose had almost completely disappeared. In both soils, decomposition rates of de-inked paper mill biosolids were greater when adding 50 than 100 Mg ha^{-1} likely due to a shortage of soil nutrients.

their chemical composition. Biosolids produced by primary wastewater treatment are composed mainly of wood fibre (cellulose, hemicellulose and lignin), having high C:N ratios (>100), and low N content (0.15-0.73). Secondary biosolids has higher N and P contents and a much lower C:N ratio than primary sludge, due to N and P addition to the waste treatment system to enhance biological degradation of C (Camberato *et al.*, 1997). Land application of primary paper mill biosolids with high C:N ratios can result in soil N immobilization (Dolar *et al.*, 1972; Shimek *et al.*, 1988; Simard *et al.*, 1998). The relatively low C:N ratio of secondary paper mill biosolids or combined indicates a good potential in its raw state for land use as a N source (Simard *et al.*, 2001). The crop response to land-applied paper mill biosolids is discussed separately as it depends on the biosolids N concentration, C:N ratio, heavy metals and amount applied.

Primary paper mill biosolids (PPMB)

Primary paper mill biosolids are considered more as a soil conditioner. Their very high C:N ratio (Table 1) can cause soil N immobilization. Several studies conducted in 80's have shown that applying PPMB with a high C:N ratio to the agricultural land will cause a net immobilization of N (King, 1984; Watson and Hoitink, 1985: Honeycutt *et al.*, 1988). Vasconcelos and Labral (1993) found that yield of *Lupin lateus* were

Table 1. Chemical analysis of primary and de-inking paper mill biosolids from different sources

Analysis	Goss *et al.*, 2003 (n=3)	Atiken *et al.*, 1998 (n =1)	Simard *et al.*, 1998 (n =3)
EC (mS cm^{-1})	0.39 - 0.87		0.09 - 0.20
pH	7.7 - 8.2	7.7	7.8 - 9.1
Dry matter (% dry)	39.7 - 49.0	31.5	
Organic Carbon (% dry)	29.7 - 33.4	31.8	42.3-44.2
Total N (% dry)	0.27 - 0.73	0.37	0.15
C:N ratio	42 - 124	86	284 - 292
Total P (%)	0.072 - 0.093	0.08	0.0096 - 0. 0097
Total K (%)	0 - 0.09	0.20	0.0029 - 0.0034
Total Ca (%)	2.86 - 5.85		0.46 - 0.56
Total Mg (%)	0 - 0.06	0.235	0.026 - 0.031
Total Na (%)	0.05 - 0.06		0.073 - 0.084

decreases with the application of paper mill biosolids application rate over 5 Mg ha^{-1}. However, 10 months after incorporation this effect was dissipated, suggesting a biodegradation of the residues.

Effect on crop growth

The effect of application of different paper mill biosolids will be different on crops, soil properties and the environment depending upon

Table 2. Chemical analysis of combined paper mill biosolids from different sources

Analysis	Simard, 2001 (n=2)	N'Dayegamiye *et al.,* 2003 (n=3)	Gagnon and Ziadi, 2004 (n=8)
pH	7.9-8.0	6.4-6.8	5.2-8.4
Dry matter (% dry)	28-32	32.4-28.5	21-41
Organic Carbon (g kg^{-1} dry)	347-496	384-397	350-530
Total N (g kg^{-1} dry)	11.8-12.4	25.2-29.8	9-36
C:N ratio	28-42	13-15	11-46
Total P (g kg^{-1} dry)	0.5-0.8 (PO_4-P)	3.01-4.0	1.4-7.8
Total K (g kg^{-1} dry)	1.8-4.6	1.01-2.1	0.5-4.6
Total Ca (g kg^{-1} dry)	2.1-2.6	6.1-48.5	1.5-29
Total Mg (g kg^{-1} dry)	0.6-2.2	0.8-0.9	0.4-2.2

Table 3. Analysis of different carbon fractions in paper mill sludge from different sources

Analysis	De-inking PMB (n=3)	Combined PMB (n=3)
Total N (g kg^{-1} dry)	3.0-3.6	25.2-29.8
Organic Carbon (g kg^{-1} dry)	381-383	384-397
C:N ratio	106-127	13-15
Cellulose (g kg^{-1} dry)	460.6-488.2	241-280
Hemicellulose (g kg^{-1} dry)	107.0-143.8	98-134
Lignin (g kg^{-1} dry)	256.5-290.5	276-355.4

