Pulp and paper mill by-products as soil amendments and plant nutrient sources

J. J. Camberato[1, 4], B. Gagnon[2], D. A. Angers[2], M. H. Chantigny[2], and W. L. Pan[3]

[1]*Pee Dee Research and Education Center, Entomology, Soils, and Plant Sciences, Clemson University, 2200 Pocket Road, Florence, SC 29506-9706, USA;* [2]*Soils and Crops Research and Development Centre, Agriculture and Agri-Food Canada, 2560 Hochelaga Blvd., Sainte-Foy, Québec, Canada G1V 2J3;* [3]*Department of Crop and Soil Sciences, Washington State University, Pullman, WA 99164-6420, USA.* *Technical Contribution No. 5126 of the Clemson University Experiment Station. Received 8 April 2005, accepted 11 April 2006.*

Camberato, J. J., Gagnon, B., Angers, D. A., Chantigny, M. H. and Pan, W. L. 2006. **Pulp and paper mill by-products as soil amendments and plant nutrient sources.** Can. J. Soil Sci. **86**: 641–653. Pulp and paper mill sludges are produced from primary and secondary treatment of wastes derived from virgin wood fiber sources, recycled paper products, and non-wood fibers. Sludges and sludge composts may be utilized in agriculture to increase soil organic matter, improve soil physical properties, provide nutrients, and increase soil pH. Positive effects of primary, deinking, and low-nutrient combined sludges on soil quality are primarily due to increased soil organic matter, aggregation, water holding capacity, infiltration rate, and cation exchange capacity. Nitrogen and P immobilization are often induced by primary and deinking sludges, but can be overcome by delayed planting, adding N and P, planting of legumes, or composting. Improved crop production obtained with secondary treatment sludges is most often attributable to enhanced nutrient availability, particularly N, but improved soil physical properties are implicated in some studies. Pulp and paper mill sludges and sludge composts are useful soil amendments and plant nutrient sources.

Key words: Paper mill sludge, soil physical properties, N and P immobilization, nutrient efficiency, land application

Camberato, J. J., Gagnon, B., Angers, D. A., Chantigny, M. H. et Pan, W. L. 2006. **Les sous-produits de papetières comme amendement pour les sols et sources d'éléments nutritifs pour les cultures**. Can. J. Soil Sci. **86**: 641–653. Les boues des usines de pâtes et papiers sont produites à partir du traitement primaire et secondaire des résidus dérivés des fibres de bois vierges, de pâtes recyclées et d'autres matériaux non ligneux. Les boues et composts de boues peuvent être utilisés en agriculture pour rehausser la matière organique du sol, améliorer les propriétés physiques, fournir des éléments nutritifs et augmenter le pH. Les effets positifs des boues primaires, des boues de désencrage et des boues mixtes à faible valeur fertilisante sur la qualité des sols sont essentiellement liés à une augmentation de la matière organique du sol, de l'agrégation, de la capacité de rétention en eau, du taux d'infiltration et de la capacité d'échange cationique. Une immobilisation de N et P est souvent rencontrée suite à l'utilisation de boues primaires et de désencrage, mais celle-ci peut être corrigée en retardant la mise en place des cultures, en ajoutant du N et du P, en plantant des légumineuses et en compostant les résidus. L'augmentation de productivité des cultures obtenue avec les boues issues du traitement secondaire est le plus souvent attribuée à la fourniture d'éléments fertilisants, particulièrement l'azote, mais une amélioration des propriétés physiques du sol est invoquée dans certaines études. Les boues et les composts de boues de papetières sont des amendements organiques intéressants pour les sols et une source importante d'éléments nutritifs pour les cultures.

Mots clés: Boues de papetières, propriétés physiques du sol, immobilisation de N et P, efficacité des éléments nutritifs, application au sol

Pulp and paper mills generate several types of sludges from primary and secondary treatment of wastes derived from virgin wood fiber sources, recycled paper products, and non-wood fibers (Table 1). Land application is becoming more frequently practiced as an outlet for sludge utilization. Surveys conducted by the National Council of the Paper Industry for Air and Stream Improvement (NCASI) indicated 3, 6, and 12% of the sludge generated by NCASI members (70% of the papermaking industry) were land applied in 1979, 1988, and 1995 (NCASI 1999). Fifty-one percent of sludge produced was landfilled in 1995 (NCASI 1999). Similar utilization occurred in Canada at this point in time (Reid 1998). Application of paper mill sludges to agricultural land has increased in recent years to 30% in the province of Québec, resulting in a comparable decrease in the amount landfilled (Fig. 1; Environnement Québec 2004a). The nutrient, heavy metal, and organic composition of paper mill sludges is determined by the fiber source and treatment process, and is quite varied (Camberato et al. 1997). Primary sludges are mainly composed of short lignin fibers, high in C but low in plant nutrients. Median N and P were reported to be 2.7 and 1.6 g kg^{-1} in a survey of 54

[4]*Current address: Department of Agronomy, Purdue University, West Lafayette, IN 47907-2054, USA (e-mail: jcambera@purdue.edu).*

Abbreviations: **CCE**, $CaCO_3$ equivalence; **CCME**, Canadian Council of Minister of the Environment; **CEC**, cation exchange capacity; **EQ**, Exceptional Quality; **MPN**, most probable number; **NCASI**, National Council of the Paper Industry for Air and Stream Improvement; **TAPPI**, Technical Association of the Pulp and Paper Industry; **TEQ**, toxic equivalent; **USEPA**, United States Environmental Protection Agency

Table 1. Estimates of proportion of papermill sludge production by type of treatment

Type	US – 1995[z] Sludge as produced	US – 1995[z] Sludge as managed	Canada – 1995[y]	Québec – 2002[x]
Primary	87%	40%	42%	5%
Deinking	NR[w]	NR[w]	12%	9%
Secondary	9%	5%	26%	1%
Combined	0%	54%	18%	85%

[z]NCASI (1999).
[y]Reid (1998).
[x]Environnement Québec (2004a).
[w]NR, not reported.

