Long term carbon sequestration potential of biosolids-amended copper and molybdenum mine tailings following mine site reclamation

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ABSTRACT

Management and reclamation of industrial mine sites for carbon (C) sequestration is an emerging technique for offsetting anthropogenic C emissions. Land application of municipal biosolids is an effective method for amending closed tailings storage facilities and providing the nutrients to establish a vegetative cover. Biosolids applications can influence the C sequestration potential of tailings and other mine wastes at the onset of reclamation by initiating soil development processes and enhancing primary productivity, thereby leading to increased accumulation of soil organic carbon (SOC) over time. The short term ecological benefits of biosolids applications are well understood, but the long-term (> 10 years) effects of biosolids on reclaimed mine soils are under-researched. The objective of this long-term study was to determine the effects of biosolids applied in 1998 at increasing rates (0, 150 and 250 dry Mg ha−1) on the C sequestration potential of a copper and molybdenum mine tailings site in the southern interior of British Columbia, Canada that is currently undergoing reclamation to a pasture-based ecosystem. We assessed changes in C pools, plant productivity and select soil physiochemical parameters at an established research site at the Bethlehem Tailings Storage Facility over a 13-year period spanning from 1998 to 2011. Tailings total C and N concentrations increased with time and were highest when biosolids were applied at 250 Mg ha−1. Carbon pools increased with increasing biosolids application and ranged from 23 to 155 Mg C ha−1 after 13 years of reclamation. The net SOC sequestration rates (i.e. the C sequestration potential) ranged from 0.72 to 6.3 Mg C ha−1 yr−1 and were highest at the 250 Mg ha−1 application rate. The C storage efficiency was higher in the 150 Mg ha−1 treatment (0.74 Mg C stored per Mg of biosolids applied), indicating that lower application rates of biosolids are more efficient at storing C than higher application rates. Aboveground plant biomass was substantially higher on biosolid-amended tailings (6 and 6.7 Mg ha−1) compared to the unamended tailings (0.39 Mg ha−1), which suggests that the increase in C pools was a direct result of organic matter inputs from enhanced plant productivity. The tailings were naturally high in Cu and Mo, and when amended with biosolids at a rate of 250 Mg ha−1, elevated levels of Zn (as compared to federal soil quality guidelines) were detected. The unamended tailings increased in alkalinity with time, whereas the pH of the biosolid-amended tailings remained stable around neutral. This study demonstrated that a single application of biosolids can facilitate plant production and the accumulation of SOC on mine tailings for more than a decade, and supports the use of biosolids for promoting long-term C sequestration on reclaimed mine sites in similar environments.

1. Introduction

In recent decades, changes in the Earth’s climate such as a rise in episodes of temperature extremes, changes in precipitation patterns, and alteration of drought regimes have been observed throughout many regions of the world (Heim, 2015; IPCC, 2014). The observed climate changes are closely associated with increases in anthropogenic carbon dioxide (CO₂) and other greenhouse gas (GHG) emissions since the pre-industrial era, which are being driven by rising populations and rapid economic growth (IPCC, 2014). Human activities, primarily fossil fuel combustion and land use changes, are responsible for an estimated 555 ± 85 Pg of CO₂ released into the atmosphere over the past 200 years (Ciais et al., 2013). These emissions have contributed significantly to the observed 45% increase in the atmospheric CO₂ concentration observed over the past two centuries (Ciais et al., 2013).
concentration (280 ± 20 prior to 1750 to 400 ppm in 2015) (Blasing, 2016; Indermühle et al., 1999). As such, there is a need to generate and implement innovative GHG mitigation strategies.

Management of terrestrial ecosystems for carbon (C) sequestration is one strategy for mitigating increases in atmospheric CO₂ concentration (Shrestha and Lal, 2006). Terrestrial ecosystems are an important component of the global C cycle because they are capable of reducing the concentration of atmospheric CO₂ and storing it in plant biomass and soils as soil organic carbon (SOC). The process where CO₂ is fixed into above-ground (leaves and shoots) and below-ground (roots) biomass through photosynthesis and assimilated into soils by decomposition and microbial activity is known as C sequestration. The amount of C stored in a terrestrial ecosystem is commonly referred to as the terrestrial C sink (Akala and Lal, 2001). An estimated total of 100–200 Gt C (1–2 Gt C·yr⁻¹) was lost from terrestrial ecosystems as a result of land use changes since the year 2000 (Strassmann et al., 2008). One study in Ohio, USA reported that land disturbance from coal mining caused a 70% decline in the local C sink (Akala and Lal, 2001). In contrast, encouraging C sequestration on such disturbed or degraded ecosystems can lead to replenishing the terrestrial C sink and help mitigate global climate change (Juwarkar et al., 2010; Shrestha and Lal, 2006).

Land disturbances from surface mining include tailings storage facilities, mined lands, waste rock piles, and road sides. Because the soils on such sites are initially low in SOC, the potential to store C over time is high (Brown and Leonard, 2004). However, plant productivity on these sites is generally difficult to restore due to a variety of physiochemical limitations, beginning with the parent material (Brashaw, 1987a). For example, tailings (fine waste materials generated from ore processing) are typically characterized by elevated levels of heavy metals, lack of organic matter and plant-essential nutrients, and extreme pH (Gardner et al., 2012, 2010; Mendez and Maier, 2008, 2007; Pedersen et al., 2017), and so they are not a suitable medium for plant growth. Successful reclamation of these mined lands can improve C sequestration and offset some of the emissions caused by mining activities (Shrestha and Lal, 2006). Furthermore, the emergence of C markets may present economic benefits to mining operations that promote C sequestration during reclamation. In one study, C sequestration rates of reclaimed coal mine soils reached up to 3.1 Mg C ha⁻¹ yr⁻¹ in a grassland ecosystem and 4 Mg C ha⁻¹ yr⁻¹ in a forested ecosystem (Akala and Lal, 2001).

Conventional techniques for reclaiming abandoned mine sites include fertilizer applications and amending the upper surface with stockpiled topsoil, but these methods are often not sufficient to produce sustained plant growth over long periods of time (Sopper, 1993). Organic soil amendments, such as compost and manure, can enhance the long-term productivity of mine tailings, and have been successful in a variety of management scenarios (Brown et al., 2007; Carson et al., 2014; Shrestha et al., 2009). In general, organic amendments improve soil conditions by providing nutrients and organic matter which can have synergistic positive effects on physical, chemical, and biological parameters relating to soil development and ecosystem recovery (Brown et al., 2003; Park et al., 2011; Sheoran et al., 2010; Tarrasón et al., 2014). Municipal sewage sludge, or biosolids, are commonly used as a soil amendment to improve the productivity of degraded agricultural and forested systems (Bai et al., 2017; Pritchard et al., 2010), and have also been used successfully on mine sites (Sopper, 1993; Tian et al., 2009). Biosolids can promote plant production by improving soil chemical properties such as organic C content, plant nutrient availability, cation exchange capacity, and soil pH (Cele and Mabota, 2016; Gardner et al., 2010). They are also capable of ameliorating harsh physical properties common of tailings such as low porosity, poor moisture retention, and reduced soil aggregation (Drozdowski et al., 2012; Gardner et al., 2010). Furthermore, biosolids can improve nitrogen (N) cycling and soil development by accelerating soil microbial activity (Gardner et al., 2010; Kim and Owens, 2010; Pepper et al., 2012). In addition to the above benefits, increases in above- and below-ground C pools resulting from biosolids application have been measured (Brown et al., 2003; Sopper, 1993). In one study, Trlica and Teshima (2011) showed that biosolids-amended copper (Cu) and molybdenum (Mo) mine tailings in British Columbia, Canada stored more C in the upper 0–15 cm layer than conventionally reclaimed tailings over an 8-year period. As such, the use of biosolids for restoring terrestrial C pools on mine tailings sites is a possible GHG mitigation strategy. Though, potential drawbacks to the use of biosolids for land reclamation have been noted, such as the tendency for metals and other contaminants to accumulate in the ecosystem (Dung et al., 2015; Sopper, 1993), reduction in soil fauna numbers (Brown et al., 2014; Waterhouse et al., 2014) and alteration of microbial communities (Shah et al., 2014), all of which warrant further investigation into the use of biosolids as a C sequestration tool.