O'Brien *et al.* (2003) applied PPMB at different rates (0, 112, 224, 336, or 448 Mg wet mass ha^{-1}) as one-time application of sludge (40% solids) was incorporated approximately 15-cm deep immediately into the soil. In each year, N was added to the field as ammonium nitrate at 200 or 400 kg N ha^{-1}. Corn (*Zea mays* 'Pioneer 35N05' was planted 17 days after the sludge was applied. Corn was again plant in the second year in the same plots. In the first season, plant density, determined at harvest, was suppressed in plots with paper sludge. In second, the year-previous additions of paper sludge had no effect on plant density. Final dry mass in first year suggested that plants recovered with time from the initially inhibiting effects of paper sludge addition. Corn ear dry mass did not differ between first and second year. They concluded that after one year of stabilization of paper sludge in the soil, perhaps more nutrients were available. Paper mill sludge application to land may benefit soil by adding nutrients, which may become available with time. The total average yield of Italian ryegrass (5 cuts in three years) was significantly reduced due the application of PPMB at 385 Mg ha^{-1} as one time application compared to control. The application of 50 kg N ha^{-1} improved the gross dry matter yield but it was still lower compared to the yield harvested from control plots (Douglas *et al.* (2003). The scale of the difference in grass yield between treatments indicated that differences in N supply were probably the main factor affecting grass production. Crop N concentrations were the lowest where PPMB was applied.

De-inking paper mill biosolids (DPMB)

A potential limitation for the use of raw de-inking sludge as a soil amendment is the possibility of N and P deficiencies for adequate plant nutrition and growth. However, when N and P fertilizers are supplemented, de-inking paper mill biosolids can be an adequate soil amendment in turf grass culture (Norrie and Gosselin, 1996). Fierro *et al.* (1997) evaluated de-inking paper mill biosolids as a soil amendment supplemented with N and P in a greenhouse study for the growth of the grasses *Agropyron elongatum* (tall wheatgrass), *Alopecurus pratensis*. (meadow foxtail), *Festuca ovina* var. *duriuscula* (hard fescue), legumes like *Galega orientalis* (galega), *Medicago lupulina* (black medic), and *Melilotus officinalis* (yellow sweet clover). Their results show that best response in sand-sludge mixtures was obtained with the mixture containing 30% biosolids (all species combined). De-inking paper mill biosolids are suitable for use as a soil amendment in revegetation studies on degraded light soils. Nitrogen and P supplements are, however, required to maintain acceptable plant growth and nutrition.

In another study Fierro *et al.* (1999) applied DPMB at 0 and 105 Mg ha^{-1} supplemented with three N rates (3, 6 and 9 kg Mg^{-1} DPMB) and 2 P rates (0.5 and 1 kg P Mg^{-1} dry DPMB) to grow tall wheat gross (*Agropyron elongatum*). They found that the biomass of the grass was increased with the application of DMPB in both growing seasons. Nitrogen and P application further increased the biomass production. Simard *et al.* (1998a) applied de-inked paper mill biosolids at 0, 5.5, 11 and 16 Mg ha^{-1} (C:N = 288) in spring along with the application of mineral N at 0, 45, 90 and 135 kg N ha^{-1} and grew barley on the amended plots. Barley yields were reduced by 50% under the high rate of de-inked paper mill biosolids application. In another study they tested the effect of de-inked paper mill biosolids on the growth of winter cabbage (Simard *et al.*, 1998b). The C:N ratio of this material was low (169) compared to that used in the previous study, and was supplemented with 3 kg N Mg^{-1} dry DPMB. De-inked paper mill biosolids were applied at 0, 7, 14 and 21 Mg ha^{-1} (C:N = 169) alone or with 0 to 235 kg N ha^{-1}. De-inked paper mill biosolids alone produced higher marketable cabbage yields than a compost of de-inked paper mill biosolids compost and dairy manure, but the yields were 20% lower than the combined paper mill biosolids. Addition of more mineral N to DMPB did not improve the yield. Soil inorganic N content 5 week after planting and at harvest was not significantly different from the control plots, suggesting that the mineral N supplement of 3 kg N Mg^{-1} DPMB prior to application was sufficient to counteract the immobilization process.

