Reclamation of metal-contaminated tailings with organic amendments: pore water quality control and phytostabilization

T.V. Rakotonimaro¹, M. Guittonny-Larchevêque¹, C.M. Neculita^{1‡} ¹ Research Institute on Mines and Environment (RIME), University of Quebec in Abitibi-Temiscamingue (UQAT)

[‡]Corresponding author: RIME-UQAT, Professor, 445 Boul. de l'Université, Rouyn-Noranda, QC, Canada, J9X5E4. Phone: +1 819 762 0971 ext. 2278, Email: Carmen-Mihaela.Neculita@uqat.ca

Abstract

Organic amendments aided with revegetation are considered as a promising prevention approach in a simultaneous physical (limitation of wind and water erosion) and chemical (acid neutralization and metals/metalloids immobilization) stabilization for the reclamation of metal-contaminated tailings. However, the inconsistency in the performance of different amendments that enhance plants growth demonstrates that further studies are still required. To better select potential organic amendments and identify future research avenues, this critical review makes a preliminary compilation of available knowledge on organic amendments and their performance. To this end, data were collected from review papers and case studies (0-5 y), with a particular focus on pore water quality and plant phytostabilization abilities (plant self-sustaining, toxicity). The screening of the most promising materials was then carried out according to whether metallic elements were mobilized/immobilized from pore water through speciation change (not including microbial mediation), or sequestered into rhizosphere or plant aboveground parts. Results showed that mixture of organic and inorganic materials are more efficient than organics alone to reclaim slightly contaminated tailings. The most promising amendments would be the combination of mature and composted animal manures with inorganic materials (hydrated lime). However, fresh compost and biosolids could enhance the release of metalloids in pore water and, possibly, in plants aerial parts. Finally, biochars could be efficient if mixed with raw industrial organics (sewage sludge) but the appropriate vegetation to be used needs to be evaluated. Further studies should focus on plant-vegetation-microbes interactions and the long term stability of organic amendments (>10-20 y).

Keywords

Phytostabilization, revegetation, mine tailings, amendment.

Introduction

Sulfide-bearing minerals in tailings are chemically instable, when exposed to weathering, and generate low quality pore water and contaminated mine drainage which may adversely impact the surroundings [1]. To prevent, limit or mitigate the environmental impacts, reclamation measures are required. Prevention of acid mine drainage (AMD) generation is the main objective of reclamation approaches that often involve the use of covers (water, organic/inorganic materials) with/without vegetation for a control of water and/or oxygen access to the reactive waste [2]. However, in the case of weathered and highly contaminated tailings, reduction of contaminants concentration from pore water may take time with the use of covers [3]. In closed and abandoned mine sites, passive treatment is also used, but the long-term performance is warranted by early implementation [4]. Alternatively, the use of organic materials, as amendments, combined with revegetation (phytostabilization) seems a promising stabilizing approach for the reclamation of metal-contaminated tailings. It offers the advantage of simultaneous physical (limitation of wind and water erosion) and chemical stabilization (acid neutralization, metals/metalloids immobilization). The mixing of tailings with organic amendments is able to improve their physicochemical (water-holding, porosity, nutrition) and biological properties (vegetation development, microbial proliferation) [5-7]. The phytostabilization, which consists of

establishing vegetative covers on the surface of the amended tailings, is used to prevent direct exposure to wind, water, human and animal contact, and to limit the mobility of contaminants through their immobilization within the rhizosphere (rhizoremediation) or accumulation by roots [6, 8, 9]. However, the vegetation should be tolerant to the presence of phytoavailable salts and metals in pore water [10]. In the same time, organic amendments can be a major source of contaminants (metallic elements, pathogens microorganisms, toxins) and their performances regarding mobilization/immobilization of metallic elements in pore water are variable depending on the type (fresh *vs* mature), source (industrial *vs* raw), physicochemical properties, durability (decomposition rate), application mode (alone or in mixture, dosage), tailings characteristics (highly contaminated *vs* slightly contaminated, fresh *vs* weathered) as well as plant species [5-8, 11-13]. In the same time, plants, through root exudates, can influence the physicochemical and microbial characteristics of soil and the redox conditions of the rhizosphere [6]. Thereby, speciation of metallic elements in pore water and their bioavailability may change after amendment and revegetation [5]. To better select appropriate materials and plant species for the reclamation of metal-contaminated mine tailings, this critical review makes a preliminary performance inventory of organic amendments and identifies future research needs for an efficient use.

