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**Growth and foliar nutrition of a hybrid poplar clone following the
application of a mixture of papermill biosolids and lime mud**

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Abstract

Fertilization of hybrid poplar (HP) plantations with papermill by-products is a promising solution to improve soil fertility and nutrient availability, increase plantation productivity, and provide added value to these materials that would otherwise be incinerated or sent to the landfill. We assessed the growth and foliar nutrition of a HP clone (*Populus x. canadensis* × *P. maximowiczii*) at six plantation sites aged three to five years in southern Quebec, Canada. Sites received a fertilization treatment consisting of a mixture of papermill biosolids (120 to 140 t ha⁻¹, depending on site) and lime mud (10 to 15 t ha⁻¹) before being planted, or no fertilization (control). Tree growth was significantly improved by fertilization, with fertilized trees showing a mean annual height increment of 1.3 m (all-site mean; SD = 0.2), compared with 0.5 m (SD = 0.4) for unfertilized trees. Foliar Ca and Mg increased following fertilization and levels met optimal thresholds at all sites, whereas N, P, and K concentrations also increased but nutritional deficiencies remained for these elements at several sites. Our results confirm the benefits of fertilizing hybrid poplars with papermill by-products, but indicate that adjustments in application rates or type of by-products could be made in order to fully satisfy nutritional requirements and thus optimize tree growth.

Keywords: fertilization; foliar nutrition; forest soil; hybrid poplar; papermill by-products.

42 **Introduction**

43 Intensive management of tree plantations can help meet the increasing demand for
44 wood products while increasing carbon sequestration, conserving larger areas of forests
45 and restoring degraded lands (Paquette and Messier 2010). However, fertilization of
46 productive plantations is often required for maintaining soil nutrient availability and
47 biomass yields over several rotations (Fox 2000). Successful fertilization programs must
48 rely on indicators for optimal nutrition (species- or clone-specific), repeated assessments
49 of the nutritional status of trees (Mead 2005) and optimization of fertilizer applications (i.e.
50 type of fertilizer, application rate, timing and frequency of application) for maximum tree
51 growth (Elferjani et al. 2013; Albaugh et al. 2018).

52 The benefits of fertilization with inorganic fertilizers have been demonstrated for a
53 wide range of environmental conditions and tree species around the world (Prietz et al.
54 2004, Fox et al. 2007, Saarsalmi et al. 2014). However, the higher demand for fertilizers
55 created by increasing areas of planted forests worldwide (FAO 2015, Payn et al. 2015)
56 could be satisfied in part by industrial by-products such as biosolids and alkaline residues
57 (e.g. lime mud/slurry and wood ash). Their recycling in tree plantations would add value
58 to what is otherwise a waste product and would decrease the high carbon footprint
59 associated with the production and use of commercial fertilizers (Chojnacka et al. 2019).
60 The development of a circular economy may become a necessary alternative to more
61 traditional disposal solutions of these by-products such as landfill and incineration (Pervaiz
62 and Sain 2015, Collivignarelli et al. 2019).

63 Municipal biosolids are commonly used in agriculture to improve soil physical,
64 chemical and biological properties, and in turn increase crop/forage growth (Lu et al.

2012). They are also successfully used for soil reconstruction and ecological restoration of decommissioned mine sites (Antonelli et al. 2018, Carabassa et al. 2020). However, the recycling of municipal biosolids is not a common practice in forestry despite that the scientific literature suggests these materials can significantly improve soil quality (e.g., lower C:N ratio and higher N mineralization) and increase tree growth (e.g. Mandre et al. 2010, Nieminen and Riäsanän 2013, Ingerslev et al. 2014, Sevel et al. 2014, Ouimet et al. 2015). Interest in municipal biosolids remains limited among forest managers likely due to environmental concerns associated with their use (e.g. nutrient runoff and eutrophication of surface waters, contamination of heavy metals and impacts on plants and soil microfauna; Su et al. 2008, Ring et al. 2013, Waterhouse et al. 2014), as well as public acceptance and relatively rigid legislations constraining their use in forested landscapes (Iranpour et al. 2004).

The use of papermill biosolids could be an attractive soil amendment alternative in agriculture and forestry. They are rich in Ca, also contain N, P and K (but in lesser amount than municipal biosolids), and due to a high fiber content they can augment soil structure, organic carbon and cation exchange capacity (Chantigny et al. 1999; Camberato et al. 2006, Gagnon and Ziadi 2020, Zhang et al. 2020). Papermill biosolids potentially minimize environmental risks compared to municipal biosolids because they contain less heavy metals than municipal biosolids and do not lead to critical levels in plants after they are applied on agricultural fields (Gagnon et al. 2013). Also, their generally high fiber content is expected to immobilize some N, thus resembling more a slow N-release inorganic fertilizer with less risk for leaching to surface waters (Lteif et al. 2007, Gagnon and Ziadi 2020). A few research trials have shown benefits of papermill biosolids on the yields of

88 agricultural crops (Price and Voroney 2007, Gagnon and Ziadi 2012, Ziadi et al. 2013).
89 Some studies have also demonstrated the positive effects of papermill biosolids on forest
90 soils (e.g. carbon, pH and calcium) and growth of trees in plantation settings (Brockway
91 and Urie 1983, Lteif et al. 2007, Ortega Rodriguez et al. 2018) and in natural stands
92 (Feldkirchner et al. 2003).