To date, few studies have investigated the long-term (> 10 years) effects of biosolids on revegetation of metal mine tailings sites, and whether positive levels of SOC, as well as plant productivity, persist (Brown et al., 2014; Pepper et al., 2013; Trlica and Teshima, 2011). Some research has addressed C sequestration on reclaimed mine lands but many of these studies were conducted on short-term (<5 years) studies (e.g. Brown et al., 2003; Drozdowski et al., 2012; Gardner et al., 2012, 2010). Here we are presented with a unique opportunity to investigate the use of biosolids as a tool for restoring terrestrial C pools on reclaimed mine tailings sites, with the overall goal of reducing atmospheric GHG concentrations and mitigating global climate change.

The purpose of this research was to assess the long-term effects of a one-time biosolids application at three rates (0, 150 and 250 Mg ha⁻¹) on the C sequestration potential of a Cu and Mo mine tailings storage site located in the southern interior of British Columbia, Canada that has used biosolids in its reclamation program. A research site from a previous study (Gardner et al., 2012, 2010) was revisited to investigate changes in C pools, plant biomass yield, and select soil physiochemical parameters spanning a 13-year reclamation period from 1998 to 2011.

2. Materials and methods

2.1. Site description

The research was conducted at Teck-Highland Valley Copper (HVC), an open pit Cu-Mo mine located in the southern interior of British Columbia (BC), Canada, approximately 80 km southwest of the city of Kamloops (Fig 1; 50°28'23"N 121°01'19"W). Mining operations were first established in the Highland Valley district over 50 years ago, and currently account for around 15% of BC's total Cu production (Mining Association of British Columbia, 2015). The mine processes ore from a low grade (0.4294% Cu over the life of the mine) Cu-Mo porphyry (Casselman et al., 1995).

The study site is located on the Bethlehem Tailings Storage Facility (50°30'43"N 120°58'29", 1475 m elevation) which is one of three decommissioned tailings sites at HVC that has been reclaimed with biosolids. The tailings site covers an area of approximately 218 ha, with an average depth of approximately 40–50 m and a maximum depth of 90 m (Jaimie Dickson, Teck-Highland Valley Partnership, personal...
communication). The mining operation used Class A and Class B biosolids from Metro Vancouver as a soil amendment for their reclamation program from 1996 to 2014 (Metro Vancouver, 2017). To ensure the protection of public health and the environment, biosolids are classified based on quality criteria, such as trace elements and pathogen reduction, prior to land application. In BC, they are classified as either Class A or B products, where Class A criteria are more stringent (Sylvis Environmental, 2008). The reclamation of the Bethlehem tailings site included one-time biosolids applications (up to 250 Mg ha$^{-1}$) or frequent (every 4 years) fertilizer treatments and seeding with a reclamation seed mix of native and/or agronomic grasses and legumes. At the time of the study, the primary land use goal for the site was to promote wildlife forage and habitat. As such, the existing vegetation has never been mowed or grazed by livestock, but the site is accessible to wildlife.

The mine site is located within the Montane Spruce and Interior Douglas-Fir biogeoclimatic zones (Government of British Columbia, Ministry of Forests 1991), and is influenced by a continental climate with dry summers, cold winters and moderately short growing seasons. Mean annual precipitation is 387 mm (54% occurring from April to October) (Table 1) and moisture deficits are common during the growing season. Predominant soils for this area consist of Eutric Brunisols and Gray Luvisols (Government of British Columbia, Ministry of Forests 1991).

### 2.2. Experimental design

A long-term study site was established on the Bethlehem tailings site in 1998 by Gardner et al. (2012, 2010) to investigate the effects of biosolids on soil and vegetation parameters during reclamation. The site consisted of eight replicates of seven treatments organized in a randomized complete block design. Each plot was 3 by 7 m, with a 1 m buffer zone between adjacent blocks. In August 1998, anaerobically digested sewage sludge (Class B biosolids) from Metro Vancouver were randomly applied to the treatment plots and incorporated into the upper 15 cm of the tailings with a tractor-mounted rotovator. The seven treatments included biosolids amendments of a single application of 50, 100, 150, 200, and 250 dry Mg ha$^{-1}$, a fertilizer application and a control with no biosolids. In June 1999, the sites were seeded at a rate of 36 kg ha$^{-1}$ with a mixture of 33.2% pubescent wheatgrass (Agropyron trichophorum (Link) Richt.), 7.5% orchard grass (Dactylis glomerata L.), 4.0% creeping red fescue (Festuca rubra L. var. rubra), 14.7% Russian wild rygrass (Elymus juncus Fisch.), 34.6% alfalfa (Medicago sativa L) and 5.9% alskie clover (Trifolium hybridum L.), determined by seed weight.

For the current study, three of the original seven treatments were selected for analysis of soil and plant productivity parameters. The treatments chosen were biosolids applications of 150 and 250 dry Mg ha$^{-1}$ (herein B150 and B250, respectively), and the control. These biosolids application rates are reflective of operational application rates used at HVC (Metro Vancouver, 2017).

#### 2.3. Tailings and biomass sampling

Field sampling methodology was guided by Gardner et al. (2010, 2012). Tailings and biomass samples were collected in September 2011, at the end of the 13th growing season following reclamation. Five random soil cores were taken from the 0–15 cm layer of each plot using a standard soil auger and compiled into a single composite sample (a total of eight composite soil samples per treatment). Subsamples of the composite samples were taken and weighed, then dried for 24 h at 105°C and re-weighed to determine soil moisture content. Photosynthetic material such as leaves and shoots were removed and the soil samples were air-dried to constant weight for 78 h. Root scores ranging from 1 to 5 (based on relative abundance) were subjectively determined for each tailings sample to quantify the abundance of belowground biomass for each treatment. Tailings samples were oven-dried for 6 h at 60°C to complete drying and passed through a hammer mill fitted with a 2 mm sieve.