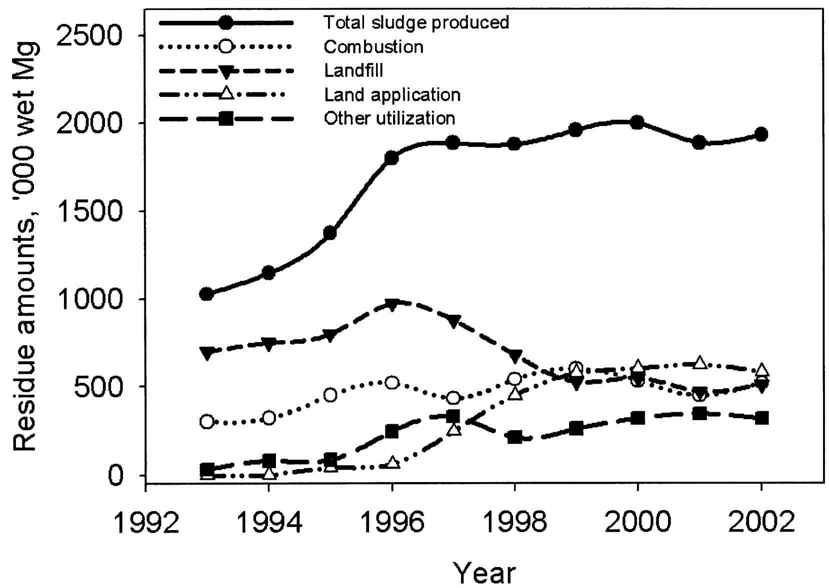

Fig. 1. Amounts of paper mill sludge generated in the province of Québec (1993–2002) and method of utilization or disposal. "Other Utilization" includes composting, application to forest and degraded sites, recycling, and reprocessing [adapted from Désilets (2003) and Environnement Québec (2003, 2004a)].

paper mills (Thacker and Vriesman 1984). Deinking sludge represents a low nutrient (~2.7 g N kg^{-1} and ~0.1 g P kg^{-1}) short fiber sludge containing some kaolin clay resulting from the recycling of used paper products (Trépanier et al. 1996b, 1998; Simard et al. 1998b; Chantigny et al. 1999; Beauchamp et al. 2002; Allahdadi et al. 2004). Additional treatment of primary and deinking sludge is often accomplished by the addition of N and P to facilitate microbial degradation, hence the term secondary sludge, and results in higher nutrient contents with median N and P levels of 23.3 and 4.2 g kg^{-1}, respectively (Thacker and Vriesman 1984). The relative proportion of primary or deinking and secondary sludge in combined sludges determines their nutrient content.

PULP AND PAPER MILL SLUDGE REGULATORY STANDARDS IN CANADA AND THE UNITED STATES

Regulations of the land application of pulp and paper sludges in Canada vary by province, but the metals criteria used in most provinces (with the notable exception of Ontario) are quite similar to those proposed in the Canadian Council of Ministers of the Environment (CCME) guideline (Table 2, Bureau de Normalisation du Québec 2005). Canadian criteria are generally more stringent for land application than those established at the national level by the United States Environmental Protection Agency (USEPA 1994). Paper mill sludges generally have metal concentrations less than municipal waste biosolids and well within regulatory limits (Camberato et al. 1997). Most paper mill sludges applied on land in the province of Quebec meet the Compost A CCME limit (Charbonneau et al. 2001).

In the province of Quebec, application of residues high in metals (listed Category C2, Table 2) is limited to <22 Mg dry ha^{-1} 5 yr^{-1} (Environnement Québec 2004b). In Nova Scotia, exceptional quality category, equivalent to Compost A, requires no approval for land application while several restrictions exist for land use of the other biosolids classes (Nova Scotia Environment and Labour 2004). In Alberta, along with metal A CCME criteria, pulp mill sludge must have a C:N ratio <75 and sodium adsorption ratio <50 (Alberta Environmental Protection 1999). Also in Alberta,

Table 2. Regulation of maximum metal concentration in biosolids utilized for land application in Canada and United States

Metal	CCME[z] Compost A	CCME[z] Compost B	Quebec Category C1	Quebec Category C2	Ontario	British Columbia Class A	British Columbia Class B	USEPA 503 EQ[y]	USEPA 503 Ceiling limit	Maine CH 419 Ceiling limit
	mg kg^{-1} (dry weight basis)									
Arsenic	13	75	13	40	170	13	75	41	75	41
Cadmium	3	20	3	10	34	3	20	39	85	39
Chromium	210	nd[x]	210	1060	2800	100	1060	1200	3000	3000
Cobalt	34	150	34	150	340	34	150	nd[x]	nd[x]	nd[x]
Copper	400	nd[x]	400	1000	1700	400	2200	1500	4300	1500
Lead	150	500	150	300	1100	150	500	300	840	300
Mercury	0.8	5	0.8	5	11	2	15	17	57	10
Molybdenum	5	20	5	20	94	5	20	[w]	75	75
Nickel	62	180	62	180	420	62	180	420	420	420
Selenium	2	14	2	14	34	2	14	36	100	100
Zinc	700	1850	700	1850	4200	500	1850	2800	7500	2800
Reference	Bureau de Normalisation du Québec (2005)		Environnement Québec (2004b)		MOE and OMAFRA (1996)	McDougall et al. (2002)		USEPA (1994)		Wright (2001)

[z] Canadian Council of Ministers of the Environment.
[y] Exceptional Quality standards.
[x] Not defined.
[w] Standard for Mo is under review.

the application rate to land is limited to <50 Mg ha^{-1} (dry weight) and the frequency of application is determined according to the rate used. The criteria for metals content in Ontario are much less stringent than in the other provinces (Table 2), but application rates of sludges with the highest allowable metal concentrations are limited to a greater extent to <8 Mg ha^{-1} (dry weight) over 5 yr (MOE and OMAFRA 1996).

Pulp and paper mill sludges are also classified according to their pathogen level (level 1: fecal coliform <1000 most probable number (MPN) g^{-1} total dry weight solids or *Salmonella* <3 MPN 4 g^{-1} total dry weight solids, level 2: fecal coliform <2 000 000 MPN g^{-1} total dry weight solids), which determines their use, separation distances and waiting periods. In addition, in Quebec, sludges are also subject to specific requirements based on odor that are more restrictive than those applied to manure (Environnement Québec 2004b).

Soil analysis is required prior to sludge application to ensure site suitability and to protect human health and the environment. The criteria examined vary with the province. In Quebec for example, only the Mehlich-III extractable P and Al and a calculation of the P saturation index are needed, in accordance with the "Regulation respecting agricultural operations" (Environnement Québec 2004b). Ontario, British Columbia, and Nova Scotia identify other standards such as pH range and maximum acceptable metal concentrations in soils (MOE and OMAFRA 1996; McDougall et al. 2002; Nova Scotia Environment and Labour 2004). In Alberta, optimum receiving soil properties include pH, sodium adsorption ratio, electrical conductivity, C/N ratio and texture, which determine the agronomic limitations for application rate (Alberta Environmental Protection 1999). For all provinces, application rate of biosolids takes into account major nutrients status of soils, composition of sludge and crops to avoid over-fertilizing and potential contamination of surface and ground waters.