Despite its poor nutrient content, DPMB can be efficiently used in legumes that rely mainly on biological N_2 fixation to meet their N requirements. Voroney and Opstal (2001) have shown that the application of DPMB significantly increased soybean yields with the increasing rates of spring applied DPMB. Goss *et al.*, (2003) reported that soybean grain yields obtained from plots receiving no DPMB and DPMB at 30 Mg ha^{-1} were not significantly different ($P<0.05$) from each other. However, this non-significant difference might be due to high variability (CV = 16.64 %) in grain yields recorded from both treated and untreated plots. Allahdadi *et al.* (2004) applied DPMB to four forage legumes (sweet clover, red clover, alfalfa and birdsfoot trefoil) and measured the crop yields and N_2 fixation. The application of DPMB generally led to similar or greater productivity and greater N_2 fixation compared with unamended controls in the first production year. They recommended the application of DPMB in fall before legume establishment in next spring.

It may also be applied well in advance of crop planting to avoid early N immobilization (Camberato *et al.*, 1997). Other alternative would be the addition of N from mineral fertilizers (Voroney and Opstal, 2001) or animal manures (Gagnon *et al.*, 2004). Simultaneous addition of farm manure and paper sludge to soil was shown to improve crop yield and soil characteristics (Carneiro and Dos Santos, 1996), and decrease N leaching likely due to the temporary immobilization of part of the available N (Cabrita *et al.*, 1996). A combination of manure and paper sludge could also fix some of the P supplied by manure, making land disposal of these organic materials a more environmentally suitable practice. Gagnon *et al.* (2004) investigated the effect of combined application of DPMB and poultry manure on corn. They applied DPMB at three different rates, control, constant rate (based on soil organic C) and variable rates (10, 20 and 30 Mg ha^{-1}). Despite early N immobilization, grain yield was not affected whereas plant P uptake was increased by poultry manure. The variable application rate treatment did not differ from the equivalent constant rate treatment for crop growth. They concluded that mixing DPMB and poultry manure might provide a cost-effective and environmentally friendly approach to land disposal of these wastes.

We applied DPMB at different rates for three years at Simcoe Research Station and grew corn on amended plots. Nitrogen fertilizer was applied at 0, 50, 100, 200 and 300 kg N ha^{-1} as UAN (28% N) to all plots for three years. Corn grain yields were significantly affected by the application of DPMB as well as N fertilizer application during all three years. Interaction between DPMB and N fertilizer application rates did not significantly affect the corn grain yields. Maximum corn grain yields (9.10 Mg ha^{-1}) in 2002 were recorded from plots received 30 Mg DPMB ha^{-1} at 200 kg N ha^{-1}. Corn grain yields in 2003 (7.58 Mg ha^{-1}) were maximum at 200 kg N ha^{-1} in plots those received DPMB at 120 Mg ha^{-1} in 2002. Corn grain yields in 2004 were maximum (7.78 Mg ha^{-1}) in plots received DPMB at 120 Mg ha^{-1} in 2002, 0 in 2003 and 15 Mg ha^{-1} in 2004 at the same rate of N application rate. Mean average corn grain yields at different PMB and N application rates show that corn grain yields were maintained on DPMB amended plots as additional amount of N was applied as fertilizer. Corn grain yields were decreased wherever DPMB was applied in the third year at > 15 Mg ha^{-1} and N available in soil might have not been enough to meet the PMB decomposition and crop demands due to immobilization (Goss and Rashid, 2004).

Combined (primary/secondary) paper mill biosolids (PSPMB)

Paper mill biosolids produced by primary wastewater treatment is composed mainly of wood fiber (cellulose, hemicellulose and lignin), has high C:N ratio and has very low total N contents. On the other hand secondary paper mill biosolids has higher N and P contents and a much lower C:N ratio than primary paper mill biosolids, because N and P are commonly added to the waste treatment system to enhance biological degradation of C (Camberato *et al.*, 1997). Land application of PPMB with high C:N ratios can result in soil N immobilization (Dolar *et al.*, 1972; Simard *et al.*, 1998a). Research on combined primary and secondary de-inking paper mill biosolids indicated that they act as a source of N and P for crops with N mineralization rates from 10 to 50% (Simard *et al.*, 1998b). The relatively low C:N ratio of secondary or combined primary/secondary paper mill biosolids indicates a good potential in its raw state for land use as a N source (Simard *et al.*, 1998b).

