Fly-ash from a pulp and paper mill: A potential liming material for agricultural soils in Western Newfoundland

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ABSTRACT

Most agricultural soils in Western Newfoundland are acidic and need lime to raise soil pH to be productive. Corner Brook Pulp and Paper Ltd produces a substantial amount of fly-ash, disposed of at a local landfill. This study was conducted to assess the potential for using fly-ash as a liming material for agricultural soil (pH 5.5) in Western Newfoundland. Heavy metal concentration in the soil and fly-ash were analysed and compared with soil and compost guidelines. As per quality guidelines, only part of the lime requirement can be substituted by fly-ash. The percentage may vary depending on initial soil pH and the desired pH for the crop to be grown. The total lime requirement can be met when fly-ash is applied combined with other soil amendments low in trace element concentration.

RÉSUMÉ

La plupart des sols de l'ouest de Terre-Neuve sont acides et nécessitent de la chaux pour augmenter le pH du sol et les rendre productifs. La papetière Corner Brook Pulp and Paper Ltd produit une quantité importante de cendres volantes qui sont éliminées dans le site d'enfouissement local. Cette étude a été réalisée pour évaluer le potentiel de l'utilisation des cendres volantes comme amendement calcaire pour les sols agricoles (pH : 5,5) dans l'ouest de Terre-Neuve. Les concentrations des métaux lourds dans le sol et dans les cendres volantes ont été analysées et comparées aux lignes directrices relatives aux sols et au compost. Pour ce qui est des lignes directrices de qualité, seulement une partie des besoins en amendement calcaire peut être comblée par les cendres volantes. Le pourcentage peut varier selon le pH initial du sol et le pH désirable pour la culture à produire. Les besoins totaux en amendement calcaire peuvent être satisfaits lorsque les cendres volantes sont combinées à d'autres amendements faibles en oligo-éléments.

KEYWORDS

Fly-ash, heavy metal, liming, pH, Western Newfoundland

MOTS CLÉS

Cendres volantes, métaux lourds, chaulage, pH, Ouest de Terre-Neuve.

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INTRODUCTION

Most agricultural soils in Western Newfoundland are strongly leached under natural conditions, and they are acidic with a loamy texture (Acton and Gregorich 1995). These acidic soils need a large amount of limestone per hectare to achieve the desired pH level for cultivation. Regular applications are required to maintain the soil pH in a desirable range for crop production (Atlantic Soil Fertility Committee 1970). There is a significant cost for farmers due to this high lime requirement. Failure to add lime to acidic soils reduces crop yields (Schwartz and Follett 1979), wasting much of the farmers' fertilizer investment and increasing the cost of production. The use of wood biomass as a fuel source for the Pulp and Paper industry has led to the production of wood ash as a by-product (Pöykiö et al. 2004). Presently, Corner Brook Pulp and Paper Ltd (CBPPL) mill uses a hog fuel and waste oil mix as a fuel source for their boiler, which generates steam for paper manufacturing and energy production. The fuel mixture comprises approximately 90% biomass (wood waste and sludge from waste treatment) and 10% waste oil (Production Manager CBPPL). Annually, due to the incineration of large amounts of biomass, a considerable amount of total ash (fly-ash and bottom ash) is generated from mill operation and disposed to landfills (Department of Environment and Climate Change, 2017). CBPPL mill currently collects 10,000 to 15,000 Mg of fly-ash each year. Ash is disposed of at the local landfill site with an approximate annual cost of $250,000 (Churchill and Kirby 2010). This material must be handled properly to prevent a severe environmental threat since leachate from wood ash may contain a high concentration of heavy metals and other contaminants (Poykio et al. 2004). There is a high potential for wood ash to be used as an alternative liming material and fertilizer in agricultural lands (Demeyer et al. 2001) due to its high pH (Campbell 1990; Pitman 2006) and thus the ability to increase the pH of acidic soils resulting in improved nutrient uptake by crop plants (Naylor and Schmidt 1989; Williams et al. 1996). Also, there is a demand for low-cost liming materials in the Atlantic region due to high soil acidity and the considerable cost of liming materials (Atlantic Soil Fertility Committee 1970).