In the United States of America, regulation of paper mill sludge land application is in many ways similar to that in Canada. Paper mill sludges are often regulated by application of the USEPA 503 standards for land application of municipal sewage sludge biosolids (USEPA 1994). Individual states can increase, but not diminish the stringency of requirements for land application (see metal concentrations for Maine CH 419 in Table 2 for example). Land application regulations take into consideration sludge characteristics, such as metal concentrations, pathogen level, and vector attractiveness. The distribution method, containerized (<1 Mg quantities) versus bulk, and the level of public access to the application site also affect land application restrictions. Other considerations include site and soil characteristics, distances from surface water and wells, depth to groundwater, potential for erosion and runoff, and proximity to floodplains or wetlands. Application rates may be limited by metal content (annual for containerized sludges or cumulative for bulk sludges), the N or P requirement of the crop to be grown, the lime requirement of the soil, or the potential for N or P pollution of ground and surface waters. Sludges that are defined "Exceptional Quality" (EQ), based on low levels of pollutants (Table 2), pathogens, and attractiveness to pathogens have lesser restrictions for land application.

Dioxin content of the sludge may also impact land application guidelines. For example the Maine Department of Environmental Protection prohibits the land application of sludges containing more than 250 pg g^{-1} dioxin toxic equivalents (TEQ) (Wright 2001). Land application of sludge with TEQ between 27 and 250 pg g^{-1} requires subsequent soil sampling to monitor accumulation of dioxin in the soil and restricted land use at soil TEQ >27 pg g^{-1}. Even though sludges with TEQ between 27 and 250 pg g^{-1} can be land applied in Maine, all meeting this criteria are landfilled (Wright 2001). Sludges containing <27 pg g^{-1} dioxin equivalents have no special restrictions. In Quebec, the dioxin and furan content should not exceed 17 and 100 μg g^{-1} in the Category C1 and C2, respectively (Environment Québec 2004b).

PULP AND PAPER MILL SLUDGES AS SOIL AMENDMENTS

Soil Organic Matter and Biological Processes

Paper mill sludge applications to soil may have profound effects on soil biological, physical, and chemical properties. Recent field experiments with extensive measurement of sludge effects on soil properties are summarized in Table 3. Increases in soil organic matter from paper mill sludge additions to soil are dependent on sludge composition and the rate, frequency, and total application. Wood fiber is approximately 15–35% hemicellulose, 40–45% cellulose, and 20–30% lignin. Sludges high in primary or deinking sludge reflect the composition of wood fiber. Thus C added to soil at high rates in the form of paper mill sludge could have persistent effects on soil properties. Decomposition of paper mill sludge occurs in a two-phase process with a rapid decomposition period of short duration preceding a slow decay over a long time. Fierro et al. (2000) found about half the sludge mass had a half-life of 0.4 yr, while the other half had a half-life of 13 yr. Similar estimates were obtained by Chantigny et al. (2000b), a rapid-decay phase with a mean residence time of 0.1–0.3 yr followed by a slow-decay phase with a mean residence time of 8.5 yr. Monitoring of soil carbohydrate composition revealed that cellulose degradation was mostly involved in the rapid decay phase, whereas lignin was responsible for the slow decay phase of deinking sludge (Chantigny et al. 2000b). More than 40% of the sludge remained in the soil 2 yr after incorporation (Chantigny et al. 1999; Fierro et al. 2000). Recalcitrance has been attributed to inclusion of clay in the sludge from the papermaking process (Fierro et al. 2000), and physical encrustation of the paper fibers by soil particles (Chantigny et al. 1999) as well as the biological recalcitrance of the C compounds themselves.

High sludge application rates have large and persistent effects on soil organic matter while low rates of application generally result in smaller increases that are mostly detectable in the year of application. Soil organic C of a fine sandy loam remained elevated 5 yr after a single application of a combined sludge [3:1 primary:secondary (vol/vol)] at 180 and 225 Mg ha^{-1}, but not at rates of 45, 90, or 135 Mg ha^{-1} (Zibilske et al. 2000). Annual or biennial applications at ≥ 45 Mg ha^{-1} increased soil C throughout the 5-yr study. The increases in soil C at the end of the study were directly proportional to the cumulative applied sludge C. For example, annual application of sludge at 225 Mg ha^{-1} more than doubled soil C from its initial level of 4%. Soil organic C content of a silty clay loam and a clay loam (0–15 cm depth) was doubled by application of a deinking sludge at 100 Mg ha^{-1} and the effect was still significant in the third year after application (Chantigny et al. 1999). In comparison, application of a deinking sludge at 6, 12, and 18 Mg ha^{-1} yr^{-1} increased soil organic matter content by only 10 g kg^{-1} in the 2 yr of application, but not by the third year when no application was made (Trépanier et al. 1996a). Similarly, soil organic C of a silt loam was increased slightly by application of a deinking sludge at 16 Mg ha^{-1} in the season of application, but not in the second season (Simard et al. 1998b). A study under progress in southern Quebec indicates that annual applications of 8 Mg ha^{-1} of combined sludge to a loam increased the soil C of the 0–10 cm surface layer by 4.1 g kg^{-1} after 3 yr of application, but had no effect in the 10–20 cm layer (N. Ziadi, personal communication). Based on the research cited above, it appears that sludge applications >20 Mg ha^{-1} yr^{-1} are necessary to substantially increase soil C in the short term in the climate of southern Canada and the northern US.

Microbial activity is stimulated by addition of paper mill sludge. Microbial biomass, CO_2 evolution, and the activity of several enzymes (fluorescein diacetate, acid phosphatase, arylsulfatase, and urease) were increased 11 mo after application of raw sludge with a C:N ratio of 109:1 to a Fredericton sandy loam (Gagnon et al. 2001). Soil amended with deinking sludge at 50 Mg ha^{-1}, compared with an unamended control, had increased microbial biomass C, hydrolysis of fluorescein diacetate, and alkaline phosphatase activity (Chantigny et al. 2000a). Application of a paper mill/swine manure compost at 11.5 Mg ha^{-1} to a sandy soil increased microbial biomass C and the activity of several enzymes by 55% and 30% (Lalande et al. 2003).