Tailings subsamples were sent to the ALS Laboratory Group (Burnaby, British Columbia, Canada) to determine pH, plant available nutrients (nitrate (NO$_3$) and phosphate (PO$_4$)) and total metal content. Tailings pH was measured using a 1:2 soil water extraction and a standard electrode (British Columbia Ministry of Water, Land and Air Protection 2003). Plant available NO$_3$ was extracted from the soil with a 2.0 M calcium chloride solution and reduced to nitrite (NO$_2$) by passing through a copperized cadmium column, then diazotized with sulfanilamide and coupled with N-(1-naphthyl) ethylenediamine dihydrochloride and measured colorimetrically at 520 nm (Alberta Agriculture, 1988). Plant available PO$_4$ was extracted from the soil using a Modified Kelowna extracting solution, then reduced with ascorbic acid and measured colorimetrically at 880 nm (Qian et al., 1994). Concentrations of arsenic (As), cadmium (Cd), chromium (Cr), cobalt (Co), Cu, lead (Pb), Mo, nickel (Ni), and zinc (Zn) were determined by strong acid digestion using a block digester at 95°C for 2 h with concentrated nitric acid and hydrochloric acid (US Environmental Protection Agency, 1994) and were analyzed using collision cell inductively coupled plasma mass spectrometry (ICP-MS) (US Environmental Protection Agency, 1998). The laboratory procedures for metals analysis were targeted to dissolve those metals that are environmentally available (strong acid leachable metals), and were similar to those employed by Gardner et al. (2010, 2012) for the 1998–2000 data used in this study. Small (approx. 3 tbsp.) subsamples were taken and crushed with a “pulverizer” (manufactured by TM Engineering, Vancouver, British Columbia, Canada). These samples were sent to Agriculture and Agri-Food Canada (Lethbridge, Alberta, Canada) for the purpose of assessing total C and total nitrogen (N) content using a modified Dumas method with a Carlo Erba NA2100 analyzer (Tabatabai and Smith, 2003). Tailings inorganic carbonate content was not measured because concentrations were expected to be insignificant.

<table>
<thead>
<tr>
<th>Climate data from the Lornex weather station$^a$ at Teck-Highland Valley Copper mine site.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1998</td>
</tr>
<tr>
<td>---</td>
</tr>
<tr>
<td>Annual precipitation (mm)</td>
</tr>
<tr>
<td>Precipitation Apr. to Oct. (mm)</td>
</tr>
<tr>
<td>Mean temperature Apr. to Oct. (°C)</td>
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<tr>
<td>Growing degree days</td>
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<tr>
<td>Freeze-free period (days)</td>
</tr>
</tbody>
</table>

$^a$ Highland Valley Lornex weather station located 5 km from study site.

2.5. Data analyses

Soluble salts (as indicated by electrical conductivity values) were high in copper, lead and zinc as well as elevated levels of Cu and Mo compared to the federal soil quality guidelines (Coulloudon et al., 1999) to determine per hectare biomass yield. Current year’s aboveground growth was clipped as close to the soil surface as possible. Samples were placed in paper bags and oven-dried for 24 h at 60°C. Dry biomass weights were measured and extrapolated to a per hectare yield basis.

2.4. Characterization of biosolids and tailings materials

Baseline physiochemical data for the biosolids and tailings materials from Gardner et al. (2010, 2012) were compiled for comparative purposes (Table 2). The tailings were classified as a silt loam texture and consisted of 60% plagioclase, 10% potassium feldspar, 10% quartz, and several other minerals including biotite, hornblende and calcite (Gardner et al., 2010). They were naturally low in C and N and had elevated levels of Cu and Mo compared to the federal soil quality guidelines. The Class B biosolids from Metro Vancouver contained ample C and N levels and were high in copper, lead and zinc as well as soluble salts (as indicated by electrical conductivity values). 

Baseline physiochemical data for the biosolids and tailings materials prior to treatment. (Table 2) was used to test for relationships between SOC pools and time; the data was fitted to a second-order logarithmic function. The data for year zero was considered anomalous (due to the high proportion of biosolids C), and so it was omitted from the data fitting. Simple linear regression was employed to analyze for correlations between C pools and aboveground biomass yields. Tailings total metal concentrations were compared to the Canadian Council of Ministers of the Environment (CCME) federal soil quality guidelines for agricultural use (Canadian Council of Ministers of the Environment, 2014).

3. Results

3.1. Total carbon

The amount of total C in the upper 0–15 cm soil layer differed by biosolids treatment in year zero (ANOVA: F = 281, P < 0.0001), with the unamended control having the least amount of C and the 250 Mg ha⁻¹ with the greatest (Table 3). A similar pattern was found in year 13 of the reclamation period (ANOVA: F = 254, P < 0.0001). Net gains in total C over the reclamation duration were significant across all treatments, including the control (Paired t-test: P < 0.0001). 

3.2. Carbon pools and sequestration rates

Both time and biosolids application rate positively influenced tailings total C pools over the 13-year reclamation period (Fig. 2). Reclamation duration and total C pools were positively correlated (ANOVA: Control, F = 12.6P < 0.001; B150, F = 36.4, P < 0.0001; B250, F = 71.0, P < 0.0001), with coefficient of determination values (R²) becoming stronger with increasing biosolids application. The C pools increased from 63 to 111 Mg ha⁻¹ (76%) in the B150 treatment, and compiled with the 2011 (year 13) data to create a time series showing changes over the 13-year reclamation period. Total C was extrapolated to a per hectare basis for each treatment using the equation

\[
Mg \text{ C ha}^{-1} = \frac{C(\%) \times \text{bulk density (Mg m}^{-3}) \times \text{soil depth (m)} \times 10^4 \text{m}^2 \text{ha}^{-1}}{100}
\]

from Akala and Lal (2001) to estimate terrestrial C pools in the upper 0–15 cm layer. Soil bulk densities from 1999 and 2000 (Gardner et al., 2010) were used to calculate C pools for years 1 and 2. Bulk density measurements were not taken in 2011, so data from a 2015 study (unpublished research) were used to calculate year 13 C pools. Total C pools for year 13 were divided into initial C, biosolids C and sequestered C. Initial C was assumed to equal the amount of C in the control plots in year zero. For simplicity, biosolids C was estimated by subtracting initial C from year zero C pools, assuming decomposition of biosolids C was negligible over the reclamation duration (Brown and Leonard, 2004). Sequestered C was determined by calculating the net gain in C over the reclamation duration. Two different linear rates of C accumulation were calculated: the “gross SOC sequestration rate” and the “net SOC sequestration rate”. The gross SOC sequestration rate included biosolids C whereas the net SOC sequestration rate did not. The rates were calculated by dividing the respective C pool by the reclamation duration of 13 years. The amount of C stored per unit weight of biosolids applied (known as the C storage efficiency) (Tian et al., 2009; Trlica, 2010) was calculated by dividing the amount of C in a given year by the amount of biosolids applied. 