93 Hybrid poplars (HP) are some of the most prominent fast-growing trees used in
94 intensive silviculture worldwide. They can establish and grow on an array of soils, but HP
95 grow best on sites with higher soil pH and nutrient availability (Camiré and Brazeau 1998,
96 Stettler et al. 1996), thus their yields are generally improved following fertilization
97 (Stanton et al. 2002, Christersson 2006, and González-García et al. 2014). However, only
98 a handful of studies tested the effects of industrial by-products on HP growth in North
99 America (Young et al. 1993, Cavaleri et al. 2004, Lteif et al. 2007, Carey 2010, Lafleur et
100 al. 2012, de Oliveira Rabel et al. 2021). To our knowledge, only Lteif et al. (2007) studied
101 the effects of papermill biosolids on hybrid poplar. Although fertilization of forest soils
102 and trees with wood ash has been largely studied and demonstrated (see review by Reid
103 and Watmough 2014), there has been very few studies investigating the response of HP
104 plantations to wood ash (Cavaleri et al. 2004, Ventura et al. 2019). The effects of wood ash
105 and lime mud on soils and crops have also been studied in combination with papermill
106 biosolids in an agricultural context (Muse and Mitchell, He et al. 2009, Gagnon and Ziadi
107 210, Ziadi et al. 2013). Lime mud (CaCO_3) is produced during the transformation of wood
108 chips into paper pulp. Wood chips are cooked with NaOH in order to extract the fibers
109 from the lignin. In the process, NaOH is transformed into Na_2CO_3 , which, through the
110 addition of CaO (quicklime), is converted back to NaOH for its reuse, and in the end CaCO_3

is formed. This lime mud has a calcium carbonate equivalent (CCE) varying between 90 and 100 % and thus a much higher effective neutralizing value than wood ash and papermill biosolids (see next section). Lime mud also contains traces of metal toxicants. In this respect, a positive outcome of mixing papermill biosolids and lime mud is expected in forest settings.

We studied soil nutrient availability, foliar nutrient status and growth of HP plantations following the application of a mixture of papermill biosolids and lime mud at six forest sites in southern Quebec, Canada. Due to the nutrient-poor and acidic nature of the soils, we expected that soil nutrient availability, notably pH and calcium, and foliar nutrition of HP would be improved following application and, in turn, that HP growth would increase as well. We anticipated the study would demonstrate that productive HP plantations growing on nutrient-poor and acidic forest soils are possible when appropriate nutrient amendments are made using by-products of the papermill industry.

Materials and Methods

Study site and experimental design

The study was conducted at six different plantation sites (Dudswell, DUD; Lingwick, LIG; Lac Louise, LL; Saint-Francois-Xavier, SFX; Stokes, STK; Weedon, WED) in the Eastern Townships region of southern Quebec, Canada (Table 1, Figure 1). The region has a warm-summer humid continental climate (Dfb in the Köppen-Geiger classification; Kottek et al. 2006) and is found within the sugar maple-basswood or the sugar maple-yellow birch forest vegetation zones (Saucier et al. 2011). Forest species composition of the mature stands present at the sites prior to harvesting and plantation

establishment comprised red maple (*Acer rubrum*), trembling aspen (*Populus tremuloides*) and balsam fir (*Abies balsamifera*). During the 1981-2010 reference period, mean summer (May-August) rainfall varied between 467 and 504 mm, depending on the site (Table 1), while mean annual rainfall ranged from 640 to 966 mm, with an additional 207 to 356 cm of snow (Meteorological Service of Canada 2016). Mean summer temperatures were between 14.7 and 16.8 °C, and mean annual temperatures were between 3.8 and 5.6 °C. Topography is relatively flat at all sites. Soils are moderately well to well drained and usually consist of loamy sand classified as Orthic Ferro-Humic Podzols (Soil Classification Working Group 1998).

Sites were clear-felled in the summer prior to plantation establishment (referred to as year 0). The treatments were established in 1- to 4-ha blocks. Each block was separated into two parts, one being a control treatment (no fertilization) and the other receiving the papermill biosolids and lime mud. Each site had one to three blocks (Table 1). The separation of the blocks into a control area and a fertilized area was due to logistical constraints of mechanical spreading in a forestry context. However, it was possible to randomly assign the treatment areas within each block. The WED site was an exception in having only one of its three blocks with a control treatment.

Papermill biosolids were applied in the fall of year 0 at a rate varying between 120 and 140 t ha⁻¹, depending on the site, in the designated area of the block. The material was applied using a mechanical spreader equipped with a design of paired propellers that evenly apply the material on the soil surface. At the same period and using the same spreader, lime mud was applied on the soil surface at a rate varying from 10 to 15 t ha⁻¹, depending on the site (Table 1). Site preparation by mounding (Sutton 1993, Bilodeau-Gauthier et al. 2011)

was done shortly after fertilization, both in control and in treated areas. In the latter, this meant that some of the applied by-products were covered by the upturned mineral soil of the mounds.

Chemical properties of papermill biosolids and lime mud used in the present study are shown in Table 2. Moisture was assessed by measuring mass before and after drying in an oven at 65°C for 72 hours. Density was measured using a water displacement method on dry material after they were sieved through a 2 mm mesh. pH was measured in water using a 1 to 5 soil to water ratio of the dry/sieved samples, whereas neutralizing capacity was assessed from the digestion of the material in diluted HNO₃ (pH 2) and back-titration to excess acidity to pH 8.3. Carbon and N were measured on finely ground (< 60 µm) subsamples by combustion at high temperature (1040°C) and infrared (C) and thermal conductivity (N) detection using a CHNS-O elemental analyzer (model EA1108, Thermo Fisons, Waltham, MA, USA), whereas loss on ignition was measured from dry/ground samples after combustion in a muffle furnace at 500°C for 5 h. Finally, total chemical composition was measured on dry/ground samples after they were fused unto 32 mm diameter beads using a 1 to 4 sample to lithium tetra(meta)borate mixture heated at 1000°C for 18 minutes. An automated X-ray fluorescence Philips PW2440 spectrometer system with a Rhodium 60 kV end window X-ray source, operating at 3 kW, was used for analysis of the beads. These characteristics are similar to those reported in the literature (cf. Lteif et al. 2007, Kuokkanen et al. 2008, Pöykiö et al. 2008). Chemical properties of the by-products were also compared to those of upper podzolic B horizons prior to fertilization at the SFX site, which is representative of the soils in the region (Table 2).