There are various advantages to diverting the ash generated by the pulp and paper industry as an alternative liming agent; not only would they save on disposal fees, but there would be environmental benefits like reduction of material landfilled, reducing the demand on the landfill site and the risk of groundwater contamination etc. Besides, farmers can save money due to increased fertilizer efficiency and cost reduction for liming material using the available ash (Alberta Agriculture 2002). Some studies have revealed that wood ash from a paper mill can have some metal contaminants (Pokio et al. 2004; Pitman 2006; Jukic et al. 2017), which can have long-term adverse effects on the ecosystem (Singh et al. 2011) when applied to agricultural soils as a liming material. That may limit the use of ash as a substitute for agricultural liming material. The general objective of this study is to assess the potential of fly-ash from CBPPL as an alternate liming material for Newfoundland agricultural soil. The specific objectives of this study were; (1) to evaluate the quality of soil and fly-ash by comparing with soil and compost quality guidelines, (2) to determine the lime requirement and the application rate of the agricultural soil, and (3) to determine the maximum allowable application rate of fly-ash to the study soil based on the heavy metal contents and the guidelines.

MATERIALS AND METHODS

Soil and fly-ash sample collection and analysis

The required soil for this study was sampled from a fallow area in the Pynn's Brook Research Station (PBRS), Pasadena (49°04'23"N, 57°33'39"W), in the Humber Valley Watershed of Western Newfoundland, Canada. The soil samples were collected from shallow depths (0-30 cm) from several places from the PBRS and combined to make a composite sample for this research to represent most of the root zone in humid regions. The collected soil samples were air-dried for one week and sieved to a particle diameter of <2 mm. Particle size analysis was carried out using the hydrometer method (Kroetsch and Wang 2007) and showed a sandy loam texture. Bulk density (BD), pH_{CaCl2}, cation exchange capacity (CEC), and soil organic matter (SOM) of the soil were analyzed using standard methods (Soil sampling methods and analysis 2007). A composite sample (n=15) of fly-ash was collected from the CBPPL mill during October 2017. The collected fly-ash sample was analyzed (n=12) for different physicochemical parameters. Heavy metals concentration (Vanadium (V), chromium (Cr), arsenic (As), selenium (Se), mercury (Hg), nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd), cobalt (Co) and lead (Pb)) in the soil sample and fly-ash sample were analyzed by using inductively coupled plasma mass spectrometry (ICP-MS) after acid digestion (EPA method 3050B).

Data analysis and interpretation

The measured heavy metal values in the soil were compared with the Canadian Council of Ministers of the Environment (CCME) Soil Quality Guidelines, and heavy metal contents in the fly-ash were compared with the CCME compost quality guidelines and the Canadian Food Inspection Agency (CFIA) fertilizer and supplement metals standards. CFIA fertilizer and supplement metals standards are predicated based on the maximum acceptable cumulative addition to soils over 45 years instead of the actual concentration of the metal in the product (Canadian Food Inspection Agency 2017). The 45-year cumulative application approach is proposed to account for the continued existence of metals once added to the environment, which eventually determines the level of contamination and, thus, long-term effects (Canadian Food Inspection Agency 2017). The maximum acceptable product metal concentration (mg metal per kg of product) was calculated using Eq. 1.

$$A = 10^6 \frac{mg}{kg} \times \frac{max.acceptable\ cumulative\ metal\ addition\ to\ soil\ over\ 45\ years\ (B)}{45\ years \times annual\ application\ rate\ (C)} \quad (1)$$

where, A - Maximum acceptable product metal concentration (mg metal per kg of product), B - CFIA Standards for maximum acceptable 45-year cumulative metal additions to the soil (kg metal ha^{-1}), C - Product maximum recommended annual application rate (kg product ha^{-1} $year^{-1}$). Based on the maximum acceptable 45-year cumulative metal additions to the soil, the possible or allowable maximum annual fly-ash application to the field was calculated using the above Eq. 1. The average metal concentration of the fly-ash was assumed as the maximum acceptable product metal concentration.