One-way ANOVA was conducted to determine if there were statistical differences between treatments within years (P < 0.05), and Tukey’s HSD test was employed to determine which treatment was most effective when statistical differences were detected. Paired t-tests were conducted to analyze differences between 1998 and 2011 parameters for the same treatments (P < 0.05). Logarithmic regression analysis was used to test for relationships between SOC pools and time; the data was fitted to a second-order logarithmic function. The data for year zero was considered anomalous (due to the high proportion of biosolids C), and so it was omitted from the data fitting. Simple linear regression was employed to analyze for correlations between C pools and aboveground biomass yields. Tailings total metal concentrations were compared to the Canadian Council of Ministers of the Environment (CCME) federal soil quality guidelines for agricultural use (Canadian Council of Ministers of the Environment, 2014).
73 to 155 Mg ha\(^{-1}\) (111\%) in the B250 treatment and 13 to 23 Mg ha\(^{-1}\) (70\%) in the control treatment. Net gains in C pools from year zero to year 13 were significant for all treatments (Paired \(t\)-test: \(P < 0.001\)). The C pools of the two biosolids treatments were statistically similar through the first two years of reclamation, but after 13 years, the B250 treatment had a significantly larger (39\%) C pool compared to the B150 treatment (ANOVA: \(F = 151, P < 0.0001\)).

### Table 3

<table>
<thead>
<tr>
<th>Parameter; Treatment (^a)</th>
<th>Year zero (^b)</th>
<th>Year 13</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Control B150 B250</td>
<td>Control B150 B250</td>
</tr>
<tr>
<td>Total C (g kg(^{-1}))</td>
<td>6.78 (0.48) C</td>
<td>69.6 (5.52) A</td>
</tr>
<tr>
<td>Total N (g kg(^{-1}))</td>
<td>0.16 (0.04) C</td>
<td>9.36 (0.73) A</td>
</tr>
<tr>
<td>C:N ratio</td>
<td>46.7 (2.74) A</td>
<td>13.7 (1.38) C</td>
</tr>
<tr>
<td>NO(_3)-N (mg kg(^{-1}))</td>
<td>1.45 (0.27) C</td>
<td>0.37 (0.06) C</td>
</tr>
<tr>
<td>PO(_4)-P (mg kg(^{-1}))</td>
<td>13.0 (1.35) B</td>
<td>0.37 (0.06) C</td>
</tr>
<tr>
<td>pH</td>
<td>7.5 (0.14) A</td>
<td>8.0 (0.10) A</td>
</tr>
</tbody>
</table>

\(^a\) Data for 1998 study year from Gardner et al. (2010, 2012).

\(^b\) B150 and B250 represent biosolids applications of 150 and 250 dry Mg ha\(^{-1}\) applied in 1998; control treatment with no biosolids application.

\(^c\) Indicates statistical significance (Paired \(t\)-test: \(P < 0.05\)) between years for that treatment.

### Fig. 3

Carbon sequestration potential of biosolids reclaimed tailings (0–15 cm depth) after 13 years of reclamation under three reclamation scenarios; the control treatment represents unamended tailings and B150 and B250 represent one-time biosolids application rates of 150 and 250 dry Mg ha\(^{-1}\). “Initial C” is the amount of C prior to biosolids application; “Biosolids C” is the C added from the one-time biosolids application and “Sequestered C” represents the net gain in C after 13 years of reclamation. The rates shown represent net soil organic carbon sequestration rates (net gain in C divided by the reclamation duration).

### Fig. 2

Time series of total carbon pools ± standard error in 0–15 cm soil depth over a 13-year reclamation period. The B150 and B250 treatments represent one-time biosolids applications of 150 and 250 dry Mg ha\(^{-1}\), respectively and the control treatment represents unamended tailings (n = 8). Time zero represents sampling conducted in fall 1998, three months following biosolids addition. Tailings were not investigated during years 3 through 12. Data for years 1, 2 and 13 were fitted using a second-order logarithmic regression curve (\(P < 0.05\)). Treatments with different letters (across graphs) within the same year are significantly different (Tukey’s HSD: \(P < 0.05\)). \(^*\) Indicates statistical significance (Paired \(t\)-test: \(P < 0.05\)) in C pool gains from year zero to year 13 for that treatment.
3.3. Tailings physical and chemical properties

Biosolids had an immediate positive effect on several of the soil parameters investigated (Table 3). In year zero, total N (ANOVA: $F = 270$, $P < 0.0001$), available N ($NO_3$) (ANOVA: $F = 270$, $P < 0.0001$), C:N ratio (ANOVA: $F = 82.4$, $P < 0.0001$) and available P ($P_2O_5$) (ANOVA: $F = 10.9$, $P < 0.001$) differed between amended and unamended tailings. The above parameters all increased with biosolids application with the exception of C:N ratio which decreased. After 13 years of reclamation, treatment effects remained prominent for total N (ANOVA: $F = 227$, $P < 0.0001$), available N (ANOVA: $F = 14.8$, $P < 0.0001$), C:N ratio (ANOVA: $F = 57.5$, $P < 0.0001$) and available P (ANOVA: $F = 764$, $P < 0.0001$).

Reclamation duration also affected several of the investigated soil parameters (Table 3). Significant net gains were detected in all treatments for total N (Paired $t$-test: control, $P < 0.05$; B150 and B250, $P < 0.001$) and available P (Paired $t$-test: control, $P < 0.05$; B150 and B250, $P < 0.001$). A significant increase in C:N ratio (Paired $t$-test: B150 and B250, $P < 0.0001$) and a significant decrease (Paired $t$-test: B150 and B250, $P < 0.0001$) in available N occurred on the amended tailings. These parameters remained relatively stable in the unamended tailings.

Prior to treatment, the pH of the unamended tailings was slightly alkaline (7.5) and the pH of the biosolids was slightly acidic (6.3) (Table 2) (also see Gardner et al. 2010). The pH of the amended tailings remained similar to the unamended tailings at the beginning of the reclamation period but differed after 13 years (ANOVA: $F = 185$, $P < 0.0001$). Tailings pH of the unamended tailings increased with time and shifted from neutral to slightly alkaline, whereas tailings pH of the biosolids amended tailings decreased but remained within the neutral range over the same time period.

Tailings bulk densities were measured by Gardner et al. (2010, 2012) in 1999 (year 1) and 2000 (year 2), and followed a decreasing trend with biosolids application rate; values were 0.7 Mg m$^{-3}$ in the B250 treatment, 0.9 Mg m$^{-3}$ in the B150 treatment and 1.3 Mg m$^{-3}$ in the control for both years. Tailings bulk densities for 2015 (used for year 13) were 0.6, 0.7, and 1.1 Mg m$^{-3}$ for the B250, B150 and control treatments, respectively.

The highest C storage efficiency (0.73 Mg C per Mg of biosolids applied) was achieved with the lower application rate.