Planting was done in the following spring (year 1) at a density of 900 trees ha⁻¹ using 1.0 to 1.5 m-long bare-rooted cuttings planted on mounds at a depth of 30 cm (thus reaching the covered by-products). The DNxM-915508 clone was used in the study. It is a combination of *Populus x. canadensis* (a hybrid of *P. deltoides* × *P. nigra*) with *P. maximowiczii*, and is produced in nurseries by the *Ministère des forêts, de la faune et des parcs du Québec* of the Quebec Government. This clone is recommended for planting in southern Quebec and has been commonly used by the forest industry and small landowners for several years.

Two to ten sampling plots of 200 m² (or 7.97 m radius) were then established in each block, half being control plots and half being fertilized plots. The plots encompassed 15 to 20 trees. Plots were positioned randomly but at least 20 m from the edge of their treatment area and at least 50 m from the edges of other plots. Because of relatively flat terrains and the substantial distance between plots, we assumed there was no transfer of papermill by-products by wind or water to the control plots.

Tree measurements

Five to 10 trees were randomly selected in each plot for further inquiries, for a total of 238 trees across the six sites. Depending on site, trees were 3 to 5 years of age (i.e. time since planting, not counting the 2 years in nursery) when their growth was assessed in the fall of 2014 or 2015. Height was measured with a telescopic measuring pole, whereas diameter at breast height (1.3 m, DBH) was measured with a manual calliper. Given the homogeneous architecture of the DNxM-915508 clone and how annual whorls of branches are formed, it was possible to also measure the height of past years, including the initial height at planting. Yet, this was done only for the sites measured in 2015 (LIG, LL and

STK). At the three sites sampled in 2014 (DUD, SFX and WED), only current total height was recorded, but not initial height. Thus, trees from the 2014 sites were attributed an initial height based on the mean (1.44 m; standard deviation = 0.3) from the 2015 sites. In order to standardize the data across sites that differed in age, the mean annual height increment (MAI) was used in the analyses and was calculated as such:

$$\text{mean annual height increment}_{yrX} = \frac{\text{total height}_{yrX} - \text{initial height}}{\text{age}_{yrX}}$$

where yrX is the year at which the measurements were done. Note that this MAI equation is essentially the same as the conventional MAI equation with the exception that it does not include the initial age term because it would be redundant in our case (i.e. initial age = 0).

Diameter at breast height was standardized by dividing it by tree age. Because initial DBH at planting is unknown, this is not an annual growth increment *per se*, but rather an ‘age-standardized DBH’. Assuming that the initial DBH was zero would have otherwise resulted in biased annual increments

In addition, at the SFX site, tree height and DBH were measured in the field every year. For the years 1 through 4, 15 to 20 trees were selected per plot, and in year 5, five trees were selected per plot. The mean annual height increment for this site was calculated using data from year 5 and the equation above.

Foliage sampling and analyses

Foliage was usually sampled during the same year that height and DBH were measured, with the exception of two sites (i.e. LIG and SFX) where it was done the previous year (Table 1). Foliage was sampled during the last two weeks of July from the same trees on which growth was measured. Ten leaves per tree were collected from random

branches in the upper third of the crown (full sunlight) and were pooled to obtain one sample per tree.

Leaves were dried at 60°C for 48 h and then finely ground using a ball mill (Mixer Mill MM 200, Retsch Inc., Newtown, PA, USA) for chemical analyses. Carbon and N were analyzed from finely ground (< 60 µm) subsamples using the EA1108 elemental analyzer as described above for papermill by-products. Calcium, Mg, K, Al and P were analyzed from digestions in concentrated HNO₃ in PTFE beakers for five hours at 120°C. A PTFE watch glass allowed a reflux of the digestate while permitting excess vapor to escape. Phosphorus was measured by colorimetry (molybdenum blue) on a continuous-flow automated analyzer (Technicon Instruments Corp., Tarrytown, NY, USA), while Ca, Mg and K (base cations) and Al were measured by inductively-coupled plasma mass spectroscopy (ICP-MS; model 820, Varian, Palo Alto, CA, USA).

Foliar nutrition was analyzed through an approach called critical value assessment (CVA; Bates 1971, van den Driessche 1974) which compares measured foliar nutrient concentrations to species-specific values representing nutritional levels for minimal or optimal growth. To this end, CVA values computed from a literature review of several *Populus* species (Camiré and Brazeau 1998) were used as they highlighted minimal and optimal nutritional requirements.

Soil sampling and analyses

Soil samples were collected at the same time as foliage, except for the SFX site where it was done the following year (Table 1). Using a soil auger, soils were sampled in the first 20 cm of mound and at the base of HP trees. This section of the mound corresponded to the podzolic B horizon. Depending on the site, three to nine soil samples

per treatment per site were collected, with the exception of the WED site where an uneven number of samples was collected between the two treatments (Table 1). As a whole, we collected a total of 60 soil samples, i.e. 28 in control and 32 in fertilized.

Soil samples were dried at 40 °C to a constant weight and sieved through a 2 mm mesh. Subsamples were used to measure soil pH in a 1:2 soil:water mixture. Carbon and N were also measured on finely ground subsamples using the EA1108 elemental analyzer. Exchangeable cations were extracted using the Mehlich III protocol (Ziadi and Tran 2008). Calcium, Mg and K were measured by ICP-MS and P was measured by colorimetry using the same protocols/instruments as for foliage.