The soil sample was analyzed for lime application requirement for two different types of forages at the Government of Newfoundland and Labrador (NL) run Soil and Plant Laboratory, located in St. John's, NL. Statistical analyses to compare the average results were performed using a one-sample t-test. The statistical analyses were carried out by using SPSS version 2010. Calcium Carbonate Equivalency (CCE) of the fly-ash sample was calculated by using a titrimetric method. The CCE of fly-ash is generally determined by the Association of Official Analytical Chemists (AOAC) for agricultural limestone. The procedure involves heating a sample in HCl and back-titration of the residual acid with NaOH. The lime requirement was converted to a fly-ash recommendation by the following calculation (Lickacz 2002) Eq. 2.

$$\text{Fly} - \text{ash application rate} = \frac{100}{CCE\ of\ ash\ (A)} \times \frac{100}{100 - moisture\ \%\ (B)} \times lime\ requirement\ (C) \quad (2)$$

where, A - CCE of the fly-ash. CCE is a measure of the liming ability of the ash compared to pure calcium carbonate, B - percent moisture in the fly-ash and C - lime requirement as provided by the soil testing laboratory.

RESULTS AND DISCUSSION

Soil analysis

The soil type was an Orthic Humo-Ferric Podzolic soil. The soil texture in the top 0–30 cm soil layer was sandy loam according to the United States Department of Agriculture (USDA) soil classification (sand=66.3% (± 3.2), silt=25.2% (± 4.6), and clay= 8.5% (± 1.3)). pH, soil OM (%), BD (g cm^{-3}) and CEC (cmol kg^{-1}) of analysed soil samples were 5.5, 3.82, 1.3 and 15.2, respectively. Available macronutrients, magnesium (Mg), calcium (Ca), potassium (K) and phosphorus (P) content (%) of the soil were 0.27, 1.1, 0.12 and 0.03, respectively**.** Soil pH affects the physical, chemical, and biological properties of soils and crop yields. Also, it plays a significant role in the solubility of nutrients and metals within the soil profile. The results indicate that the soil is strongly acidic, with a pH of 5.5. In strongly acid soils, the availability of the macronutrients and micronutrients such as molybdenum and boron are reduced (Lohry 2007). Soil sample analysis shows that available nutrient content is very low. At this pH level, certain micronutrient deficiencies are common on some sandy soils. Low pH in topsoil may affect microbial activity, most notably decreasing legume nodulation (Bargaz et al. 2018). This soil needs a lime application to increase the soil pH to the desired level for effective crop production. Soil pH also influences the CEC of soil colloids that have a pH-dependent type of charge (Karak et al. 2005). CEC is another important chemical property of soil affecting crop productivity, and studied soil in our research has a low CEC. Soils with low CEC are more likely to develop deficiencies in K and Mg (Lombin 1979). Additionally, the studied soil has a relatively low SOM content as well which is 3.82%, resulting in relatively poor soil in terms of agriculture.

The CCME has developed soil quality guidelines depending on land use. The agricultural soil quality guidelines are the most restrictive compared to other guidelines (residential, parkland, commercial or industrial). Heavy metal concentrations within the sampled soils and the CCME quality standards for the agriculture soil are presented in Table 1. In the soil studied, Cd has the lowest mean concentration (0.25 ± 0.11 mg kg^{-1}), while the highest concentrations were recorded for Zn (63.02 ± 7.42 mg kg^{-1}). There were no detectable amounts of Se and Hg observed in the analyzed soil sample. According to the CCME Quality guidelines for agricultural soils, heavy metal concentrations in the studied soil are generally below the threshold values as shown in Table 1. The soil can be considered for agricultural purposes.

Fly-ash analysis

About 88% of biomass and 12% of waste oil (on average) were used as a boiler fuel during the sampling period. The fuel mix (typically biomass, 90% and waste oil, 10%) used in the CBPPL boiler varies depending on boiler conditions, moisture content of feedstock and steam and power requirements of the mill (Churchill and Kirby 2010). The

Table 1. Heavy metal contents in soil samples. (n=7). Mean values are presented with standard deviation and CCME limits for agricultural soils (ND = Not detected).