Few studies have examined the impacts of paper mill sludge spreading on macrobiota, and those conducted have been on forested or reclamation sites, not agricultural land. The application of paper mill sludges has been shown to have little effect on macrobiota in these ecosystems. For instance, no effect of paper mill sludge application was detected on deer mouse populations or several bird species in a Wisconsin red pine plantation (Thiel et al. 1988, 1989). Similarly, paper mill sludge spread to regenerate a spruce-fir forest in Maine had no effect on breeding bird density, small mammal numbers, or invertebrate abundance (Vera and Servello 1994). The number of omnivorous ground-gleaning birds was increased at the expense of the number of insectivorous ground-gleaning birds due to an increase in shrubby vegetation with sludge application in this study. Paper mill sludge applied to reclaim a coal mine in Ohio had little impact on fish, frogs, algae, or vegetation in a drainage lake, nor did dioxin or furan accumulate in rodents, insects, earthworms, or plants (McFadden et al. 1995).

Soil Bulk Density and Aggregation

Improved soil physical properties, such as lower bulk density and greater soil aggregation, reflect beneficial increases in soil organic matter and microbial activity from paper mill sludge applications. Bulk density of a sandy minesoil decreased from 1.7 to 1.3 Mg m^{-3} through 2 yr after application of deinking sludge at 105 Mg ha^{-1} (Fierro et al. 1999). Primary sludge applied at 160 Mg ha^{-1} to a gravelly loam potato (*Solanum tuberosum* L.) soil decreased bulk density of the plow layer (2–10 cm depth) from 1.21 Mg m^{-3} in the unamended control to 1.01 Mg m^{-3}, and also resulted in greater total porosity and saturated hydraulic conductivity (Chow et al. 2003). This improvement in soil properties

Table 3. Overview of selected field experiments examining the impact of papermill sludges on various soil biological, physical, and chemical properties

Sludge type	Rate (Mg ha^{-1})	Soil texture[z]	SOC[y]	MBC[x]	Enz. Act.[w]	Aggr.[v]	Soil water	BD[u]	Soil nutr.[t]	Citation
Deinking	32–95	cl, sil	✓				✓		✓	Aitken et al. (1998)
Deinking/poultry manure compost	14–42	sil			✓		✓		✓	Baziramakenga et al. (2001)
Deinking	50 & 100	sicl, cl	✓	✓	✓	✓	✓			Chantigny et al. (1999, 2000a)
Primary	20–160	l	✓			✓	✓	✓		Chow et al. (2003)
Combined/compost	22–45	ls	✓				✓	✓		Foley and Cooperband (2002)
Mechanical/compost	45 & 90	sl	✓	✓	✓	✓				Gagnon et al. (2001)
Combined	8.5–34	s			✓				✓	Gagnon et al. (2003)
Deinking/swine manure compost	12–35	ls		✓	✓					Lalande et al. (2003)
Deinking/secondary	8–24	sic,sl,l				✓	✓	✓		Nemati et al. (2000)
Deinking	5–16	sl	✓				✓		✓	Simard et al. (1998b)
Combined	45–225	fsl	✓			✓	✓	✓		Zibilske et al. (2000)

[z]Soil texture abbreviations, c = clay, si = silt, l = loam, s = sand, f = fine.
[y]Soil organic carbon.
[x]Microbial biomass carbon.
[w]Enzyme activity.
[v]Aggregation and aggregate stability.
[u]Bulk density.
[t]Soil nutrient levels.

resulted in a twofold time delay in runoff initiation and a 23 to 71% reduction in runoff loss with simulated rainfall. In this study, the proportion of soil aggregates in the 1–5 mm size category was increased by sludge application in comparison with the unamended control (37.5 versus 29.8%), while the proportion of aggregates <0.5 mm was decreased. Similar improvements in soil aggregation were obtained in silty clay loam and clay loam soils 1 yr after application of a deinking sludge at 50 or 100 Mg ha^{-1}, compared with the unamended soils (Chantigny et al. 1999). An increase in aggregates >1 mm arose from a decrease in aggregates in the 53 to 250 µm size fraction, and to a lesser extent from a decrease in the 250 to 1000 µm fraction; aggregate stability gradually decreased over time but was still significant 3 yr after sludge application. Sludge from a thermo-mechanical pulping newsprint process (C:N ratio of 109:1) and sludge compost (C:N ratio of 42:1), applied at 45 or 90 Mg ha^{-1}, increased the proportion of aggregates >250 µm to the same extent, in comparison with the unamended control (Gagnon et al. 2001). Although beneficial effects persisted through the 3 yr of potato cultivation, the impact of the sludge on aggregation diminished over this time period.

Soil structural stability was increased by 20% after two annual applications of deinking sludge at 18 Mg ha^{-1}, but this effect persisted <1 yr after the last application (Trépanier et al. 1998). Wet aggregate stability of a silty clay and a loamy soil was increased 15–17% by application of a combined sludge (C:N ratio of 160:1, 85% deinking sludge/15% secondary sludge) at 8 to 24 Mg ha^{-1} (Nemati et al. 2000). Enhanced stability persisted for about 1 yr, indicating annual applications were necessary for continuous benefit. Greater aggregation occurred primarily during summer months, which prompted the authors to conclude that the effects were most likely due to the production of binding substances through microbial decomposition. The abundance of aggregates >5 mm and the mean weight diameter of aggregates were reported to decrease when the fresh combined paper sludge was added to soil with mineral N fertilizer (Bipfubusa et al. 2005), also implying that benefits to aggregation are dependent on relatively undecomposed sludge. Lower sludge rates resulting in lesser increases in soil organic matter in the studies of Trépanier et al. (1996a) and Nemati et al. (2000) may explain the less persistent effects on soil aggregation in comparison with the research of Chantigny et al. (1999) and Gagnon et al. (2001). Zibilske et al. (2000) clearly showed that the onset and magnitude of decreased bulk density and increased aggregation of a fine sandy loam were dependent on the rate (45 to 235 Mg ha^{-1}) and frequency (once, biennially, and annually) of combined sludge application.

Soil Water Holding Capacity

The water holding capacity of deinking paper mill sludge was reported to be 0.36 cm^3 cm^{-3} at –33 kPa and 0.26 cm^3 cm^{-3} at –1500 kPa (Trépanier et al. 1996a), greater than most mineral soils. Therefore, most soils amended with papermill sludge are expected to have increased soil water holding capacity. For example, incorporation of a combined sludge at 246 Mg ha^{-1}, having a water holding capacity of 0.19 and 0.12 cm^3 cm^{-3} at –33 and –1500 kPa, increased soil water 20 and 74% at –33 and –1500 kPa, respectively (Zhang et al. 1993). Soil water contents of a silty clay loam and a clay loam were increased by 50 and 100 Mg ha^{-1} deinking sludge from 271 to 726 d after sludge application, compared with the unamended control (Chantigny et al. 2000a). Increased total soil water may create more favorable conditions for soil organisms, particularly during drought (Chantigny et al. 2000a).