Statistical analyses

Linear mixed effects models were performed to test the effects of papermill by-products on mean annual height increments, age-standardized DBH, foliar elemental concentrations and soil chemical properties. We used the function ‘lmer’ from the R package ‘lme4’ (Bates et al. 2015) for these analyses. For growth and foliar nutrition, the tree was the basic sampling unit. There were some missing observations because 3 trees died within control plots and two others were too short to allow measuring DBH ($n = 238$). In the case of foliage, however, adjacent trees were selected to act as replacements for the aforementioned dead trees. Fixed effects included treatment (two levels: control and fertilized) and age (as a categorical variable: three levels). Site and plot were modelled as random effects with plot nested into site (Zuur et al. 2009), with probability levels resolved by Restricted Maximum Likelihood (REML, Searle et al. 1992; Wolfinger et al. 1994). Incorporating a hierarchical, or nested, random-effects structure in a mixed-effects model allows circumventing shortcomings of the repeated measures method (Kristensen and

271 Hansen 2004, Millar and Anderson 2004, Paterson and Lello 2003). This way, the variance
272 associated with each hierarchical level of the random-effects structure can be assessed via
273 the model output, and compared to the variance associated with the fixed effects (see the
274 tables of variance for the mixed models in the Results section). Degrees of freedom for
275 calculating *P*-values of linear mixed effect models were approximated after Kenward and
276 Roger (1997) using the R package ‘pbkrtest’ (Halekoh and Hojsgaard 2014) and were
277 obtained through type III F-tests using the ‘anova’ function from the ‘lmerTest’ package
278 (Kuznetsova et al. 2016).

279 In addition, relationships between tree growth and foliar nutrients were tested
280 through least squares linear regressions (via orthogonal factorization method or QR,
281 Becker et al. 1988), with height as the response variable, foliar nutrient concentration as
282 the fixed effect (one model per nutrient), and site as a random effect. Mean values per plot
283 were used to circumvent the variability associated with the different number of trees per
284 plot across sites ($n = 40$). Marginal R^2 (variance explained by the fixed effects) and
285 conditional R^2 (variance explained by both fixed and random effects) were calculated using
286 the methods described in Nakagawa and Schielzeth (2013) and Johnson (2014), which are
287 implemented in the R package ‘piecewiseSEM’ (Lefcheck 2015).

288 For all analyses, normality and homoscedasticity of residuals were verified visually
289 with, respectively, a quantile-quantile plot and a predicted vs residual values plot. All
290 statistical analyses were conducted on R 4.0.3 (R Core Team 2020). When warranted, data
291 were log-transformed in order to conform to normality or homoscedasticity assumptions
292 (soil Ca and Mg, and foliar Ca/Al). In one instance, a few outliers had to be removed (soil

K), while the 'age' effect had to be introduced as a continuous variable (soil P) in order for the model to converge properly.

Results

Tree growth

Hybrid poplar mean annual height increment range from as low as 23.8 cm in the control plots at the LIG site to as high as 154 cm in the fertilized plots at the LL site (Figure 2a, Table 3). The smallest age-standardized DBH was also in the control plots at the LIG site (0.4 cm), whereas the largest age-standardized DBH was in the fertilized plots at the LL site (2.1 cm, Figure 2b, Table 3).

Hybrid poplar mean annual height increment and age-standardized DBH were significantly increased by the application of papermill by-products (Tables 3, 4 and S1). There was an increase at all sites. The smallest (36.4 cm) and largest (90.4 cm) differences in annual height increment between the control and fertilized plots were at the SFX and LL sites, respectively. The smallest (0.5 cm) and largest (1.5 cm) differences in age-standardized DBH between the control and fertilized plots were at the SFX and DUD sites, respectively.

Fertilization with papermill by-products also homogenized hybrid poplar growth between sites. Without fertilization, mean annual height increment ranged from about 30 cm to almost 120 cm across the sites, while it ranged from about 100 cm to 154 cm with fertilization. Variation and homogenization of the data was similar for age-standardized DBH, which ranged between 0.4 cm and 1.4 cm under control conditions and between 1.3 cm and 2.1 cm under fertilization.

Fertilization with the papermill by-products explained 19.0 and 24.1 % of the variability in DBH and height growth variables, respectively, which was the highest percentage for a single fixed or random effect in these models (Table S1). Site, age and plot generally explained only a small amount of the variation in annual height increment and age-standardized DBH, i.e. about 14 % for the three factors together, with plot making the most of that value (9.7 % for DBH and 12.8 % for height). The residuals amounted to about two-thirds of the total variance (62 % for height and 66 % for DBH).

At the SFX site where we measured height and DBH growth annually from years 1 to 5, HP trees receiving papermill by-products exhibited higher growth starting in year 1 (Figure 3). However, due to the variability in the data and the small relative magnitude of the gain from fertilization in early years, significant differences in total height and DBH between fertilized and control plots were detected only for years 4 and 5.

Foliar nutrition

Foliar N, P, Ca and K concentrations in the control plots were generally below the minimal requirements, whereas foliar Mg concentrations were all on or above the minimal requirement (Figure 5). Application of papermill by-products favored an increase above the minimal Ca requirement for hybrid poplar at all six sites, whereas it helped meet N requirement at three sites (DUD, LL and WED), K requirement at three sites (DUD, LL and WED) as well as P requirement at one site (LL). It also largely decreased foliar Al at four sites (DUD, LIG, LL and STK; Figure 4). All sites exhibited foliar Al concentrations below 0.02 mg g⁻¹ in the fertilized plots, with the exception of the LIG site (0.05 mg g⁻¹).