Method

In order to achieve the objectives of this study, data compiled from review papers (10) and case studies (54) were collected, with a particular focus on pore water quality and plant phytostabilization abilities. Case studies involved laboratory [12, 14, 15], greenhouse [16–18], and field experiments [19–22]. Pore water quality was monitored through the evolution of physicochemical parameters, including pH, electrical conductivity (EC), oxydoreduction potential (ORP), water-holding capacity, cationic exchange capacity (CEC), and concentrations of dissolved organic carbon (DOC), nutrients (Ca, Mg, N, P, K), metals/metalloids (including trace elements- TEs) and organic matter (OM). Phytostabilization failure or success is evaluated through plant aerial growth (comprising germination), biomass production and bioaccumulation (metallic elements concentration in shoot/root tissues), as well as mid-term evolution i.e. 0-5 y (plant self-sustaining, toxicity). Following data synthesis, the sorting of the most promising organic amendments was undertaken depending on whether particular metallic elements were mobilized/immobilized from pore water through speciation change (not including microbial mediation), or sequestered into rhizosphere or plant aboveground parts. Advantages and limits of organic amendments used for the reclamation of metal-contaminated mine tailings were also recorded.

Results

Tailings are fine-grained (<2 mm) materials obtained after extraction of valuable metals/minerals from metalliferous ores. They lack OM, nutrients, soil organisms, physical structure and have low hydraulic conductivity [10, 16, 23, 24], which limit vegetation development. Tailings pore water can exhibit extremely low to neutral pH and high EC, as well as slight to high concentrations of contaminants (metallic elements) [16, 18, 22]. Organic material influences the evolution of physicochemical characteristics of tailings and the speciation of metallic elements, which in its turn has impact on pore water quality and plant development.

Physicochemical characteristics of tailings pore water after addition of organic amendments

Inventory of various organic amendments already tested for tailings reclamation includes composts (green wastes, municipal solid waste, olive mill waste, sewage sludge, spent mushroom), biosolids (sewage sludge, food wastes, sanitary wastes, anaerobic digestate), manures (cow, cattle, pig, poultry), biochars (rice straw, hardwood, oak tree), slurry (pig, cattle) and peat [5–7]. They are used alone, in mixture with organic amendments of the same/different sources [6, 17, 21, 25] or combined with inorganic (alkaline) materials (hydrated lime/limestone, red mud, marble waste) [7, 12, 26-29].

Single organic amendments were generally found less effective to treat pore water relative to mixtures [5, 30], but usually increased pore water pH [12, 17, 22, 31]. However, depending on the applied dose, the pH of the organic amendments itself and the physicochemical characteristics of the tailings (particularly when extremely acidic), the pore water pH hardly increased or even decreased [18, 32, 33]. Indeed, composted sewage sludge (pH 8.2) used alone at a lower dose (<10% dry weight), barely increased the pH of extremely acidic tailings [18]. In addition, mixed compost (cattle manure+ green waste, steer manure) +/- vegetation (grasses) would be efficient only for a very short term (6 months) before acidification of the tailings occur [33]. Finally, the application of green agriculture wastes (plant remains + strawberry substrate; pH 4.9-7.2) + rockwool or limestone (mixture dose of 30-75 t/ha), showing low pH buffering and in a short period of time (~13 months) [32]. Peat (pH 5) can also entail pH decrease [34]. Nevertheless, biochars or inorganic materials could provide additional alkalinity, i.e. soil pH was found significantly higher and stable [17, 20, 31, 32]. In this sense, biochars, alone or with raw organics, are advantageous due to their high pH (7.65-11.3). However, because of their high content in ash, they could increase EC, depending on their source [6, 35, 36]. Hence, biochars manufactured with wood and paper sludge yielded high EC, whereas lower EC was found for those from manures (poultry, cow) and sewage sludge [36-38]. Composts and biosolids are the most common employed amendments, but they could significantly increase the EC depending on their raw materials [5, 15, 16, 27, 39]. Nonetheless, mature compost (e.g., solid olive mill-waste) would not alter pore water EC [12, 22]. Biosolids, compost (green wastes, sewage sludge, solid olive-mill waste) +/inorganic materials and manures increased DOC [6, 20, 22, 27, 29], but not slurries (e.g., pig slurry) [29]. The high DOC in tailings pore water would denote low proportion of humified OM in biosolids as well as manures, indicating their fresh nature [27]. At the same time, biochars decreased pore water DOC (down to 45%) when produced at more than 600°C [6, 31].