Increased plant available water, rather than total water holding capacity, is the factor most likely to influence crop growth. Five annual applications of sludge at 225 Mg ha^{-1} to a fine sandy loam increased available water (defined as that held between –60 and –1500 kPa pressure) to about 49% (dry weight basis) compared with 26% in the unamended control (Zibilske et al. 2000). The plant available water holding capacity of a sandpit soil (0.94 kg kg^{-1} medium sand) was increased by 42, 27, and 72% at 4, 359, and 743 d after incorporation of a deinking sludge at 105 Mg ha^{-1}, and according to the authors likely contributed to the significant improvement in plant responses (Fierro et al. 1999). The slow decay of the sludge and/or the presence of clay in the sludge contributed to the persistent impact on soil water holding capacity (Fierro et al. 2000).

Some researchers have concluded that the increases in soil water holding capacity with sludge application are primarily due to the presence of relatively undecomposed sludge (Aitken et al. 1998; Chantigny et al. 2000a; Baziramakenga et al. 2001). Thus, this benefit diminishes as the sludge is decomposed. Increased soil water content of a silt loam was achieved with a deinking sludge compost at 14–42 Mg ha^{-1} 2 mo after application, but the benefit was greatly diminished by 4 mo after application and negligible the next season (Baziramakenga et al. 2001). Application of paper sludge at 32, 63, and 95 Mg ha^{-1} increased soil moisture content of a clay loam and a sandy silt loam at 6 mo, but not in the second or third season after application (Aitken et al. 1998).

Reduced irrigation requirement is another potential benefit of increased soil water holding capacity due to paper mill sludge application. Plant available water of a low organic matter Plainfield loamy sand was increased from ~0.07 to ~0.09 cm^3 cm^{-3} by two applications of combined paper mill sludge compost at 78 Mg ha^{-1}, in comparison with the unamended soil (Foley and Cooperband 2002). Thus, it was estimated that fewer irrigations (6.6 vs. 8.2) and lower water use (169 vs. 184 mm) would be required for potato production with paper mill sludge compost than with the unamended control. Benefits in irrigation efficiency and crop growth from sludge-enhanced soil water holding capacity are more likely in soils of inherently low water holding capacity than those with high water holding capacity.

Cation Exchange Capacity

Reported cation exchange capacities (CEC) of paper mill sludges vary widely: 297 cmol (+) kg^{-1} (Field et al. 1996), 62.8 cmol (+) kg^{-1} (Feagley et al. 1994a), 12–42 cmol (+) kg^{-1} (Campbell et al. 1995), 10.6 cmol (+) kg^{-1} (Cavaleri et al. 2004); 5.3 cmol (+) kg^{-1} (Fierro et al. 1999). Differences in CEC may reflect differences in sludge composition, organic matter fraction and/or clay content, or differences in measurement method. Care should be taken in interpreting sludge CEC calculated from extractable bases. This procedure, commonly used for soils, may not be accurate for sludges containing high concentrations of salts and carbonates (Sumner and Miller 1996). Irrespective of the difficulty in measuring sludge CEC, soil amendment with paper mill sludge typically results in substantial increases in soil CEC. For example, large applications of raw primary clarifier sludge (approximately 448 and 672 Mg ha^{-1}) increased soil CEC ~4 to ~12 cmol (+) kg^{-1} after 4 yr of simulated weathering at low and high application rates, respectively (Einspahr et al. 1984). Composting or allowing worms to degrade the sludge prior to incorporation doubled and tripled the increase in soil CEC in comparison with raw sludge. The 10–25% kaolin clay content of this sludge, as well as its organic matter content, likely contributed to increased soil CEC. A deinking sludge [CEC = 5.3 cmol (+) kg^{-1}] increased soil CEC about 2 cmol (+) kg^{-1} when applied at 105 Mg ha^{-1} (Fierro et al. 1999). Increasing the CEC of inherently low CEC soils, such as the sandpit soil used in this study, is more likely to benefit crop production than increasing the CEC of initially high CEC soils.

Liming Benefits

Paper mill sludges may increase soil pH substantially. The alkalinity of paper mill sludge typically arises from causticizing materials in the pulping process and/or $CaCO_3$ used in the paper finishing process. Irrespective of the source of alkalinity (NaOH, CaO, $CaCO_3$, etc.), the liming value of sludge is represented by the term "$CaCO_3$ equivalence (CCE)", indicating its neutralizing value relative to an equivalent weight of pure $CaCO_3$. The CCE of a primary sludge from a bleached Kraft mill in Portugal was reported to range between 25 and 50% (Valente et al. 1987), a primary sludge from Louisiana was 33% (Feagley et al. 1994a), a combined sludge from Maine was 14.6% (Carpenter and Fernandez 2000), a primary sludge from Oregon was 12.7% (Sherwood 1992), a deinking sludge from Wisconsin was 9.2% (Diehn 1991), and a deinking sludge from the United Kingdom was 5.9% (Aitken et al. 1998).

Primary clarifier sludge applied to a silt loam soil at 13 and 26 Mg ha^{-1} increased the pH of the upper 15 cm depth 0.4 and 0.8 units, respectively (Legendre et al. 2004). Deinking sludge at 105 Mg ha^{-1} increased the soil pH of the upper 20 cm of an abandoned sandpit from 4.7 to >6.0, 2 yr after application (Fierro et al. 1999). Soil pH in the upper 20 cm was increased from 2.9 to 7.3 after application of a combined sludge at 860 Mg ha^{-1} to an abandoned coal minesoil for at least 59 mo (Kost et al. 1997). A more modest increase in soil pH of 0.4 pH units was recorded at 42 Mg ha^{-1} for a deinking sludge/poultry manure compost (Baziramakenga et al. 2001). In most cases, pH increases due to sludge addition do not impact crop growth because the initial soil pH is adequate and the subsequent pH with amendment is not excessive (Legendre et al. 2004). However, when initial soil pH is low, crop response to increased pH with sludge application may be substantial (Fierro et al. 1999).

CROP RESPONSES TO PULP AND PAPER MILL SLUDGES

Low Nutrient Sludges

Although primary and deinking sludges frequently improve soil physical, chemical, and biological properties (Table 3), beneficial crop responses to low nutrient sludges have been limited. Immobilization of N is thought to be responsible for the detrimental effects on crop production. Seven field studies conducted with high C:N paper mill sludges are summarized in Table 4. Reduced crop yield occurred in the year of sludge application in five of seven studies and was attributed to reduced N availability. This occurred despite adding N fertilizer to sludge treatments and beneficial effects of sludge on soil properties. The negative effects of sludge application on N availability generally dissipated in the next

Table 4. Crop yield responses to application of low nutrient papermill sludge in field trials.