Fertilization with the papermill by-products significantly increased foliar N, P, Ca and Mg and significantly decreased foliar Al concentrations (Tables 4 and S2). However,

fertilization had no significant effect on foliar K concentrations. Age did not significantly affect the foliar concentrations of the studied elements. The proportion of the variance explained by fertilization was highest for foliar Ca (26.7 %) and Al (16.8 %), with lower values for the other elements (1.0 to 6.4 %; Table S2). Plot was the second factor explaining the most variation in foliar concentrations for most elements (8.8 to 15.2 %), except for foliar Mg where site explained slightly more (19.6 %). The residuals amounted to almost two-thirds of the total variance in foliar K, Ca, Mg and Al (58.2 to 67.8 %), and over three-quarters of the total variance in foliar N and P (80 to 92 %).

Linear mixed models revealed significant relationships between hybrid poplar height and foliar N, P, Ca, Mg and Al concentrations as well as N/P and Ca/Al ratio (Figure 5, Table S3). Except for foliar Mg, these models were all very robust, with conditional R^2 ranging from 0.59 (Al) to 0.72 (N) and explaining between 42.7 % (Al) and 57.1 % (Ca) of the variation in tree height. Foliar Mg, in turn, explained 18.8 % of the variation in tree height (conditional $R^2 = 0.46$). Site, which was used as a random effect, explained only 0.7 % to 7.8 % of the variation in the height of hybrid poplar trees. There was no relationship between tree height and foliar K (Figure 5, Table S3).

Soil chemical composition

Soil pH and exchangeable K, Ca and Mg concentrations were significantly increased following the application of papermill by-products, whereas soil exchangeable Al concentrations was significantly decreased (Figure 6, Table S4). Fertilization and age did not have a significant effect on soil total N or exchangeable P concentrations (Figure 6, Tables 4 and S4). The divergence in soil pH, Ca and Al between the control and fertilized plots was apparent at all sites except for SFX and STK. Mean soil pH was consistently near

6 or above at all sites except STK, whereas all sites except SFX had a pH near or below 5 in the control plots. In regard to soil K, differences were observed at all sites except for DUD and SFX, whereas the LIG and WED sites were the ones which responded the most to fertilization in regard to soil Mg (Figure 6).

Fertilization with the papermill by-products significantly explained soil K, Ca, Mg, Al, as well as soil pH (Table S4). The proportion of the variation explained by fertilization was highest in the case of soil Ca (45.4 %), Mg (16.1 %) and pH (20.5 %), and soil lower for K (12.2 %) and Al (8.3 %). Age explained an additional 18.3 % of the variation in soil exchangeable K concentrations, whereas site explained an additional 10.4 %, 45.3 % and 25.3 % of the variation in soil pH, exchangeable Mg and Al concentrations, respectively.

Discussion

Fertilization effects on tree growth

Our results suggest that the HP clone 915508 responded positively to the application of a mixture of papermill biosolids and lime mud. Depending on the growth variable and site, a 1.3- to 4.2-fold increase was observed. Despite the slightly unbalanced experimental design in terms of number of replications per site and differences in stand age between sites, we detected no significant effect of age, site and plot on trees (growth and foliar nutrition) or soil chemical composition. The SFX site exhibited the highest growth of all sites under control conditions and it appeared to respond the least to fertilization in terms of growth. In fact, considering all six sites, there seemed to be a clear trend in the size of the growth response to fertilization relative to the inherent growth (control plots). Specifically, the three sites with the lowest inherent height growth (DUD, LIG and STK)

responded more positively to fertilization (gains of ~ 300 % in height) than the three sites with the highest inherent height growth (LL, SFX and WED; gains of ~ 30 to 180 %).

The use of papermill by-products reduced the variability in both annual height increment and age-standardized DBH between the sites. Within-site variability in height growth was also reduced following fertilization. For five sites, we found that the coefficient of variation in mean annual height increment was lower in fertilized plots compared with the control plots. Less variability in growth between and within the sites can be beneficial for the management of these plantations, allowing for more consistent yields and more accurate harvest projections.

The gains in height growth from fertilization reported in our study are comparable to gains found in the literature for HP receiving mineral (commercial) amendments, although the responses at the DUD, LIG and STK sites are at the higher end of the response gradient (cf. Brown and van den Driessche 2002, with 21% gains, and Coyle and Coleman 2005, with 200% gains). In a study of HP plantations at higher latitudes in the southern boreal forest of Quebec, but on similar podzolic soils and following the same mechanical site preparation (mounding), Bilodeau-Gauthier et al. (2011) found that fertilizing hybrid poplars with granulated N and P fertilizers increased height growth by 30% in the year following fertilization.

Hybrid poplar plantations in Larchevêque et al. (2011) were younger (i.e. 2 years), which could explain the small, or lack of, effect of sheep manure on tree height. However, the increase in diameter was considerable (21% gain) with the largest application rate. Larchevêque et al. (2011) also tested mineral amendments and concluded that they were more effective than sheep manure to favor HP growth, possibly because mineral fertilizers

released nutrients faster. Lafleur et al. (2012) tested the effects of hog manure on HP growth and concluded a maximum increase of only 13 %. In both studies, growth was significantly less than at our sites. However, studying the response of HP to fertilization with municipal biosolids, Carey (2010) reported a large increase (83%) in height growth after a single growing season. Moreover, Lteif et al. (2007) tested various mixtures of pig slurry and paper biosolids on HP growth and reported gains between 56 % and 287 % in stem biomass. Similarly, in a short-term greenhouse study by Cavaleri et al. (2004), HP cuttings grown in pots filled with forest soil and fertilized with a combination of wood ash and municipal biosolids exhibited a 213 % gain in growth relative to control pots. We believe that the large and prolonged responses at our study site are due to the fact that HP plantations are grown in a forestry context on poor soils, whereas many of the studies investigating the effects of fertilization on HP growth were conducted in an agricultural settings where soils are generally more productive and are associated to a history of fertilization.