Crop response to land-applied PSPMB has been variable and depended on the sludge N concentration, C:N ratio and amount of PSPMB applied (Camberato *et al.*, 1997). The potential available N content of the combined primary and secondary sludge has to be quantified to avoid excessive application, which could result in crop yield loss and NO_3-N leaching. Bellamy *et al.*, (1995) conducted a series of experiment to assess the response of grain corn to PSPMB (90% primary and 10% secondary; low N content) plus mineral fertilizer application. Mixed paper mill biosolids were applied at 0, 12 and 24 dry Mg ha^{-1} followed by applications of 0, 100 and 200 kg N ha^{-1} as ammonium nitrate. The application of PSPMB at 12 Mg ha^{-1} in combination with N at 100 kg ha^{-1} produced highest corn grain yields.

Simard (2001) conducted a 3-yr experiment to determine the effects of PSPMB application on crop yields, N uptake and N recovery. The PSPMB was applied in 1996 on a Bedford silty clay (Humic Gleysol) cropped to winter cabbage (*Brassica oleracea* var *capitata* L. 'Bartolo') at 0, 8, 16, 32, and 64 Mg ha^{-1} (dry basis). In 1997, PSPMB was applied at 44% of the 1996 rates to the same plots and cropped to sweet corn (*Zea mays* L. 'Delectable'). No PSPMB was applied in 1998 to evaluate residual effects on corn. Treatments with ammonium nitrate at 50, 100 and 200% of N fertilizer recommendations were also included each year as a reference for crop response. The PS had a C:N ratio of 42:1 in 1996 and of 28:1 in 1997. Cabbage and corn marketable yields and N uptake increased with increasing amounts of PSPMB applied. PSPMB supplemented with ammonium nitrate (AN) further increased cabbage yields. The two PSPMB applications also had a very significant residual effect on corn yield in the third year, although supplemental AN at 150 kg N ha^{-1} tended to further increase yields. The apparent total N recovery by the three crops was similar for both PSPMB and AN (i.e., 34 vs. 38%).

Arfaoui *et al.* (2001) compared three PSPMB (100, 200 and 400 kg N ha^{-1}) with calcium ammonium nitrate (CAN) at 0, 50, 100 and 200 kg N ha^{-1}) to grow mixed grass alfalfa (*Medicago sativa* L.) sward in 1997 and 1998 on a Bedford clay loam (Humic Gleysol). The PSPMB and CAN induced a significant linear increase in forage dry matter yield and relative yield. Nitrogen concentration in forage tissues was increased by PSPMB and CAN inputs in 1997, but was decreased in 1998. They concluded that PSPMB can be an efficient N source for grass-alfalfa swards on fine-textured soils.

N'Dayegamiye *et al.* (2003) applied PSPMB (30, 60 and 90 Mg ha^{-1}) alone and in combination with N fertilizer (90 and 135 kg N ha^{-1}, respectively, for 60 and 30 Mg ha^{-1}) to corn. Treatments also included a control without PSPMB or N fertilizer, and a complete mineral N fertilizer (180 kg N ha^{-1}) as recommended for corn. Corn yields were significantly increased by 1.5–5 Mg ha^{-1} when PSPMB was applied alone compared to the unfertilized control. However, corn yields and N uptakes were highest from the application of PSPMB in combination with N fertilizer. Biennial applications at 60 to 90 ha^{-1} significantly increased corn yields and N uptake, which suggest high

PSPMB residual effect; however, these increases were lower than those obtained with annual PSPMB applications. The results indicate high N supplying capacity and high residual N effects of PSPMB, which probably influenced corn yields and N nutrition.

Gagnon *et al.* (2003) conducted a field study to compare the effect of PSPMB with mineral fertilizers on the blueberry yield and soil chemical properties and enzyme activities of a sandy soil in the Saguenay-Lac Saint-Jean area (Quebec, Canada). The PSPMB was applied in the spring at 0, 8.5, 17 and 34 Mg ha^{-1} and mineral fetilizer was applied at 0, 13, 26 and 52 kg N ha^{-1}. The highest fresh fruit yields were obtained at 8.5 and 17 Mg PSPMB ha^{-1}. The 34 Mg PSPMB ha^{-1} treatment produced berry yield comparable to the control. They suggested that PSPMB, when used at low rates, improved low bush blueberry yield on this less fertile sandy soil.