3.4. Plant productivity

The control plots were observed to be sparse in vegetative cover while the plots amended with biosolids were dominated by agronomic grasses. Mean biomass yields across all treatments ranged from 0.0020 to 0.44 Mg ha$^{-1}$ in year zero and from 0.39 to 6.67 Mg ha$^{-1}$ in year 13 (Fig. 5). The addition of biosolids resulted in a significantly higher biomass yield compared to the unamended control across all study years, with the most prominent increase occurring in year 13 (ANOVA: $F = 25.6$, $P < 0.0001$). There were no statistical differences in plant productivity between the two biosolids application rates during any of the years investigated. Biomass productivity increased with time (Paired $t$-test: control, $P = 0.043$; B150 and B250, $P < 0.05$). Biomass yields on the amended tailings were 12–15 times greater in year 13 than in year 1. Surprisingly, plant productivity on the unamended control plots improved substantially (upwards of 200 times the initial yield in year 1) over the reclamation duration. Net differences in biomass yields as compared to the control treatment were 5.61 and 6.28 Mg ha$^{-1}$ for the B150 and B250 treatments, respectively in year 13. A positive correlation existed between tailings C pools and aboveground biomass gained from year zero to year 13 for that treatment. Year zero is excluded because plots were not seeded until year 1. Data for years 1 and 2 were obtained from Gardner et al. (2010). Biomass was sampled at the end of each growing season.

3.5. Tailings metals concentrations

Tailings elemental concentrations of select metals in the 0–15 cm layer for years 1 and 13 of reclamation are reported in comparison to federal soil quality guidelines for agriculture use (Canadian Council of Ministers of the Environment, 2014) in Table 4. Copper, Mo and Zn were the only metals of which total concentrations exceeded the soil quality guidelines after 13 years of reclamation; Cu and Mo were high on all treatments (including the control) while Zn was in exceedance for the B250 treatment only. Concentrations of Cr, Pb, Ni and Zn increased following biosolids application. After 13 years, Mo was the only element with a lower concentration on the amended tailings (relative to the control); all other metals were significantly higher. Treatment effects (ANOVA: $P < 0.05$) were detected for all metals except for Cu;
the concentration of Cu in the amended tailings was similar to the unamended material. In assessing the change in metal concentration over the reclamation period, it was found that concentrations of metals remained statistically unchanged in the biosolids treatments with the exception of Co which increased by 42% from year 1 to year 13 in the B150 treatment (Paired t-test: $P < 0.0001$). It appears that Co increased with time in the B250 treatment as well, but due to the year 1 data being below the detection limit, statistical analysis was not conducted on this parameter. In the unamended tailings, significant changes in metal concentrations were detected for Cr which decreased by 57% (Paired t-test: $P < 0.05$) and Mo which increased by 61% (Paired t-test: $P < 0.05$) while all other metal concentrations remained relatively stable.

4. Discussion

4.1. Effect of biosolids on tailings total carbon and C pools

The one-time biosolids application resulted in higher total C concentrations (Table 3) and C pools (Fig. 2) in the 0–15 cm layer of the tailings when compared to the unamended control, both in the short term (immediately following application) and the long term (after 13 years). The initial increase in C was the direct result of organic matter addition from the biosolids, while the net gain over time can be attributed to increases in SOC from plant biomass (i.e. roots and litter) coupled with improved soil structure and function (Juwarkar et al., 2010; Shrestha et al., 2009; Tian et al., 2009). The rapid decrease in C pools from year zero to year 1 of reclamation can be partially attributed to microbial decomposition and mineralization of the added biosolids C (Gardner et al., 2010). Sopper (1993) stated that, at the onset of mine site reclamation, most of the SOC originates from the biosolids amendment but it is plant biomass that contributes to the buildup in SOC over the long-term. This is supported by the observed increase in plant productivity on the biosolids plots throughout the reclamation period (Fig. 5) and the positive correlation between plant biomass and SOC pools (Fig. 6). The increase in total C in the upper soil layer is an indication that these tailings are developing improved soil characteristics. Shrestha and Lal (2010) stated that SOC accumulates rapidly in reclaimed mine soils, and that it can take as little as 10–15 years for the reclaimed material to resemble a native soil. Soil development and succession on anthropogenic soils, such as reclaimed mine tailings, is an emerging area of research (Howard, 2017).

4.2. Carbon sequestration rates and equilibrium soil carbon content

This study demonstrated that biosolid-amended tailings were capable of storing five to nine times more C per year than the unamended tailings after 13 years of reclamation. When extrapolated to the site level, the entire 218 ha tailings facility has the capacity to offset 1366 Mg of C emissions per year in the upper 15 cm (while consuming about 55,000 Mg of biosolids in the process) during the first 13 years of reclamation. The net SOC sequestration rates of 3.7 and 6.3 Mg ha$^{-1}$yr$^{-1}$ (B150 and B250, respectively) obtained over the duration of this study were nearly threefold those reported by Akala and Lal (2001), however, in their study, topsoil was used as a soil amendment (rather than biosolids) and the reclamation duration extended beyond 20 years. The Tian et al. (2009) study assessed a 34-year reclamation project on strip mined agricultural land in Illinois, USA and also found a net gain in SOC, with sequestration rates reaching up to 3.4 Mg ha$^{-1}$yr$^{-1}$, but biosolids were used in repeated applications.

Since most restored mine sites are relatively young compared to native ecosystems, it is not known if C sequestration rates will persist, and when C pools will equilibrate over time (Brown and Leonard, 2004). In the current study, the logarithmic models (Fig. 2) indicated that SOC sequestration rates diminished with time, following a one-

### Table 4

<table>
<thead>
<tr>
<th>Element; Treatment</th>
<th>Year 1</th>
<th>Year 13</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Control</td>
<td>B150</td>
</tr>
<tr>
<td>As</td>
<td>&lt; 5.0</td>
<td>&lt; 5.0</td>
</tr>
<tr>
<td>Cd</td>
<td>&lt; 1.0</td>
<td>1.50 (0.50)</td>
</tr>
<tr>
<td>Cr</td>
<td>9.38 (1.46) B 15.4 (2.23) A 16.4 (1.18) A 3.98 (0.22) C * 12.2 (1.14) B 17.9 (1.6) A 64</td>
<td></td>
</tr>
<tr>
<td>Co</td>
<td>&lt; 1.0</td>
<td>1.38 (0.18)</td>
</tr>
<tr>
<td>Cu</td>
<td>605 (51.1) A 696 (50.8) A 710 (41.1) A 627 (42.7) A 676 (57.8) A 680 (59.4) A 63</td>
<td></td>
</tr>
<tr>
<td>Pb</td>
<td>2.13 (0.23) B 13.9 (1.95) A 17.8 (2.13) A 1.89 (0.12) B 16.09 (2.42) A 23.6 (3.15) A 70</td>
<td></td>
</tr>
<tr>
<td>Mo</td>
<td>28.8 (2.76) A 28.2 (2.66) A 27.3 (2.16) A 46.51 (7.5) A 28.35 (4.41) AB 27.1 (3.11) B 5</td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>&lt; 1.0</td>
<td>8.25 (0.94) A 9.13 (0.44) A 3.71 (0.19) C 6.64 (0.46) B 8.64 (0.66) B 45</td>
</tr>
<tr>
<td>Zn</td>
<td>14.8 (1.39) B 136 (20.2) A 184 (15.8) A 24.3 (5.1) C 151 (18.7) B 237 (24.8) A 200</td>
<td></td>
</tr>
</tbody>
</table>

Treatments with different letters within the same year are significantly different (Tukey’s HSD: $P < 0.05$). $\bar{x}$ standard error of the mean, $n = 8$.