At the SFX site, the benefits of fertilization were statistically significant only in years 4 and 5. First, the variability in the data and the small relative magnitude of the gain in years 1 to 3 likely prevented detecting significant differences between the fertilized and control plots. Second, Henry (1991) proposed a slow mineralization of nutrients, leading to a delayed uptake by the trees. Papermill biosolids may thus not result in an immediate response in tree growth, but it would be expected to have a more lasting effect than mineral amendments. In this respect, our results at the SFX site suggest a Type 2 response, meaning that the difference between fertilized and control trees increased over time (Mead 2005).

Fertilization effects on soil chemistry and foliar nutrition

Lime mud was mixed with paper biosolids and in turn, its effects on soil chemistry cannot be directly observed. However, due to its high neutralizing capacity (91%), lime mud (CaCO_3) is expected to have a significant positive effect on the soil acid-base status by increasing pH and exchangeable Ca concentrations and lowering exchangeable Al concentrations. During its rapid dissolution, lime provides Ca^{2+} in the soil solution, which displaces acidity (Al^{3+} and H^+) adsorbed on soil exchange surfaces (Havlin et al. 2005). Hydroxyl ions produced during lime dissolution then react with Al^{3+} and H^+ to form $\text{Al}(\text{OH})_3$ precipitates and water (autoprotolysis), respectively. The improved soil acid-base status had a positive effect on foliar nutrition of HP by increasing Ca concentrations above the minimal requirement for HP (Camiré and Brazeau 1998) and largely decreasing Al concentrations at some site where it was intrinsically high (e.g. DUD, LIG and STK). Foliar Ca, Al and Ca/Al were the most robust predictor variables to model HP height growth across the six study sites. Higher foliar Ca levels and Ca/Al ratio and lower Al levels led to greater HP productivity. Higher Ca availability following fertilization of acidic forest soils was shown to lead to increased HP growth (Bilodeau-Gauthier et al. 2011; Elferjani et al. 2013), whereas low Ca/Al ratio are associated with an increased probability for detrimental effects on tree growth (Cronan and Grigal 1995). Similarly, the alkalinizing effect of lime mud used in our study was beneficial for HP nutrition and growth.

The application of papermill biosolids and lime mud also increased soil exchangeable Mg and K concentrations, but not N and P. Conversely, foliar concentrations of Mg, N and P were significantly increased following the application of papermill by-products, but not K. Foliar Mg concentrations were on or above the minimal requirement (Camiré and Brazeau 1998), whether plots were fertilized or not. This is likely due to the

influence of serpentine rocks $[(\text{Mg,Fe,Ni})_3\text{Si}_2\text{O}_5(\text{OH})_4]$ on the soil chemical signature in the area (Rajakaruna and Harris 2009). At the SFX site, fertilization resulted in a decrease in foliar Mg, probably due to Ca antagonism over Mg in the soil and in tree leaves and roots (Diem and Godbold 1993, Loide 2004). These two elements compete for soil exchange sites and for root uptake pathways (Jakobsen 1993, Fageria 2001). For example, McLaren et al. (2007) observed an increase in soil Ca following application of municipal biosolids in a pine plantation, but the increased dominance of Ca over Mg on soil exchange surfaces favored leaching of Mg. Soil and foliar Ca and Mg levels at the SFX sites were higher in control plots than at any other site; in turn, an increase in the activity of Ca in the soil may have more readily affected soil Mg availability and uptake by tree roots.

There was a positive effect of fertilization on foliar K concentrations at the DUD, LL, SFX and WED sites, but a substantial decrease at the LIG site and no change at the STK site. Foliar K concentrations also remained below the minimal requirement for most sites (Camiré and Brazeau 1998). There are cases of uptake interferences of K due to a dominance in N (e.g. Kelty et al. 2004) and Ca (e.g. Arnold and Diest 1993). We suspect the interference of Ca to be the main mechanisms at play at our sites. Potassium is not strongly adsorbed on soil exchange surfaces compared to Ca and thus can be leached to greater depths following liming. For example, Arnold and Diest (1993) found foliar K concentrations in *Pinus sylvestris* to increase following fertilization with chemical fertilizers, but this increase was more limited when chemical fertilizers were mixed with lime. Thus, the decrease in foliar K concentrations at the LIG site could be due to a similar mechanism. Antagonistic interactions of Mg with other nutrients due to an excess in Mg can also lead to deficiencies (Venkatesan and Jayaganesh 2010). In this respect, we also

believe that the serpentinic soils, rich in Mg, also combine with lime and high Ca activity to interfere with K uptake in the studied HP plantations.

Foliar P concentrations in control plots were well below the minimal nutritional requirements (Camiré and Brazeau 1998). This is because at low soil pH such as the ones at the study sites, P availability to plant roots is limited due to adsorption of PO_4^{3-} onto active surfaces of Fe and Al sesquioxides and/or precipitation of PO_4^{3-} within these mineral structures (Cade-Menun et al. 2000, Devau et al. 2009). Phosphorus mobility and availability are thus at a maximum at soil pH values varying between 6 and 7 (Tinker and Nye 2000, Havlin et al. 2005). Papermill biosolids and lime mud are only small sources of P (0.47 % and 0.60 %, respectively). The overall increase in foliar P levels at the study sites is thus believed to be due to the combination of adding a small source of P to the soil, through both by-products, and adjusting its pH, via lime mud, to levels closer to 6 or above. With regard to N, its content in papermill biosolids is about 1.2 %. On the one hand, the added N can be (1) immobilized in organic matter or by microorganisms in N-poor and acidic forest soils (Henry 1991, Cabral et al. 1998) and/or (2) captured by the fibers of the papermill biosolids (Lteif et al. 2007, Gagnon and Ziadi 2020). However, the C/N ratio of the papermill biosolids used in our study is approximately 18, which is conducive to some N release. Microbial activity releasing more N is also expected at the new pH values adjusted with the lime mud (Binkley and Fisher, 2013; Brais et al., 2015). In this respect, the relatively small (but consistent) increase in foliar N following papermill biosolids, i.e. close to the minimal N requirement, likely reflects material properties that resemble those of a slow release fertilizer. Hybrid poplar growth in Bilodeau-Gauthier et al. (2011) has

responded earlier (1-2 years) than in the current study (4 years) likely due to a faster release of nutrients from granulated fertilizers compared to papermill biosolids.