Gagnon *et al.* (2004) in their review paper concluded that PSPMB when applied at appropriate rates (10 Mg ha^{-1}) increased plant growth by supplying nutrients. Crops with long cropping seasons and high nutrient requirements may benefit more from PSPMB applications. They further suggested that soils with low organic C have a greater potential to respond to PSPMB additions.

Composted paper mill biosolids (CPMB)

Compost is the product of degradation of fresh organic matters by microorganisms. Field scale studies have demonstrated the feasability of pulp and paper industry biosolids for composting (Campbell *et al.*, 1995; Line, 1995; Rantala *et al.*, 1999). Composting could potentially reduce the C:N ratio, volume, and odour of paper mill biosolids and potential soil N immobilization in paper mill biosolid-amended soils, thus producing a marketable material suitable for horticulture and agricultural uses. In addition, composting stabilizes the organic C, so compost may have a longer impact on soil organic matter content than raw materials on coarse-textured soils (Barker 1997). Paper mill biosolids alone can not easily be composted successfully due to high C:N ratio in primary or de-inked paper mill biosolids.

Addition of materials with high N contents enhances the microbial breakdown of biosolids without harmful consequences for N uptake by plants (Zhang *et al.*, 1993; Feagley *et al.*, 1994, Bellamy *et al.*, 1995; Norrie and Gosselin, 1996). Nitrogen reinforcement can be achieved by the addition of materials containing high N such as cattle, poultry and swine manure and swage sludge (Arrouge *et al.*, 1998; Sesay *et al.*, 1997). Co-composting is a good method to produce a stabilized material with a more balanced nutrient composition than raw paper mill biosolids. The co-composting of the paper mill biosolids and hardwood sawdust can also be successfully done if aeration, moisture, and bio-available C:N ratios are optimized to reduce losses of N (Dinel *et al.*, 2004).

Simard *et al.* (1998) applied paper mill biosolids composted with dairy manure at 0, 10 and 15 Mg dry ha^{-1} before winter cabbage (*Brassica oleracea var. capitata* L.) planting. The cabbage yield was significantly lower compared to mineral N fertilizer (185-235 kg N ha^{-1}) and raw CPMB amended with 3 kg N Mg^{-1} dry CPMB. They observed significant N immobilization early in the season where composted paper mill biosolids were applied, however, no significant difference in soil inorganic N between treatments.

Rantala *et al.* (1999) applied five CPMB to barley at two different rates based on the amount of P and K supplemented with N (90 kg ha^{-1}). Barley yields with the application of CPMB were same as produced by NPK fertilization in the first year. They also observed a net N immobilization in soil during the growing season and concluded that the composts might be immature to some extent for crop production.

Brito (2001) investigated the crop responses to soil amended with municipal solid waste compost and CPMB with bark in pot experiments using lettuce (*Lactuca sativa* L) cvs. Animo and Jory and summer cabbage (*Brassica oleracea* L. var. *capitata* L) cv. Lima. Dry matter accumulation generally increased with increasing concentrations of CPMB. Lettuce dry matter accumulation further increased when composted paper mill sludge treatments were amended with N fertilizer (0.1 and 0.2 g N kg^{-1}) but declined with further amounts. Dry matter accumulation of lettuce and N accumulation of cabbage could be described as a function of compost nitrogen content and electrical conductivity.

Lalande *et al.* (2003) applied CPMB to a Bevin loamy sand soil (Orthic Humo-Ferric Podzol) to determine the effects of a co-composted paper mill biosolids with hog manure, applied alone or in combination with mineral fertilizers for potato and wheat yields. The CPMB was applied in the spring of 1997 at rates of 0, 11.5, 23 and 34.5 Mg dry ha^{-1}, with and without fertilizer equivalent to 150 N-200 P_2O_5-200 K_2O kg ha^{-1}. Potatoes (*Solanum tuberosum* L.) were planted the first year and the residual effect of CMPB was evaluated on a spring wheat (*Triticum aestivum* L.) crop in 1998. The addition of CPMB at 11.5 Mg ha^{-1} produced the highest marketable potato tuber yield with fertilizer supplement. Highest wheat grain yield was obtained with the 23 Mg CPMB ha^{-1} level in the following season.