Sludge type	Soil[z] texture	Crop	C:N	Appl. freq.[y]	Rate, dry (Mg ha^{-1})	Yield of crop with sludge versus control[x] and causal factor[w] — Years after application: 0	1	2	Citation
Primary	sil	Sugarcane	70:1	Once	13 & 26	=	=	=	Legendre et al. (2004)
Deinking (site 1)	cl	Spring barley (yr 0), winter barley (yr 1),	86:1	Once	32	↓N	=	↑N	Aitken et al. (1998)
		spring barley (yr 2)			64 & 95	↓N	=	=	
Deinking	s	Tall wheatgrass	114:1	Once	105	↑SSP	↑SSP	NE	Fierro et al. (1999)
Primary recycle	NR[v]	Spring oat (yr 0), winter wheat (yr 1)	169:1	Once	56–168	↓N	=	NE	Field et al. (1996)
Deinking	cl, sicl	Sweet & red clover	240:1	Once	50 & 100	=	↑N	NE	Allahdadi et al. (2004)
		Alfalfa, trefoil				↓N	↑N	NE	
		Bromegrass				↓N	↓N	NE	
Deinking	sl	Barley (yr 0), Strawberry (yr 1)	288:1	Once	5–16	↓N	=	NE	Simard et al. (1998b)
Primary	sil	Cotton	323:1	Once	134	↓N	=	↑N	Boquet et al. (2001)

[z] Soil texture abbreviations, c = clay, si = silt, l = loam, s = sand.
[y]Frequency of sludge application.
[x]The effects of sludge on crop yield are compared with the highest yield obtained with mineral fertilizers, except where noted.
↑, ↓, and = denote increased, decreased, and equivalent yield.
[w]Causal factor is that attributed by the authors or evident from the data. An "'N"' in combination with a downward ↓ indicates a yield decrease due to decreased N supply, an "'N'" with an upward ↑ indicates a yield increase due to enhanced N supply, and "'SPP"' with an upward ↑ indicates a yield increase due to improved soil physical properties. NE = not evaluated.
[v]Not reported.

cropping season, when yields were equivalent to those obtained with standard fertilizer treatments. A slight increase in crop yield 2 yr after sludge application was realized in the only two studies conducted for three or more seasons. Perhaps the modest enhancement in crop yield should not be unexpected given the inherently good soil properties of the arable soils used in these studies.

In contrast to the studies noted above, tremendous improvements in tall wheatgrass [*Elytrigia elongata* (Host) Nevski] establishment and growth were obtained with sludge in the restoration of a sandpit soil (Fierro et al. 1999). Incorporation of a deinking sludge improved many properties of the sand soil that potentially limited plant production, including pH, CEC, water holding capacity, and bulk density. Additionally, adequate supplemental N and P were provided based on sludge rate, so these nutrients were not limiting.

Nutrient Immobilization

The most important factor limiting positive crop responses to low nutrient concentration paper mill sludges is the immobilization of nutrients. Nitrogen is the most frequently occurring sludge-induced deficiency and therefore the best understood. Deficiencies in P and S are also thought to result from additions of paper mill sludges low in these nutrients, but little is known about their frequency of occurrence.

Organic materials with C:N ratios >20–30:1 generally result in some period of N immobilization (Alexander 1977), and primary sludges typically greatly exceed this ratio. Therefore, the amount and duration of N immobilization can be extreme. A primary sludge with a C:N ratio of 480:1 immobilized N at all application rates (17–267 g sludge kg soil^{-1}) for at least 250 d (Zibilske 1987). Increasing application rate increased both the amount and duration of N immobilization. Net immobilization occurred throughout the 250-d incubation period and no mineral N was detected in the soil at the highest application rate at 25°C, whereas net immobilization lasted about 60 d at the lowest rates and was followed by a net N mineralization phase. At 12°C, the immobilization period at lower sludge rates was longer, about 100 d. Effects of temperature on N immobilization were somewhat different for a higher N content paper mill sludge (C:N ratio of 43:1) (Zibilske 1997). Faster microbial growth at 25°C resulted in N immobilization for the first 40 d of incubation but no immobilization occurred at 12°C. After 40 d, the amount and rate of N release from the sludge was greater at 25°C than at 12°C (1.2 vs. 0.4 mg N kg soil^{-1} d^{-1}) at a sludge application rate of 160 g kg soil^{-1}.

The amount and duration of N immobilization during the decomposition of sludge are dependent upon several factors including the amount of sludge, its C:N ratio, soil inorganic N supply and soil type. The occurrence of immobilization as it relates to the timing and quantity of N needed by the crop determines the impact on crop production. Nitrogen deficiency occurs when low-N sludges are added shortly before planting of annual crops. For example, a raw deinking sludge with a C:N ratio of 288:1 applied at 16 Mg ha^{-1} without additional N decreased barley (*Hordeum vulgare* L.) yield by 50% (Simard et al. 1998b). Spring barley yield was decreased by deinking sludge (C:N ratio of 86:1) at 32, 63, and 95 Mg ha^{-1} with fertilizer N at 0 or 40 kg ha^{-1} (Aitken et al. 1998).

One strategy suggested for overcoming N immobilization is a fallow period after primary sludge application to allow reduction of the sludge C:N ratio (Dolar et al. 1972). However, this approach is complicated by the great variation in duration of the immobilization period. For example, Simpson et al. (1983) found sludge application should precede planting by 4 wk for primary sludge~~s~~ and only 2 wk for secondary sludge~~s~~. Generally, estimates of the immobilization period are greater; hence the recommended delay between sludge application and planting is necessarily longer: 60 d (Hatch and Pepin 1985), 90 d (Dolar et al. 1972), or several months (Shimek et al. 1988). In some instances, the negative effects of N immobilization are not completely overcome until the next growing season. Nearly half the fertilizer N added to primary sludge amended soil was immobilized in the first season after application, but almost none was immobilized in the second year (Henry 1991). The negative impact of application of a deinking sludge with a C:N ratio of 86:1 at 32, 63, and 95 Mg ha^{-1} on cereal and linseed (*Linum usitatissimum L.)* yields was diminished in the second and third year after application compared with the first season (Aitken et al. 1998). Primary paper mill sludge (C:N = 323:1) at 134 Mg ha^{-1} decreased cotton (*Gossypium hirsutum* L.) lint yield by 70% in the year of application, but had no effect on yield in the following 4 yr (Boquet et al. 2001).