Organic amendments supply nutrients (N, P, and K), organic-C to sterile tailings. Compost (rabbit and horse manure, pruning residue, seaweeds, and fruit wastes) +/- biochar and pig slurry increased total C (TC) and N (TN) [17, 29], whereas industrial fertilizer + paper-mill sludge did not increase neither OM or N and P of the tailings [40]. Pig manure and sewage sludge provided low organic-C and TN whereas olive residues and pig slurry increased soil organic-C, dissolved N, K and P [25, 27, 29]. The use of compost alone (mushroom, solid olive-mill waste) increased dissolved P, which concentration did not change when the compost was combined with inorganic material (red mud derivative) [12, 21, 29]. An increase in extractable P in pore water from compost (municipal green waste, organic waste, pine bark) alone or mixed with peat may increase the risk of eutrophication [15]. Composts (solid olive-mill waste), red mud, olive processing solid waste, sewage sludge, paper mill residues also increased soluble K (up to 50- fold), and N (up to 94%) [12, 29, 34]. Biochars provided low nutrients but could improve CEC and water holding capacity (biochars from pruning residues, manures) [36]. Compost could increase also water holding capacity, CEC, and improve aeration of mine tailings [8, 41].

The performance of organic amendments in mobilizing/immobilizing metallic elements is widely variable depending on their nature and source, as well as tailings characteristics. Several factors that could impact the mobility/immobility of metallic elements in tailings pore water were identified such as the pH, EC, CEC, DOC, OM, nutrient content (N, P, K), metal concentrations as well as humification degree. With respect to redox conditions, the mechanisms of metallic elements immobilization during application of organic amendments and revegetation are sorption, complexation and precipitation [5, 18]. The pH is the main factor affecting precipitation, while both OM and soil pH affect adsorption site [5, 17]. For slightly acidic and neutral tailings, amendments containing 14% of OM would be sufficient to reclaim slightly contaminated tailings [16]. Contents in humic acids, OM, DOC and pH affect metals complexation [5]. Availability of Mn is influenced by both pH and ORP of soil while Cu is mainly controlled by OM and is less pH-dependent relative to Cd and Zn [29].

Most of organic amendments mobilize metalloids (As, Sb, Se) depending on P, Fe, OM, DOC concentrations, pH and redox condition [5, 6, 12, 22]. The mobilization of As by using various composts was closely linked to soil pH, which threshold value for higher release started at pH more than 6 [42]. Compost is advantageous to immobilize As, providing that tailings contain high Fe

concentrations (>14%) [22]. Indeed, less As mobilization was noted in tailings containing Fe oxides/hydroxides due to their positive surface charge that could sorb negatively charged As [12].

The sewage sludge alone would not be effective to immobilize metals (Al, Cu, Mn and Pb) in oxidized tailings [14]. Composted yard waste was suggested for Zn and Pb immobilizing in contaminated tailings while composted cattle manure was rather used for Pb [43]. Chicken manure compost would promote Cd speciation (soluble, extractable or residual) in soil at a moderate dose (54 t/ha) [44]. Due to its content in hydroxyl, phosphoryl and phenolic functional groups, spent mushroom compost could immobilize Cd, Pb, Cr by sorption [21]. Biosolids increased Cd and Zn concentrations in pore water [45]. Biochars have been shown to be promising amendments due to their high efficiency in improving physicochemical characteristics of mine tailings (e.g., pH, CEC), but their performance to mobilize/immobilize metallic elements depends on their nature (feedstock and pyrolysis parameter) [7, 36]. Indeed, biochar made from rice, husk and bran would decreased pore water concentrations of Pb, Cd and Zn, whereas straw char-biochar would increase Pb by up to 26% [46]. A combination of biochar and green waste could immobilize Cd and Zn but not Cu and As [6, 47]. Biochar from fir tree + manure reduce Cd and Zn concentrations in soil, which was not the case for biochar from fir tree alone [35, 36]. Better treatment of metallic elements in tailings pore water was found when a mixture between organic amendments (raw/industrial) or with inorganic materials was used. Compost and calcium carbonate improved plant biomass (aerial and roots), for example bushes (e.g., Atriplex halimus L.) [12, 22]. However, the reclamation of highly contaminated tailings with 40-100 t/ha of compost + lime would increase shoot concentrations of As, Cu and Pb in herbs (e.g., Medicago sativa L.) and Zn in shrubs (e.g., Cistus ladanifer L.) [18]. Compost combined with biochar could bind Cu and limit mobility of Pb and Ni [17]. In the long term (≈ 20 y), a mixture of compost (preferably mature)/biosolids (high quality i.e. low metallic elements concentrations) with inorganic materials (lime or gypsum) could immobilize heavy metals (Cu, Cd, Zn and Pb) in tailings with low Fe and Al concentrations [5, 12]. However, compost from green wastes, catering wastes, municipal solid wastes, mixed with lime increased total As and Zn [42, 44].