Foliar N/P ratio ranged from 8 to 25, depending on sites/plots and treatments. According to Güsewell (2004), a N/P ratio from 10 to 20 is the optimal balance between the two nutrients for terrestrial plants, with values under 10 and over 20 representing N- and P-limitations, respectively. This is apparently true for short-term fertilization experiments. All N/P ratio above 20 in our study were control plots, whereas all N/P ratio under 10 were fertilized plots. Based on this premise, applying papermill by-products at the proposed rates in the studied HP plantations is associated to a small risk of creating limitations in P, whereas not proceeding to any fertilization exposes the trees to possible N limitations. Considering that the overall effect of fertilization was to bring foliar N and P concentrations closer to their minimal nutritional requirements, we argue that (1) the application of papermill biosolids and lime mud at respective rates of 120-140 and 10-15 t ha⁻¹ is a reliable fertilization strategy for HP plantations growing on acidic forest soils, and that (2) imbalances between N and P are a more marginal concern because both nutrients in HP foliage are generally below or well below the minimal requirements and because the immediate objective is to raise their levels.

Management implications

Because our results show that P availability, and to a lesser extent N and K availabilities, were not optimal for HP trees, we suggest that the use of a fertilization by-product with higher nutrient concentrations than papermill biosolids could be considered as a means to further improving foliar nutrition and thus completely mitigating deficiencies. One option would be to use municipal biosolids, which can reach N and P

concentrations as high as 6 % and 3 %, respectively (McLaughlin 1984, CCME 2010), although their application requires extra care that could make them impractical. Increasing nutrient supply to roots could also involve more thorough weed control (Pinno et al. 2010) and a fertilization schedule that better distributes the nutrients over time.

In the latter, the idea would be to supply trees with nutrients at the right time and amounts, e.g. supplying extra amounts of P in the early years to boost root production. For example, instead of applying the full dose of biosolids at the onset of the plantation, it could be split to allow for a second application at a smaller rate three to four years following tree establishment, i.e. when trees have an increasing nutrient demand but are still small enough for the spreader to circulate between tree rows without physically damaging their branches. We believe this more targeted fertilization strategy would better fulfill N and P demands of the larger trees while attenuating leaching to surface and ground waters due to a tighter nutrient cycling provided by the trees and the well-established understory vegetation. As a whole, a first fertilization with papermill by-products would provide an initial alkalinization of the soils along with a slow release fertilizer, whereas a second (delayed) fertilization would provide a rapid access to nutrients limiting HP growth a few years after plantation establishment. The control of understory competition could be conducted before the second application to render more nutrients available to HP trees, if needed.

Conclusion

Growth of the DNxM-915508 HP clone planted on nutrient-poor and acidic forest soils in southern Quebec was significantly increased following fertilization with papermill biosolids and lime mud. The benefits of applying a mixture of papermill biosolids at a rate

varying between 120 and 140 t ha⁻¹ and lime mud at a rate between 10 and 15 t ha⁻¹ on HP growth were associated with an overall improvement of the soil acid-base status and of foliar nutrition. Notably, Ca and Mg levels in foliage increased to optimal thresholds at all sites. On average, mean annual height increment was 1.3 m for fertilized plots compared to 0.5 m for control plots. However, despite increases in foliar levels, our results suggest that HP growth was not optimized because of remaining nutrient deficiencies, mainly in P but in N and K as well, following fertilization with papermill by-products. Although adjustments in application rates or type of by-products could be made to optimize HP nutrition and growth, recycling of papermill by-products has great potential for intensive plantation forestry and augmenting C sequestration in forests. Given that HP trees are ultimately intended to provide pulp for the paper industry, fertilization of HP plantations with industrial by-products is a good example of circular economy. Further research could compare how various clones planted on various soils respond to these by-products.

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Figure 1. Location of the six plantation sites in southern Quebec, Canada. Map created by the authors with ArcMap 10.2.2 (Esri 2014) using open-source shapefiles from the Government of Quebec (2021) for water bodies and from StatSilk (2021) for political boundaries.

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856 plots (black dots) across the six plantations. Vertical dashed lines in (g) represent lower
857 and upper thresholds of an adequate balance between N and P (Güsewell 2004).
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859 following Nakagawa and Schielzeth (2013).

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861 **Figure 6.** Differences in soil pH, total nitrogen and exchangeable P, K, Ca, Mg and Al
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Table 1. Description of study sites and experimental designs.