* B150 and B250 represent biosolids applications of 150 and 250 dry Mg ha$^{-1}$ applied in 1998; control treatment with no biosolids application.

* CCME, Canadian Council of Ministers of the Environment (CCME) soil quality guidelines for agricultural use (2014). Bolded values are in exceedance of the CCME guideline.

* Indicates statistical significance (Paired t-test: $P < 0.05$) between years for that treatment.
time biosolid application, but that SOC pools will continue to increase indefinitely. This contradicts previous research (Shrestha and Lal, 2006), which has shown that SOC pools equilibrate after some time following reclamation.

Ussiri and Lal (2005) stated that equilibrium C pools on mine soils are determined by several factors including climate, soil properties and land use. One way to estimate the achievable equilibrium C content of a given site is to assess nearby undisturbed areas. However, since mine tailings reclamation has traditionally involved seeding with non-native species (Huff, 2009), reclaimed sites differ substantially from pre-disturbance native ecosystems. Our site fits this example because the tailings were reclaimed using agronomic grasses and legumes to achieve a pasture-based end land use, whereas the neighbouring undisturbed ecosystem is primarily forested. Therefore, the C measurements of the adjacent native ecosystem are not representative of the equilibrium C stocks achievable on the mine tailings site.

In one study, Akala and Lal (2001) considered the “equilibrium C content” when modelling temporal changes in SOC pools on reclaimed coal mine soils. They used a first-order kinetic equation which incorporated the equilibrium SOC content of two different land uses, “pasture” and “forest”, which were derived from long-term studies. The values were 55 Mg C ha\(^{-1}\) for the pasture ecosystem and 60 Mg C ha\(^{-1}\) for the forested ecosystem. In this study, the C pools were nearly threefold these values after only 13 years. Akala and Lal’s (2001) models predicted that SOC sequestration rates would decline after 20–30 years of reclamation, and that it could take up to 110–140 years for the SOC pools to reach equilibrium. Further, Trlica and Teshima, (2011) noted lower sequestration rates on older mine sites (< 31 years) reclaimed with biosolids, which suggests that SOC accumulation on our site will continue to diminish over time. It is understood that C sequestration slows down when rates of organic matter input, decomposition and SOC losses (e.g. mineralization, leaching and volatilization) approach a steady state (Ussiri and Lal, 2005).

In the current study, the unamended control plots were also a positive C sink over the reclamation duration. In a long term study of several mine sites, Trlica and Teshima (2011) suggested that, given adequate time, the SOC pools of the unamended soils could reach levels similar to biosolids-amended soils. However, the amount of time needed is generally unknown and likely varies based on site-specific environmental conditions including climate, topography, and the nature of the mine soils being reclaimed; in a study on strip-mined soils, C content of the treated soils remained higher than the untreated soils after 21 years (Malik and Scullion, 1998). Extrapolation of the logarithmic models in this study indicates that SOC pools on the control plots will take about \(1 \times 10^{22}\) years to reach the same level as the B150 treatment.

### 4.3. Soil carbon pool composition

Several factors affect how much of the biosolids C remains in the soil after application, and so it is difficult to estimate the relative composition of biosolids C in relation to the C sequestered from the atmosphere (Brown and Leonard, 2004; Tian et al., 2009). Some studies have analyzed carbon-13 isotopes to differentiate biosolids C from plant biomass C (Dai et al., 2009; Gerzabek et al., 2001). Another study introduced mathematical equations based on empirical models to estimate the amount of biosolids C remaining at a given time following application (Tian et al., 2009) and stated that biosolids decomposition rates were often overestimated. In consideration of C credit policymakers, Brown and Leonard (2004) proposed the idea to calculate C stocks by assuming biosolids C is stable and does not decompose over time when managed for land reclamation. Based on the latter, when calculating our C pool composition (Fig. 3), we assumed that the C remaining from the biosolids was equal to the gain in C immediately following amendment addition in year zero, and that the net gain between year 1 and year 13 resulted solely from primary productivity. By separating our C pools into biosolids C and the SOC gained from atmospheric sequestration, we were able to derive two separate linear C sequestration rates for our site: the “gross C sequestration rate” and the “net SOC sequestration rate”. These results can be useful for carbon credit managers and climate researchers alike; while the gross C sequestration rates can be useful for calculating C credits (Brown and Leonard, 2004), the net SOC sequestration rates give an indication as to the amount of GHGs that can potentially be removed from the atmosphere when long term vegetative cover is encouraged on mine tailings through the use of biosolids.

Depending on the type of parent material and soil depth, inorganic C can make up a significant proportion of total C. In one study conducted on reclaimed coal mine soils in Ohio, USA, inorganic carbon made up almost 30% of the total C in the 0–15 cm layer and increased with soil depth (Shrestha and Lal, 2010). Because of this finding, the researchers cautioned against estimating SOC pools without correcting for the inorganic carbon component. One limitation of this study is that we did not account for inorganic C when calculating our total C pools (Fig. 2). Although, since we considered the initial C in the unamended tailings as being primarily inorganic C (because there was no vegetative cover prior to treatment), we are confident that the inferred C pool composition and net SOC sequestration rates (Fig. 3) are representative of actual conditions. Furthermore, given the pH of < 7.5 on the amended tailings, the potential for carbonate formation on this site is low (Ippolito et al., 2010).

#### 4.4. Effect of biosolids on C storage efficiency

Since C storage efficiency (amount of C stored per weight of biosolids applied) is dependant on SOC pools, the C efficiency values also increased with time for both treatments. Although the highest C sinks and sequestration rates were achieved when biosolids were applied at the high application rate of 250 Mg ha\(^{-1}\), there was evidence that the lower application rate was more efficient at storing C. The highest C efficiency (0.73 Mg C per Mg biosolids) was achieved when amendments were applied at the low application rate of 150 Mg ha\(^{-1}\). Increasing the biosolids application rate by 67% (from 150 to 250 Mg ha\(^{-1}\)) resulted in only a 39% increase in SOC pools and no change in aboveground biomass, which indicates that the additional biosolids do not translate directly to increased productivity. Furthermore, these results suggest that the effect of biosolids additions on SOC sequestration is not linear, and that there is an optimum application rate at which SOC sequestration is maximized; above this threshold, the gain in stored SOC per additional unit weight of biosolids begins to diminish. When extrapolating beyond study years, the models predict that C storage efficiency will be higher in the B250 treatment after ~30 years, which indicates that the higher application rate may be more beneficial in the long-term.