Supplemental Nitrogen to Overcome Immobilization

One of the simplest and most frequently used approaches to overcome immobilization is addition of supplemental N to compensate for the low N content of the sludge. For example, Simard et al. (1998b) found that, at the standard rate of fertilization (70 kg N ha^{-1}), increasing sludge rate reduced barley grain yield due to a reduction in plant N availability. Supplementing the standard N rate with another 135 kg N ha^{-1} provided enough N to overcome immobilization so that barley yields were not reduced by increasing sludge rate. Supplemental N may also come from farm manures with high N and low C:N ratios, such as swine or poultry manure, which can be added simultaneously to soil (Carneiro and Dos Santos 1996; Ferguson 1997; Gagnon et al. 2004).

The amount of N needed to overcome the high C:N ratio of primary sludges varies substantially based on research and experience, from 0.9 kg N Mg^{-1} sludge to more than 8 kg N Mg^{-1} (Table 5). Supplemental N needs are likely dependent on the C:N ratio of the sludge, the application rate, the soil type and residual N content, the cropping system, and temperature and moisture conditions.

Use of Legumes to Overcome Nitrogen Immobilization

Another approach to overcome the high C:N ratio of some paper mill sludges is to grow a legume, since they are less reliant on soil NO_3 and NH_4 for growth than non-legumes. For example, the application of paper mill sludge with a C:N

Table 5. Amount of supplemental nitrogen required to overcome nitrogen immobilization with land application of papermill sludges of various C:N ratios.

Sludge type	C:N	Sludge rate (Mg ha^{-1})	Supplemental N (kg N Mg^{-1} sludge)	References
Deinking	86:1	32	1.3	Aitken et al. (1998)
Recycle	125:1	12	8.3	Bellamy et al. (1995)
Primary	151:1	56	3.0	Dolar et al. (1972)
Recycle	169:1	112	0.9	Field et al. (1996)
Deinking	169:1	7–21	≤3.0	Simard et al. (1998a)
Deinking	240:1	–	5.3–8.4	Fierro et al. (1997)
Deinking	288:1	16	4.3–5.6	Simard et al. (1998b)
Deinking	344:1	12	7.5	Trépanier et al. (1996a)

ratio of 240:1 at 50 or 100 Mg ha^{-1} completely eliminated growth of the non-legume bromegrass (*Bromus inermis* L.), but growth of legumes such as alfalfa (*Medicago officinalis* L.), birdsfoot trefoil (*Lotus corniculatus* L.), red clover (*Trifolium pratense* L.), and yellow sweet clover [*Melilotus officinalis* (L.) Lam.] persisted, albeit with some reduction in the growth of alfalfa and birdsfoot trefoil (Allahdadi et al. 2004). The authors attributed the growth reduction in alfalfa and birdsfoot trefoil to a greater dependence of these crops on soil-derived N relative to the other legumes. Sufficient sludge decomposition had occurred by the second cutting in the year after sludge application so that N immobilization no longer reduced growth of any of the legumes.

Subterranean clover (*Trifolium subterranean* L.W. woogenelux) growth was decreased after addition of sludge with a C:P ratio of 1580:1 (Feagley et al. 1994b) suggesting this legume, like alfalfa and birdsfoot trefoil (Allahdadi et al. 2004), might have greater dependence on soil derived N. Tissue N concentration decreased substantially as sludge rate increased in support of this theory; however, tissue P concentration was also severely decreased by sludge application. Addition of fertilizer P substantially increased dry matter production and tissue N and P concentration, suggesting decreased P availability, at least in part, caused reduced growth at high sludge rates. Similar results were obtained with application of a paper mill sludge (C:P ratio of 943:1) to yellow lupin (*Lupinus luteus* L.) (Vasconcelos and Cabral 1993). Reduced P availability after addition of a primary deinking sludge (C:P ratio of 6400:1) was proven to impact the growth of three legumes [galega (*Galega orientalis* Lam.), black medic (*Medicago lupulina* L.), and yellow sweet clover] in studies conducted in sand and soil media (Fierro et al. 1997). The legumes required an additional 0.5 to 1.2 g P kg^{-1} of sludge to attain dry matter levels approaching that obtained in soil without sludge. Microbial immobilization of soil P is likely to occur with addition of organic substrates with a C:P ratio >300 (Alexander 1977), even if they are not always observed (Gagnon et al. 2004). Phosphorus immobilization due to high C:P ratio paper mill sludge addition may not interfere with plant growth in P-rich soils.

Composting

Composting primary sludges with secondary sludges (Campbell et al. 1991; Sesay et al. 1997; Vagstad et al. 2001), animal manures and other waste products (Campbell et al. 1995; Valente et al. 1987; Sesay et al. 1997; Simard et al. 1998a; Baziramakenga et al. 2001; Baziramakenga and Simard 2001; Das et al. 2001; Lalande et al. 2003), municipal biosolids (Line 1995), or fertilizers (Valente et al. 1987; Brouillette et al. 1996; Das et al. 2001; Gagnon et al. 2001) has been studied as a means of overcoming the low nutrient content of primary sludges. Nitrogen immobilization induced by primary sludges can be reduced if the sludge is first co-composted with N-rich wastes. Likewise, N availability from the low C:N materials alone, such as animal manure or fertilizers, is moderated by composting with the primary sludge. Composting manures and other high-nutrient waste products with primary sludges has other benefits as well, including reduced mass (Campbell et al. 1995; Line 1995; Brouillette et al. 1996; Das et al. 2001) and reduced concentration of organic compounds, such as resinic acids and polycyclic aromatic hydrocarbons (Beauchamp et al. 2002). Other potential benefits are reduced pathogen content and odor.

Composting and curing a primary sludge (initial C:N ratio 270:1) for a total of 18 wk with ash, tailings, and slaughterhouse waste reduced the C:N ratio to between 14:1 and 67:1 dependent on the amounts of ash added to the mixture (Campbell et al. 1995). Ten months of composting a deinking sludge (C:N ratio of 330:1) with N-P-K fertilizers reduced the C:N ratio to a 40:1 to 60:1 range depending on the aeration conditions and position in the compost pile (Brouillette et al. 1996). Cellulose, hemicellulose, and lignin were decreased to 57, 61, and 85% of their initial values. The cellulose content of another primary sludge was decreased by 50% when composted with cattle manure and fertilizer for 75 to 140 d (Valente et al. 1987). The persistence of lignin during the composting process may explain why the effects of composted paper mill sludges on soil physical and biochemical properties may be no different from those of raw sludges (Gagnon et al. 2001).