Sippola *et al.* (2003) applied five paper mill biosolids composted with bark (different ratios) to barley at two different rates based on the amount of P. Lower rate of CPMB was based on an annual P fertilization rate and the high rate was double or triple the low rate depending on the mineral N concentration of compost. Total contents of N, P, K and Ca in composts were 8.8-17.5, 0-7-3.9, 1.5-6.5 and 4-25 g kg^{-1} dry matter, respectively. There was no significant difference in grain yields between plots received mineral fertilization and compost treatments supplemented with mineral fertilizers. However, there was a decreased fertilization effect of some of the composts on straw yield during first experimental year indicating N immobilization. Composted paper mill biosolids has a definitive nutrient value for crops provided the application rate is appropriate and supplementation with mineral N fertilizer is used when C:N ratio is not optimal. Composting with manure may be considered, however, CPMB provides no advantage over the raw material apart from possibly reducing soil immobilization (Gagnon, *et al.*, 2001; Nemati *et al.*, 2000).

Effect on soil and environment

Structural stability is a very important soil physical property that describes the ability of the soil to retain its arrangement of solids and voids when exposed to different stresses (Kay, 1990). The maintenance of adequate levels of organic matter in soils has been identified as a key to productive, sustainable agricultural systems (Doran and Scot-Smith, 1987). The soils under intensive continuous cultivation are in a net deficit of C and require external sources of organic matter to equilibrate their deficient C balance. Carbon sequestration can be enhanced by the application of organic materials to soil (Janzen *et al.*, 1998). Addition of organic amendments is an effective way to restore soil physical and biochemical properties, and consequently improve crop productivity (Lalande *et al.*, 1998; Gagnon *et al.*, 2001). With the aim of increasing the organic matter level in agricultural land, there has been a growing interest in the advantages of using organic residues and industrial waste rich in C, such as paper mill biosolids may provide an interesting solution to soil degradation problems (Trépanier *et al.*, 1998).

Heavy Metals

One of the major public concerns over the use of paper mill sludge on agricultural land is the potential for heavy metal contamination of water and plants. Contents of heavy metals and organic toxic compounds in paper mill sludges are generally low (Trépanier *et al.*, 1996; Cabral *et al.*, 1998; Demeyer and Verloo, 1999) and comparable to those found in livestock manure (Bellamy *et al.*, 1995). Concentrations of heavy metals in soil amended with paper mill biosolids or plants grown in these soils have usually been below established standards (Simard *et al.*, 1998; Baziramakenga and Simard 2001). However, our results shows that copper levels in soil (estimated after crop harvest) after the application of de-inking paper mill biosolids at 135 Mg ha^{-1} exceeded the permissible limits (Goss and Rashid, 2004). These results suggest that de-inking paper mill biosolids should not be applied in heavy quantities as a single dose.

NO_3-N leaching

Agricultural soil is one of the main contributors of nitrate nitrogen (NO_3-N) to natural waters. An important task is therefore to find realistic means of reducing NO_3-N leaching from agricultural systems. Reduction of NO_3-N leaching through modification of soil biological processes is one possible counter measure (Kirchmann *et al.*, 2002). One manageable part of the NO_3-N leaching problem is to control nitrification. This can be achieved by decreasing net mineralization or increase net N immobilization in soil. This is possible through the application of organic materials rich in microbial available energy and with low N content. Primary and de-inked paper mill biosolids have low N contents and contain a large proportion of easily degradable fibers that may facilitate net immobilization if applied on soils. The possibility of using paper-mill wastes for reducing N leaching has been recognized (Zibilske, 1987; Feagley *et al.*, 1994; Bellamy *et al.*, 1995; Harrison *et al.*, 1996; Busscher *et al.*, 1999).