Similar results were obtained by (Trlica, 2010) who found that the increase in soil C per Mg of additional biosolids applied was minimal. In their study, C efficiency rates were derived from five different mine sites across North America (including HVC) that were 2–30 years into reclamation. Their C storage efficiency rates ranged from 0.03 to 0.31 Mg C stored per Mg of amendment in the 0–15 cm layer and decreased with soil depth. They concluded that the application rate at which C storage is optimized varies from site to site and depends on local factors such as climate, topography and the composition of the biosolids used. In another study, Tian et al. (2009) reported a similar trend of decreasing C storage efficiency with cumulative biosolids applications and a mean C storage efficiency of 0.08 Mg C per Mg of biosolids on strip-mined soils. The small C storage efficiency value reported in the latter study can be attributed to their lower estimates of SOC pools, which excluded the estimated amount of C remaining from the biosolids.
4.5. Long term soil carbon storage mechanisms

Though long term C sequestration research is limited on biosolids-reclaimed mine tailings sites (Trlica and Teshima, 2011), some of the same mechanisms for enhancing C storage that were effective on other sites are also applicable to this study. In a literature review, Shrestha and Lal (2006) identified soil aggregation as a critical mechanism for maintaining C sequestration on reclaimed mine soils over the long term. This is because soil aggregates prevent the mineralization of C which allows SOC to persist for long periods of time without being leached through the soil or lost to the atmosphere. According to Denef et al. (2002) soil aggregation can be enhanced in degraded soils by improving SOC concentration and promoting belowground plant root and fungal growth. The positive long-term plant and soil data in this study suggests that biosolids are capable of initiating similar soil development processes. In fact, increases in soil aggregation and structural stability as a result of biosolids application have been demonstrated (Sort and Alcaniz, 1999).

Soil texture is also an important soil physical property that determines the C sequestration potential of newly reclaimed mine soils. In one study, Shrestha and Lal (2011) noted a positive correlation between clay content and SOC accumulation. This is because organic C compounds adhere to clay and colloids more readily than to larger soil particles (Shrestha and Lal, 2006). In this study, the tailings were a silt loam texture which typically contains 0–25% clay particles (Brady, 1990), therefore the potential for C retention is expected to be moderate.

In addition to soil physical properties, land use and soil management practices are also important drivers of long-term SOC sequestration on reclaimed mine soils (Shrestha and Lal, 2011). Sites which are reclaimed to agricultural crops and which are cultivated regularly will typically store less C because conventional tillage practices can break up soil aggregates, leading to increased C mineralization (Usiri and Lal, 2005). On the other hand, when permanent vegetation cover with minimum tillage or no tillage is encouraged, the site will have a higher potential for long term C sequestration because the biomass is returned to the soil where it is decomposed and becomes SOC over time (Brown and Leonard, 2004). With regard to soil amendment type, biosolids have the advantage because certain organic constituents (e.g. humic and fulvic acids) tend to decompose at a slower rate compared to other amendments (Brown et al., 2003; Tian et al., 2009). Other variables such as climate, decomposition rates and soil type can also affect long-term C storage following amendment addition (Gerzabek et al., 2001; Trlica and Teshima, 2011).

4.6. Effect of biosolids on soil physiochemical properties

The C sequestration potential of a reclaimed mine site is dependent on soil productivity and revegetation success (Shrestha and Lal, 2006). In our study, soil chemical parameters that are important for plant growth including pH, total C, total N, available N and available P concentrations were positively influenced by biosolids application (Table 3).

4.6.1. Tailings pH levels

When tailings were amended with biosolids, pH remained relatively stable and within the neutral range (6.5–7.5) throughout the course of reclamation. Without biosolids, the pH of the tailings increased with time. According to Sopper (1993), a pH ranging from slightly acidic to slightly alkaline (6.5–8.0) is preferred by most grasses and legumes planted during mine reclamation. Soil pH plays an important role in promoting nutrient cycling (Sheoran et al., 2010; Shrestha and Lal, 2011) and limiting metal availability (Bolan et al., 2014). For example, at low pH (< 5.5) metals such as Cd, Cu and Zn become readily more available to plants and increase the probability of phytotoxicity (Chang and Page, 2000), while limiting the availability of important plant nutrients such as P (Bolan et al., 2014; Munshower, 1994). Furthermore, rapid changes in soil pH can inhibit root symbioses with certain microbial communities (e.g. mycorrhizal fungi, N-fixing bacteria) (Shrestha and Lal, 2006). The direction, and the change in pH over time is related to the initial pH of the parent material, and the pH of the amendment applied to the soil (Zebarth et al., 1999) as well as any natural chemical processes, such as humification or weathering, occurring over time.

The increase in soil pH on the unamended tailings can be attributed to weathering of the parent material and is indicative of the chemically unstable nature of mine tailings (Nancuecho and Johnson, 2011). A study conducted on manganese mine tailings observed a similar increase in pH (from 6.9 to 7.5) over a 20 year reclamation period, and attributed it to changes in carbonate activity due to weathering of the parent material (Juwarkar et al., 2010). Mine spoils, particularly those derived from coal mining, can contain high amounts of carbonates which causes increases in soil pH (Shrestha and Lal, 2010). However, the rate of carbonate formation significantly decreases when soil pH levels become lower than 7.5 (Ippolito et al., 2010). Based on this there is, in this study, the potential for carbonate activity in the unamended tailings but not in the amended tailings. It is possible that the establishment of a vegetative cover aided to reduce weathering while increasing soil organic matter content which acted to buffer pH levels around the neutral range.

4.6.2. Plant nutrients

Total concentrations of N and available P, as well as the C:N ratio of the 0–15 cm layer were positively affected by biosolids addition in the short term and in the long term. Similar to total C, the initial increase in total N was a direct result of organic matter addition from the biosolids, whereas the gain over time was due to increases in plant biomass organic matter and enhanced rates of decomposition (Drozdzowski et al., 2012; Shrestha et al., 2009). In a review of several studies of reclaimed mine sites, a similar positive relationship between reclamation duration and total N was highlighted by (Shrestha and Lal, 2010, 2006). It is possible that the establishment of legumes (e.g. M. sativa) and an active N-fixing microbial community are responsible for a portion of the gain in total N. The decline in available N (NO₃) content over the reclamation period can be attributed to a combination of natural processes such as leaching, plant uptake, volatilization and denitrification. The observed loss of available N (NO₃) with time provides some indication that the current rate of soil and plant productivity may be indefinite. Nitrogen deficiency is common on mine soil ecosystems and is a core cause of poor plant productivity during reclamation (Bradshaw, 1987b). Shrestha et al. (2009) demonstrated a positive relationship between plant tissue N content and SOC content. In this study, C and N levels increased simultaneously over time and with biosolids addition, which is consistent with their findings.

The concentration of total N in the unamended tailings was extremely low and the C:N ratio was well beyond the range of < 20:1 preferred by most plants (Munshower, 1994). According to Brown et al. (2003), a C:N ratio of around 12:1 indicates proper soil functioning while a stable ratio (at a given value) indicates a self-sustaining vegetative cover, both of which are important components for enhancing SOC sequestration. In our study, similar C:N ratios were achieved after 13 years of reclamation with biosolids. These results indicate that biosolids are capable of stabilizing OM levels over the long-term.

Shrestha and Lal (2006) showed available P decreasing with reclamation age across several sites which contradicts what occurred on the biosolids-amended mine tailings assessed at our site. This could be due to the tendency for biosolids P to persist in the upper soil layer following application (Elliott et al., 2002). Phosphorus plays an important role in biological metabolism and is often a limiting nutrient in terrestrial ecosystems, but in excess quantities, P can be harmful to the environment. It is normal for biosolids applications to result in excess P because application rates are adjusted to meet plant N demands and not...
P (Sopper, 1993).