Interaction between plants and evolution of soil pore water quality needs close appraisal since plants survival is closely related to the mobilization/immobilization of metallic elements in the pore water.

Interactions of amended tailings and plants

Organic amendments enhanced chemical stability of tailings, and improved their physical structure, and plant establishment [48]. Plant candidates should be native, metals/metalloids and salinity tolerant [8, 12]. Noteworthy, the main objective of phytostabilization is the immobilization of contaminants in plants roots instead of shoots. Plants absorb soil nutrients in pore water (in particular mineral N and P) from tailings and organic amendments. They uptake bioavailable metals from soil pore water, hence limiting metal leaching, thereby reducing indirectly pore water contamination [6, 21]. The stabilizing effect of plants on potential contaminants in tailings also includes water absorption by roots, controlling erosion, increasing macropores in the root zone, and binding metals through root exudates with the aid of OM from the organic amendments [6, 8, 16, 48, 49]. Amended tailings with low compaction and bulk density promote root development [19]. A contradictory plant effect is the possible acidification of the rhizosphere due to uptake of cations that could increase metals concentration (often Zn) in soil pore water [17].

Depending on the type of organic amendments that changes tailings physicochemical properties and metals speciation, shoot metals concentration (frequently Zn) in plants growing on amended tailings might be significant, with respect to nutrients particularly N and P, DOC, organic acid contents, and plant species [5, 6, 12, 27]. Amended tailings with high P would have a greater capacity to decrease metal translocation from roots to shoots by the formation of insoluble metal-phosphate in the roots [50]. However, desorption of As could be enhanced, increasing plant uptake due to site sorption competition with P [6]. The C/N ratio of organic amendment and tailings mixtures controls N availability to plants from OM mineralization. Low C/N ratios associated to biosolids and manures + inorganic materials can improve soil fertility while olive residues and mature compost can increase significantly the C/N (>30, translated to low N mineralization) and reduce plant aerial biomass [27, 41,

51, 52]. Plants can also influence C/N ratio (e.g. mustard increased it) [17]. Amended tailings with biosolids, grape residues, olive mill residues, goat manure (alone or in mixture) having high DOC that could form organo-metallic complexes might be unsuccessful for plant growth since the new formed metal species could be adsorbed by plant roots, thus increasing phytotoxicity [27, 45]. DOC can often mobilize Cd and Zn and Cu of soil pore water, thus exposing plants to toxicity risks [27, 45]. For example, tailings amended with fresh organic amendments could contain considerable fulvic acid which increases metals solubility and bioavailability to plants [27]. Limed tailings (e.g., before compost application) would be an option to lessen metals availability, accelerate soil development and achieve successful vegetation growth (*Cistus ladanifer L., Medicago sativa L.*) [18]. Animal manures (cow, goat, pig, poultry) with low contents of metallic elements, mixed with inorganic materials, could sustain plant growth and eventually decrease metal concentrations in shoots (e.g., Pb, Zn), especially when applied for highly acidic and pyritic mine tailings reclamation [6, 25].