High Nutrient Sludges

Secondary and combined sludges and sludge composts can be significant sources of essential plant nutrients, particularly N, P, and K. Secondary and combined sludges typically increase soil N levels with no immobilization or only a short period of immobilization. As the C:N ratio approaches 20:1, the duration and impact of N immobilization decrease. For example, a combined primary/secondary sludge with a moderate C:N ratio (28:1 to 42:1) was an effective source of N for cabbage (*Brassica oleracea* var. *capitata* L.) and sweet corn (*Zea mays* L.) (Simard 2001). Apparent N recovery from the sludge (34%) was comparable with that from ammonium nitrate fertilizer (38%).

Sweet corn showed N deficiency early in the season after application of a primary/secondary sludge (C:N ratio of 25:1), but not after temperatures warmed (Thiel 1984). Immobilization of soil nitrates occurred in June after winter and spring application of sludge with a C:N of 23:1 (Bowen et al. 1996). However, potato yield and N removal indicated that N mineralization was sufficient to supply N equivalent to 180 kg N ha^{-1} as ammonium nitrate. Nitrogen uptake efficiencies of three combined paper mill sludges applied at total N rates of 131, 120, and 88 kg N ha^{-1} to a timothy (*Phleum pratense* L.) dominated grass-alfalfa sward were 9, 39, and 58%, respectively, in the first year of application, compared with 35% for ammonium nitrate at 100 kg N ha^{-1} (Arfaoui et al. 2001). Differences in N availability among the three sludges were not predictable from their N contents (35, 17, and 34 g kg^{-1}) or C:N ratios (14:1, 28:1, and 15:1). Apparent N use efficiency of paper mill sludges ranged from 16 to 69% in eight experiments conducted in Eastern Canada summarized by Gagnon and Ziadi (2004).

In the absence of immobilization, the time course of N release from secondary, combined, and composted paper mill sludges generally follows a two-phase model with the rate of N release being faster in the initial rather than the latter phase (Zhang et al. 1993; Zibilske 1997). Explanations for this phenomenon include the exhaustion of easily degradable C substrates and/or depletion of soil nutrients such as N, P, or S by microorganisms as sludge decomposition proceeds. The rate and total quantity of inorganic N released from a secondary treatment sludge was greater at 25°C than 12°C over a 229-d incubation period (Zibilske 1997). The rate of N release at 25°C was 1.3 mg N kg $soil^{-1}$ d^{-1} at a sludge application rate of 90 g kg $soil^{-1}$, compared with 0.7 mg N kg $soil^{-1}$ d^{-1} at 12°C with the same sludge rate. Nitrogen mineralization from a secondary sludge (C:N ratio of 5:1) averaged 71%, 12 mo after application to sandy and clay soils in a maritime climate (Henry 1991).

Combined, secondary and composted sludges can also be significant sources of other plant nutrients. A combined sludge applied at 8.5 and 17.5 Mg ha^{-1} provided enough P (15 and 30 kg P ha^{-1}) and K (10 and 20 kg K ha^{-1}) to meet the needs for these nutrients for native lowbush blueberry (*Vaccinium angustifolium* Ait.) (Gagnon et al. 2003). Berry yields were equivalent to or greater than those achieved with mineral fertilizer supplying comparable levels of available N, P, and K. A deinking sludge/poultry manure compost applied at 14, 28, and 42 Mg ha^{-1} provided P at 25, 50, and 75 kg ha^{-1} and K at 12.5, 25, and 37.5 kg ha^{-1} (Baziramakenga and Simard 2001). Total uptake of P and K from the compost by snap bean (*Phaseolus vulgaris* L.) was no different from that from mineral fertilizer in this study. Recovery of P and K added in compost ranged from 12.5 to 5.3% P and from 58 to 36% K, for low to high application rates, respectively. Mehlich-3 extractable P was enhanced by the addition of compost, perhaps due in part to a reduction in P sorption onto soil mineral particles as well as addition of soluble P (Baziramakenga et al. 2001).

Some studies indicated that higher crop yields could be achieved when combined or composted paper sludge were applied in combination with mineral N fertilizers than when these materials were applied alone (Simard et al. 1998a; N'Dayegamiye et al. 2003). This observed synergistic effect might be attributed to closer synchronization of nutrient supply and crop demand and/or improved soil physical conditions and microbial activities. However, given the substantial improvements in soil quality obtained with application of paper mill sludges previously noted, it is surprising that this synergism has been the exception, not the rule.

ADDITIONAL USES OF PULP AND PAPER MILL SLUDGES

Several projects have investigated the use of paper mill sludges in plant and soil systems other than traditional agricultural systems. Paper mill sludges have been evaluated and used to successfully amend disturbed soils, fostering revegetation and reducing erosion (Feagley et al. 1994a, b; Kost et al. 1997; Fierro et al. 1999). Artificial soils manufactured from paper mill sludges in combination with other residuals were found superior to natural topsoil for reclaiming a gravel pit (Carpenter and Fernandez 2000). Paper mill sludges have also been used as components of nursery container potting mixes (Chong and Cline 1993; Bellamy et al. 1995) and in the establishment of turfgrasses (Norrie and Gosselin 1996). Sludges left on the soil surface as mulch (Campbell et al. 1995) benefit crop growth through conservation of moisture (Henry 1991; Gagnon et al. 2003). Application of primary paper mill sludges to soils high in NO_3-N has been proposed as a method for reducing NO_3-N movement to ground water (Zibilske 1987; Cabrita et al. 1996).

SUMMARY

Paper mill sludge composition varies with pulp and paper making processes and waste treatment. The high content of recalcitrant organic matter in paper mill sludges, particularly those high in primary and deinking sludge, has profound and persistent benefits on soil physical and chemical properties. Substantial increases in soil organic matter content and soil aggregation often result in lower soil bulk density and increased total and available water. Improved aggregate formation and stability strengthen soil structure, thereby reducing surface runoff and erosion. Paper mill sludges may also have a liming capacity, which can be important in acidic soils. Nutrient immobilization, particularly N but also P and S, is an overriding concern with low nutrient sludges and typically reduces crop production in the year of sludge application. Supplemental fertilization, delayed planting of the crop, use of legumes, and composting are successful approaches for reducing the impact of nutrient immobilization on crop production. Finally, paper mill sludges combined with secondary treatment sludges and/or composted with fertilizers or other high-nutrient by-products can provide significant amounts of N, P, and K to crops while still enhancing soil physical properties.

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