Table 4. Heavy metal concentrations in paper mill sludge from different paper mills

Metal	Abitibi† sludge (n=3)	Calger‡ sludge (n=3)	Powell‡ River primary (n=3)	Powell‡ River secondary (n=3)	Paprican‡ primary (n = 4)	Paprican‡ secondary (n=3)	CMCS limit¶
	Concentration of metals in sludge sample (mg kg^{-1})						
Arsenic	<1.0		0.09-1.7	0.2-0.98	0.5-1.1	0.9-1.2	20
Cadmium	<1.0		0.25-2.5	0.25-2.5	0.4-5.0	1-14	3
Chromium	5.2-12		17-29	31-46	8-73	20-40	750
Cobalt	<1.5-2.5		1-5	<1	<1-4	<1-3	40
Copper	250-310		15-26	12-30	11-46	14-65	150
Lead	8.3-10		1-10	4-10	3-92	5-37	375
Mercury	0.57-0.87		0.02-0.2	0.14-0.25	0.02-0.10	0.1-1	0.8
Molybdenum	2.5-3.8		4-20	<4	<2	<2-10	5
Nickel	2.9-5.6		7-10	<2-9	8-56	9-38	150
Selenium	<1.0		<0.5-2.0	<0.5-2.0	0.05-2.7		
Zinc	130-250		26-38	30-79	30-94	88-475	600

Abitibi†: Goss *et al.* (2003)
Calger‡: What we know about paper mill biosolids. 1997. Reach for Unbleached! http://www.rfu.org/MonSolid.htm#Pulp%20Mill%20Sludge%20Land%20Applicatio
CMCS limits¶ = Criteria for Managing Contaminated Sites Limits in British Columbia

Vinten *et al.* (1998) applied paper mill biosolids at three different rates (0, 12.7 and 44.4 Mg ha^{-1}) to an intensive vegetable growing area in Scotland vulnerable to nitrate leaching (14 mg NO_3-N L^{-1}) and measured the NO_3-N leaching losses. Soil solution samples were collected from soil solution samplers installed in the field. Their results suggest that a significant reduction in NO_3-N leaching occurred as a result of the paper waste treatment. The application of paper mill biosolids at highest rate of application (44.4 Mg ha^{-1}) resulted in a decrease in N leaching from 177 to 94 and 227 to 152 kg ha^{-1} at two sites, respectively. Smallest NO_3-N leaching loss was observed where paper mill biosolids were mixed in soil by conventional plowing. Deep plowing significantly increased the NO_3-N leaching losses in both with and without paper mill biosolids application.

Baggs *et al.* (2002) observed a continuous decline in NO_3-N contents in soil throughout the experimental period. They estimated that application of PPMB in autumn reduced leaching between October 1994 and March 1995 by up to 90 kg N ha^{-1}. However, they suspected that much of the reduction in soil NO_3-N might be due to higher denitrification associated with the hot spots of microbial activity as a result of high input of organic carbon.

Kirchmann and Bergstrom (2003) tested four different types of paper mill wastes (secondary sludge, primary, de-inked fiber sludge and wood waste) for their suitability to control the NO_3-N leaching losses. Primary fiber sludge immobilized most N in soil as compared with deinked fiber sludge and wood waste, both on the basis of dry matter, C and added cellulose-C. Secondary sludge released N in soil in accordance with its high N content. With the aim of reducing NO_3-N leaching from agricultural soil in a cold temperate climate during autumn/winter/early spring through temporal N immobilization, only primary fiber sludge and de-inked fiber was found to be potentially suitable of the organic wastes tested.

Conclusions

The main impacts of paper mill biosolids application to agricultural soils can be summarized as follows:

It is useful soil amendment which increases soil organic matter content, improving water holding capacity, structure and bulk density of soil. Successive application of paper mill biosolids (primary and deinking) will be beneficial for poor textured and structured soils, such as sandy and heavy clayey soils to improve the physical conditions of these soils.

Crop yields will be affected depending on the type of paper mill biosolids applied and how it is applied such as co-application with some other materials. Primary paper mill biosolids and deinking paper mill biosolids will reduce the crop yields (except leguminous crops) due to N immobilization. However, secondary paper mill biosolids, combined (primary and secondary mixed) and composted paper mill biosolids will increase the crop yields due to higher N availability to crops.

Successive application of paper mill biosolids may contaminate the soil with heavy metals and other organic compounds. Ground water may also get contaminated (secondary and combined sludge applied). Nitrogen and P immobilization (primary and deinking paper mill biosolids) by microorganisms due to high C:N and C:P ratio of these solids, will ultimately result in lower crop yields.

Finally, successive land application of paper mill biosolids will always need a convenient monitoring in terms of amount used and methods and time of application to avoid soil and water pollution.

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