4.7. Effect of biosolids on aboveground plant productivity

A key to improving C sequestration is to encourage plant growth, and subsequently, the amount of biomass returned to the soils (Shrestha and Lal, 2006). The amount of organic carbon in soils is directly related to biomass yield because higher rates of organic matter inputs occur with increasing productivity. Further, root growth into the soil profile reduces the bulk density and provides a major SOC input (Akala and Lal, 2001). In our study, biosolids positively influenced plant productivity and biomass yields were positively correlated with C pools (Fig. 6). Shrestha et al. (2009) demonstrated a similar relationship between aboveground plant biomass and SOC sequestration rates over time using other organic amendments. Our data indicates that the use of biosolids as a soil amendment was beneficial, not only for improving terrestrial C pools but also for meeting reclamation targets with regards to plant productivity.

The increase in aboveground plant biomass on the amended tailings (Fig. 5) can be attributed to improvements in soil physiochemical parameters (e.g. bulk density, pH, plant available nutrients) resulting from the application of biosolids (Gardner et al., 2010) (Table 3). In a greenhouse study conducted with iron ore mine tailings obtained from a mine in Swaziland, Celé and Maboeta (2016) reported improvements in soil parameters that included water holding capacity, cation exchange capacity and concentrations of plant nutrients (e.g. ammonium, magnesium, calcium and phosphorus) which, together, had a positive effect on plant growth. Although the amount of biomass did not differ between the two application rates, we observed a greater amount of roots in the soil samples taken from the B250 plots (data not shown). This suggests that, similar to Skousen and Clinger’s (1993) study, increasing the biosolids application rate resulted in increases in belowground productivity.

Achieving a productive and self-sustainable plant community is one outcome of successful reclamation and has additional benefits of offsetting GHG emissions through atmospheric C sequestration. In addition to enhancing terrestrial C stocks, the increased plant cover can have secondary ecological benefits such as increase in soil poor space, moderation of soil temperatures, improved moisture content and reduced erosion, which can ultimately lead to higher revegetation success and C sequestration potential (Sopper, 1993).

The plant productivity results compliment the results from the soil physiochemical analysis and suggest that a single application of biosolids (at a sufficient rate) is capable of initiating plant establishment and sustaining a long-term (> 10 years) C sequestration on mine tailings.

4.8. Effect of biosolids on tailings metal concentrations

The addition of Metro Vancouver biosolids led to increased levels of As, Cd, Cr, Co, Pb, Ni and Zn in comparison to the unamended tailings after 13 years of reclamation (Table 4). Of these metals, Cu, Mo and Zn were the only elements to exceed the federal soil quality guidelines for agricultural land use (Canadian Council of Ministers of the Environment, 2014); tailings concentrations of Cu and Mo were in excess of certain metals in the treated tailings (Table 2) (also see Gardner et al., 2012). It is common for biosolids to contain an array of trace elements that originate from industrial and domestic wastewater (Ce and Maboeta, 2016; Sopper, 1993). The rise in concentrations of heavy metals, such as Cu, Zn and Cd, with biosolids application is well documented (Brown et al., 2014; Gardner et al., 2012; Sopper, 1993), especially on sites where applications were repeated several times (Ipolito et al., 2010). It is common for metals to persist in the upper soil layer for numerous years after the application of biosolids because after being released from this material through decomposition, they are slowly adsorbed by inorganic and organic particles contained in the soil-biosolids matrix (Haynes et al., 2009). The pH of the soil influences the availability of heavy metals and whether or not they will be leached from the rooting zone (Haynes et al., 2009; Zebarth et al., 1999).

In addition to accumulating in the soils (Dung et al., 2015), heavy metals can be taken up by plants which can lead to phytotoxicity and reduce plant productivity (Hodson, 2012). Also, important is that soil fauna (e.g. earthworms) and microbial communities can be negatively impacted by certain metals found in biosolids (Bai et al., 2017; Shah et al., 2014; Waterhouse et al., 2014), which can subsequently lead to reduce soil function and overall reclamation success (Celé and Maboeta, 2016). The metals most often associated with phytotoxicity and having a potential to directly inhibit plant growth are: As, Cd, Cu, Ni and Zn (Chang and Page, 2000). On this site, there is minimal risk in regards to phytotoxicity from these metals because at neutral pH these metals are less available for plant uptake (Bolan et al., 2014; Haynes et al., 2009; Mendez and Maier, 2007).

Our results suggest that there is risk of further buildup of metals (particularly Cu and Zn) at the mine site when biosolids are applied at the higher rate of 250 dry Mg ha⁻¹, or when applications are repeated, but this depends on the trace element composition of the biosolids used. Despite the elevated levels of Cu and Zn, there was no direct evidence of inhibited plant productivity or C sequestration capacity resulting from metals contamination.

4.9. Implications of biosolids as a tool for long-term carbon sequestration and greenhouse gas mitigation

As a means to mitigate global climate change and human-induced increases in atmospheric GHG concentrations, several researchers have stressed the importance of managing terrestrial ecosystems for C sequestration (Post and Kwon, 2000; Shrestha and Lal, 2006; Tian et al., 2009; Trlica, 2016). Tian et al. (2009) mentioned that even a small change in the terrestrial SOC pool can contribute to a reduction in atmospheric C, thus it is essential to promote primary productivity on land disturbed by human activities. Shrestha and Lal (2006) highlighted the idea of managing abandoned mine sites for C storage, and in a later study, Shrestha et al. (2009) promoted the use of organic amendments as a means to enhance C sequestration rates. Further, Tian et al. (2009) suggested that biosolids are ideal for promoting long-term SOC sequestration on degraded sites because of their tendency to decompose slowly and retain organic matter and plant nutrients over long periods of time. Though land application of biosolids is often scrutinized (Robinson et al., 2012), uncovered biosolids stockpiles (landfilled or temporarily stored) are a significant source of GHG emissions (Majumder et al., 2014) which only exacerbates the current GHG problem. Brown et al. (2014) determined that a net savings in GHG emissions can be achieved through land application of biosolids during revegetation of mine tailings sites. This is especially important considering the rate of global biosolids production steadily increases from year to year (Wang et al., 2008).

Our research supports the argument for land application of biosolids as a tool to enhance long-term C sequestration potential of reclaimed mine tailings and other degraded sites. The results of this study are distinctive as there have been very few studies which have assessed the long-term (> 10 years) effects of biosolids on productivity and SOC sequestration of mine tailings storage facilities located in dry environments (Brown et al., 2014; Pepper et al., 2013). The Brown et al. (2014) study was conducted on Pb and Zn mine wastes in Missouri, USA; they reported positive effects on plant yield and SOC levels after twelve years of reclamation with a single application of biosolids. Pepper et al. (2013, 2012) found enhanced microbial activity and sustained plant cover on an arid copper mine tailings site after 8–10 years following a biosolids amendment. Our study fits into this body of long-term research and provides useful information for estimating C pools and GHG
The increased risks of elevated concentrations of Cu and Zn when using this rates may be more economical. At the higher application rate, there are sequestration rate, but greater C storage e more moderate rate of 150 Mg ha