Feature		Site					
		DUD	LIG	LL	SFX	STK	WED
Tree age at the time of measurement or sampling	growth	5	3	3	5	4	4
	foliage	5	2	3	4	4	4
	soil	5	2	3	5	4	4
Application rate in the fertilized plots (t ha ⁻¹ , fresh weight) †	PB	120	140	140	125	140	120
	LM	10	10	15	12	10	10
Nb. of blocks		1	3	2	1	2	3 ††
Nb. of plots per block		6	2	2	10	4	2
Nb. of trees per plot		5	8	10	5	5	5
Total nb. of trees		30	48	40	50	40	30
Nb. of soil samples per treatment		3	9	6	5	4	1 or 5 †††
Latitude, N		45°36'54"	45°40'02"	45°43'43"	45°34'01"	45°35'09"	45°38'02"
Longitude, W		71°30'41"	71°16'56"	71°23'40"	72°04'59"	71°42'57"	71°24'54"
Altitude, m a.s.l.		230	365	275	270	315	285
Mean summer temperature, °C		15.6	14.7	14.7	16.8	15.6	15.6
Mean annual temperature, °C		4.4	3.8	3.8	5.6	4.4	4.4
Summer rainfall, mm		467	504	504	475	467	467
Annual rainfall, mm		940	966	966	940	940	940
Annual snowfall, cm		220	359	359	207	220	220

† Typical moisture content is 75% w/w for paper biosolids (PB) and 30% for lime mud (LM).

†† Only one of these three blocks had a control plot.

††† One sample for the control, five samples for the fertilized treatment.

Table 2. Chemical properties of papermill biosolids and lime mud as well as of a podzolic B horizon sampled from a control plot at the SFX site. Values are means from three to ten samples tested for each material, depending on variable.

Variable †	Biosolids	Lime mud	Soil (B horizon)
Density, kg m ³	900	1050	1100
Moisture, %	□75	□30	n.a.
pH	7.90	11.0	5.70
Neutralizing capacity, %	24	91	n.a.
N, %	1.23	0.02	0.20
C, %	21.9	11.2	2.97
C/N	17.8	560	14.8
Loss on ignition, %	53	43	8.50
SiO ₂ , %	29.27	1.02	73.1
Al ₂ O ₃ , %	6.30	0.35	9.52
CaO, %	10.3	53.3	0.93
K ₂ O, %	0.88	0.07	1.84
MgO, %	2.41	0.78	0.71
MnO, %	0.08	0.03	0.07
Na ₂ O, %	0.88	1.01	1.75
P ₂ O ₅ , %	0.47	0.60	0.10
Sum, %	53.1	57.5	92.8

Table 3. Summary of hybrid poplar growth per site and treatment. Values for annual height increment and age-standardized diameter at breast height (DBH) are means $\pm$ standard deviation (SD). The coefficient of variation (CV) is the SD divided by the mean and expressed as a percentage. The gain in growth compares the means of fertilized vs control plots, either in absolute (cm) or relative (%) terms.

Variable	Treatment / Measurement	Site					
		DUD	LIG	LL	SFX	STK	WED
Annual height increment, cm	control	30.2 $\pm$ 14	23.8 $\pm$ 19	63.8 $\pm$ 30	118 $\pm$ 30	27.5 $\pm$ 16	64.3 $\pm$ 10
	fertilized	120 $\pm$ 19	99.9 $\pm$ 35	154 $\pm$ 23	154 $\pm$ 12	109 $\pm$ 16	143 $\pm$ 41
Age-standardized DBH, cm	control	0.5 $\pm$ 0.2	0.4 $\pm$ 0.2	0.8 $\pm$ 0.3	1.4 $\pm$ 0.4	0.4 $\pm$ 0.1	0.8 $\pm$ 0.2
	fertilized	1.9 $\pm$ 0.4	1.6 $\pm$ 1.3	2.1 $\pm$ 0.5	1.9 $\pm$ 0.4	1.5 $\pm$ 0.3	1.9 $\pm$ 0.7
CV, % (height)	control	46.3	79.7	47.1	25.5	58.2	15.6
	fertilized	15.9	35.0	14.9	7.8	14.7	28.7
CV, % (DBH)	control	43.5	53.3	39.8	28.1	25.4	25.8
	fertilized	20.6	83.0	23.5	21.0	20.1	36.7
Gain in growth, cm	height	89.3	76.1	90.4	36.4	81.3	78.7
	DBH	1.5	1.2	1.4	0.5	1.1	1.1
Gain in growth, %	height	296	319	142	31	296	122
	DBH	323	317	182	34	280	146

Table 4. Partial results of the linear mixed models that tested the effect of (a) fertilization on tree growth, foliar nutrition, and soil chemistry, and (b) foliar nutrition on tree growth.

Complete results are available in supplementary materials.

Explanatory variable	Response variable	F-value	P-value
(a)			
Fertilization	Tree growth		
	Annual height increment	74.5	< 0.0001
	Age-standardized DBH	54.3	< 0.0001
	Foliar nutrition		
	Foliar N	12.6	0.00073
	Foliar P	40.5	< 0.0001
	Foliar K	2.79	0.099
	Foliar Ca	86.6	< 0.0001
	Foliar Mg	20.5	< 0.0001
	Foliar Al	40.8	< 0.001
	Soil chemistry		
	Soil N	7.48	0.23
	Soil P	0.669	0.51
	Soil K	8.14	0.0063
	Soil Ca	48.5	< 0.0001
	Soil Mg	14.2	0.00041
	Soil Al	7.34	0.009
	Soil pH	15.2	0.00027
(b)			
Foliar N	Annual height increment	41.9	< 0.0001
Foliar P	Annual height increment	38.5	< 0.0001
Foliar K	Annual height increment	0.208	0.65
Foliar Ca	Annual height increment	44.7	< 0.0001
Foliar Mg	Annual height increment	7.89	0.032
Foliar Al	Annual height increment	25.7	< 0.0001
Foliar N/P	Annual height increment	17.1	0.0002
Foliar Ca/Al	Annual height increment	58.6	< 0.0001

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279x215mm (96 x 96 DPI)

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238x286mm (96 x 96 DPI)

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190x237mm (120 x 120 DPI)

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298x269mm (120 x 120 DPI)

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246x497mm (120 x 120 DPI)

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378x441mm (96 x 96 DPI)