Elements	Concentration (mg kg^{-1})	CCME limits for Agriculture Soil (mg kg^{-1})
Cr	17.18 ± 2.74	64
Ni	20.14 ± 4.26	45
Co	4.59 ± 1.1	40
Cu	8.04 ± 1.8	63
Zn	63.02 ± 7.42	200
As	2.87 ± 0.89	12
Cd	0.25 ± 0.11	1.4
Pb	8.66 ± 1.15	70
Se	ND	1
V	27.47 ± 2.32	130
Hg	ND	6.6

biomass portion of the fuel mix is a mixture of hog fuel and dewatered secondary sludge from the secondary effluent treatment system. Hog fuel is a mix of coarse chips of bark from trees, sawdust, and wood fibre. Waste oil is a high-viscosity residual oil used at the CBPPL for enhancing the combustibility of wet biomass. Waste oil may contain varying types, and amounts of contaminants and the amount of oil used may affect the contaminant levels in the fly-ash. In the present study, the pH of the fly-ash sample was measured as 10.0; it indicates that fly-ash was highly alkaline and can be used for the reclamation of acidic soils. Fly ashes differ widely in their pH (3.8-12.8) and their chemical characteristics depending on their source (Yunusa et al. 2012), thus directly affecting the availability of macro and micronutrients. The concentrations of the Ca, K, and Mg (macro-nutrients) in the fly-ash from the CBPPL were 8.98, 4.09 and 3.01%, respectively and correspondingly 8, 34 and 11 times higher than in the soil collected from the field in the PBRS. Thus, because of its high nutrient content, the utilization of fly-ash as a nutrient source can also be recommended.

The heavy metal analysis results of the composite fly-ash sample used in this study are given in Table 2. In the fly-ash sample, Hg has the lowest mean concentration (0.21 ± 0.11 mg kg^{-1}), while the highest content was recorded for Zn (1061.63 ± 89.28 mg kg^{-1}). There was no detectable amount of Se and Th observed in the analyzed fly-ash sample. Fly-ash is chemically composed of many trace and heavy metals in variable proportions (Basu et al. 2009; Demeyer et al. 2001; Kishor et al. 2010). The results of fly-ash analyses have shown a complex and heterogeneous nature of this material. High levels of trace elements (As, Cd, Cr, Pb, Hg, Se, and Mo) seem to be the biggest problem for the agricultural use of fly-ash (Aitken and Bell 1985; Carlson and Adriano 1993). Regulations in some provinces limit the input of heavy metals into the soil and may restrict the use of fly-ash. In the present study, the composite fly-ash sample showed a CCE value of 42%. In general, wood ash has a CCE ranging from 35% to 85% (Nkana et al. 1998) and is commonly used as a liming amendment. Unlike limestone, wood ashes contain carbonates and other components that may react with acid and affect the CCE determination. Due to its chemical properties (pH and CCE), this fly-ash can be used as a liming material and would be available to farmers at a very low cost. However, to use this fly-ash as a liming material or soil amendment, it would need to meet a quality guideline set by the jurisdiction of usages, such as the CCME or CFIA quality guidelines.

Comparison of heavy metal contents with CCME compost quality guidelines

Two compost categories (A and B) have been developed based on trace element concentrations and foreign matter by CCME (Canadian Council of Ministers of the Environment 2005). According to the one-sample t-test, measured heavy metal concentrations in the fly-ash sample are significantly ($p < 0.000$) below the CCME compost category B guidelines. It could potentially be used following the category B compost regulations. Under the categories B, fly-ash can be used as a soil amendment with additional control. When compared with the CCME compost category A guidelines, one sample t-test showed that most of the metals were within the allowable limits (measured heavy metal concentrations in the fly-ash sample were significantly ($p < 0.000$) below the threshold values) except for Ni, Zn and Mo concentrations. The concentration of Ni and Zn in fly-ash was significantly ($p < 0.000$) higher than the compost A guidelines. The mean concentration of Mo was slightly higher than the guidelines values, but the difference was not significant. If the fly-ash was to be incorporated with other components that are low in the above elements (compost, manure, sludge or organic waste), the final compost might pass as a category A compost. However, Zn may be present in other natural materials used as an additive. It is an essential element, present in the tissues of animals and plants even at normal, ambient concentrations. If plants and animals are exposed to high concentrations of Zn, significant bioaccumulation can result, with possible toxic effects (Wuana and Okieimen 2011). Therefore if a compost material is prepared, the metal analysis still should be required.

Fly-ash application based on CFIA fertilizer and supplement metals standards

Accumulation of metals of concern in soil over the long period can have long-term effects on the ecosystem and cause environmental toxicity. The application rate of a product is the main factor in defining acceptable product metal concentrations. Table 3 shows the maximum acceptable cumulative metal additions to the soil over 45 years and the maximum allowable annual application of CBPPL fly-ash based on each average element

Table 2. The concentrations of the heavy metals in fly-ash from CBPPL. Mean values are presented with standard deviation (n = 12) and CCME compost guidelines (ND = Not detected, NA = Not available).

Elements	Concentration (mg kg^{-1})	CCME compost	
		A	B
Cr	129.84 ± 17.47	210	1060
Ni	109.90 ± 18.41	62	180
Co	19.22 ± 1.63	34	150
Cu	197.41 ± 10.76	400	2200
Zn	1061.63 ± 89.28	700	1850
As	4.91 ± 0.76	13	75
Cd	0.93 ± 0.14	3	20
Pb	12.67 ± 0.8	150	500
Mo	5.6 ± 1.8	5	20
Hg	0.21 ± 0.11	0.8	5
V	58.0 ± 4.5	NA	NA
Se	ND	NA	NA
Tl	ND	NA	NA

concentration in fly-ash (Canadian Food Inspection Agency 2017). When considering all the elements based on their average concentration in the sampled fly-ash individually, Ni and Zn limit the amount of annual application to the field. According to soil and fly-ash testing results, only 7.27 Mg of fly-ash can be applied per hectare field annually. Once mixed with the other components (compost, manure, sludge or organic waste), which are low in the above elements (Ni and Zn), the application rate should be increased as the total metal concentrations in the compost material would be lower than that of the pure fly-ash.

Lime requirement and fly-ash recommendation

According to the soil analysis report from the Soil and Plant Laboratory, the lime application recommendations for the legume forage and mixed forage were 14.8 and 7.3 Mg ha^{-1}, respectively (Table 4). Results of soil tests in Canada's Atlantic region show that approximately 70% of the soils need 2 to 8 Mg of limestone per hectare to correct present soil pH to desired levels. In extreme cases, 16 Mg or even more may be required per hectare (Atlantic Soil Fertility Committee 1970). The rate of lime needed to bring about a desired soil pH change is determined by several factors, including; (a) the change in the pH required, (b) the buffer capacity of the soil, (c) the chemical composition of the liming material, and (d) fineness of the liming material. Legume forages need higher lime requirement than mixed forages because legume forages typically require a higher soil pH. For optimum production compared to other forage types, alfalfa, a legume, requires 6.6-7.0 pH, while clovers and birdsfoot trefoil can withstand slightly more acidic conditions (Turkington and Franko 1980).

According to the fly-ash analysis result and soil lime requirement, the fly-ash application rate was calculated using Eq. 2. This ash recommendation is based on the lime requirement and specific for the studied soil, fly-ash used and selected crop species. This application rate may vary with soil type and pH, the type of plant grown, and the quality of ash (CCE % and moisture %). Table 4 shows the lime requirement and the fly-ash recommendation for the studied soil. When fly-ash is used as a liming material, special attention must be given to soil sampling, the lime requirement test and application and incorporation of the fly-ash (Lickacz 2002). A thorough understanding of each factor is essential to achieve the maximum benefit from the use of fly-ash. With fly-ash, the moisture and purity need to be assessed when determining application rates.

According to the CFIA standards, only part of the fly-ash requirement can be substituted by CBPPL fly-ash, 20% and 40.7% for legume forage and mixed forage, respectively. The total lime requirement can be met when fly-ash is mixed with and applied with other soil amendments low in trace element concentration (example: Agricultural lime, compost, organic waste and sludge). Some agricultural soils may have a low lime requirement or ash requirement, depending on their initial pH and the desired pH. The desired pH will be determined by the crop, which will be grown in the field. Some crops need slightly acidic conditions, where CBPPL fly-ash can substitute most of the lime requirement.

Table 3. CFIA Fertilizer and Supplement Metals Standards and Examples of Maximum Acceptable Metal Concentrations Based on Annual Application Rates ND = Not detected (Canadian Food Inspection Agency 2017), average metal concentrations in the composite fly-ash sample from CBPPL and the maximum allowable annual application rates.

Metal	Maximum acceptable cumulative metal additions to the soil over 45 years (kg ha^{-1})	Average metal concentration in fly-ash (mg kg^{-1})	Maximum allowable annual application of CBPPL fly-ash Mg $ha^{-1}year^{-1}$
As	15	4.91 ± 0.76	66.67
Cd	4	0.93 ± 0.14	88.89
Cr	210	129.84 ± 17.47	35.90
Co	30	19.22 ± 1.63	33.33
Cu	150	197.41 ± 10.76	16.84
Hg	1	0.21±0.11	111.11
Mo	4	5.6 ± 1.8	15.87
Ni	36	109.90 ± 18.41	7.27
Pb	100	12.67 ± 0.8	170.94
Se	2.8	ND	-
Tl	1	ND	-
V	130	58.0 ± 4.5	49.81
Zn	370	1061.63 ± 89.28	7.97

At present, the use of wood ash as a soil amendment is more common in agricultural soils than forest soils in Canada. Over the last few decades, however, several research trials have been established in British Columbia, Saskatchewan, Manitoba, Ontario and Quebec provinces to

Table 4. Lime requirement and ash recommendation for the studied soil (Soil and Plant Laboratory).

Crop to be grown	Lime requirement (Mg ha^{-1})	Ash recommendation (Mg ha-1)	Allowable ash application (Mg ha^{-1}) (Based on CFIA standards)	Allowable ash application (%) (Based on CFIA standards)
Legume forage	14.8	36.25	7.27	20
Mixed forage	7.3	17.88	7.27	40.7

examine the effects of wood ash applications on forest soil's physicochemical properties, soil biodiversity, vegetation communities, tree growth and water quality. Table 5 shows the use of wood ash as a soil amendment in Canadian provinces and territories. The application of fly-ash as a soil amendment is administrated by the Environmental Protection Act (EPA), including the Environmental Assessment Regulations under the Environmental Protection Act of Newfoundland (NL). To date, no specific guidance has been developed for fly-ash applications on forest or agricultural soils in the province (Hannam et al. 2016). Although soil applications of fly-ash are not specifically mentioned in the EPA, likely, any large-scale use of fly-ash as a soil amendment in NL would require an Environmental Assessment (Hannam et al. 2016).

CONCLUSIONS

Using fly-ash as a soil amendment and liming material can increase soil pH. The metal concentrations in the CBPPL fly-ash are lower than those in the CCME compost categories B guidelines so that the fly-ash could potentially be used in the category B compost. The concentration of Ni, Zn and Mo in fly-ash were slightly higher than those in the CCME compost A guidelines. If the fly-ash can be mixed with the other soil amendments low in these elements before land application, then the final product may pass as a category A compost. According to the CFIA standards, only part of the ash requirement can be substituted by the CBPPL fly-ash. The percentage may vary depending on soil's initial soil pH and desired pH and which crop is grown. Beyond agricultural application, maximum allowable ash can be applied annually to the forest and marginal lands to improve their fertility and productivity.

Table 5. Use of wood ash as a soil amendment in Canadian province and territories (Hannam et al. 2016).

Province	Ash used as a soil amendment	Common uses of applies	
		Purpose	Soil type
Alberta	Yes	Liming	Agriculture
British Columbia	Yes	Liming	Agriculture/ Forestry
Manitoba	No	-	-
New Brunswick	Yes	Fertilizing /Liming	Agriculture
Newfoundland & Labrador	No	-	-
Nova Scotia	Yes	-	Agriculture
Northwest Territory	No	-	-
Nunavut	No	-	-
Ontario	No	-	-
Prince Edward Island	No	-	-
Quebec	Yes	Liming	Agriculture/ Forestry
Saskatchewan	No	-	-
Yukon Territory	No	-	-

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