Dosage of organic amendments with/without inorganic amendments and plant effect could impact redox condition of the tailings and, thus, metals speciation. A small amount of organic amendments (organic-C <2% dry wt. in mixture) could be advantageous to create reducing environment, by promoting SO₄ reduction in unoxidized tailings [53]. Biochars are promising organic amendments because of their capacity to immobilize a wide variation of metallic elements (Cu, Cd, Pb, Ni, Zn, Tl) in tailings, except for As [20, 31, 35, 36, 55, 55]. The accumulation of metallic elements (e.g., As) in shoots is reduced, depending on the type of the selected plant (e.g., Solanum lycopersicum L.) [47]. However, depending on the type of biochar (fir tree-biochar), higher metals concentration (Cd, Pb, Zn) could be found in shoots (Anthyllis vulneraria, subsp. polyphylla (Dc.) Nyman, Noccaea rotundifolium (L.) Moench subsp. cepaeifolium, Poa alpina L. subsp. alpina (L.) [35, 36]. A dosage of 1.5 % dry wt. biochars (pruning residues, fir tree, manure + fir tree) would be enough to obtain satisfactory aerial mass [35, 36]. The application of high doses (>10%) of raw hardwood biochars for the rehabilitation of highly contaminated tailings could inhibit bioavailable metals/nutrients for plant uptake, especially when combined with grass [20]. An optimal dose of 10% would exhibit greater grass growth in the condition to be mixed with other organic amendments at a higher dose [20]. Recommended dose of chicken manure compost was 54 t/ha and 90-200 t/ha for biosolids to limit Cd and Zn (>15 mg/kg) in Triticum aestivum L. and Lolium perenne L. shoots during the reclamation of slight to moderately contaminated tailings [15, 44, 45]. Addition of 20 t/ha of red mud alone or in combination with compost (olive mill-waste) increased Mn and Tl in shoots, especially Fe and Pb in Zygophyllum fabago L. during reclamation of highly contaminated tailings [22]. Better biomass production could be obtained with 5% dry wt. of compost (sewage sludge) + lime [18]. Pig manure could promote microbial biomass and activity, and establishment of native vegetation, independently of the dose [25]. In summary, organic amendments can change metal speciation (soluble, extractable, residual, insoluble) in soil to make them accessible or unavailable for plants according to the applied doses. A classification of organic amendments is then necessary in order to accommodate the selected type along with the appropriate plants for tailings reclamation.

Discussion

Application of organic amendments coupled to phytostabilization is site-specific and should be adapted to the addressed contaminants in tailings (acidic/neutral, low/highly contaminated, metals/metalloids). The selection criteria consider principally the capacity of organic amendments to mobilize/immobilize metals according to their quality including contents in metals, nature (fresh *vs* mature), nutrients (particularly N and P), OM, DOC, humification degree, the mode of application (alone or in mixture), C/N ratio, dosage, and the appropriate type of vegetation. Even though biosolids and manures are the two major sources of metals/metalloids among organic amendments [5], the use of animal manures could be advantageous since the diversity of contaminants might be lower. Biosolids might contain numerous metallic elements (e.g., Al, Fe, Cu, Mn, Zn, Ni, Cr, Pb in sewage sludge), whereas contaminants encountered in manures only originate from additives in industrial animal diet (e.g., As to prevent coccidiosis in poultry or Cu to treat weakness of dairy cattle) [5, 14].

Organic amendments alone would be appropriate for tailings reclamation in single metal contamination rather than for multi-elements. Compost alone could be used to reclaim slightly acidic to neutral and marginally contaminated tailings (at >15% dose) bud should be combined with woody plants (e.g., willow species) instead of grasses. However, the concern about using compost, as well as pig slurry, is the leaching of As. Nonetheless, compost released lower As than pig slurry due to its low mineralisable organic-C [56]. Biochar combusted at lower temperature (<500°C) and produced with a mixture of manures and residues would be performant due to its high OM (≈50%) and carbon contents [35, 36, 53]. Previous studies also reported that low pyrolysis temperature biochars (cottonseed hull combusted at 350°C) contain high oxygen, available P, K, and Ca, and could immobilize Cu, Ni, Cd and Pb [5, 6, 49, 57]. Application of organic amendments in mixture, either between organic amendments or with inorganic materials for an additional alkalinity, is recommended. Biochars would be more efficient when combined with other raw organic amendments such as compost or aged manure for extremely acidic and highly contaminated tailings reclamation [17]. A combination of fresh compost and biosolids could increase contaminants input. Inorganic amendments, particularly limestone, are advantageous as during OM mineralization, the liberation of CO₂ enhances CaCO₃ dissolution and metal carbonates formation (e.g., ZnCO3 and PbCO3) [58]. Limestone combined with organic amendments also moderates the buffering of pH (and high OM contents) if used to reclaim slightly acidic tailings, as well as its capacity to immobilize Mn by precipitation and complexation [18]. Biochar could also be used as alkaline material substitute but a high increase in pH could mobilize metallic elements in tailings such as As (III) [36]. Anaerobic and high quality biosolids mixed with inorganic materials could be applied only for low Fe and Al contaminated tailings [8].

Mature and stable organic amendments, with a higher humification degree (thus low DOC) should be used in order to obtain stable chelate, preventing metals transfer to shoots [27]. Thus, composted organic amendments are suggested rather than raw ones because they have more stable OM, have higher CEC, and limited spread of pathogens [51, 59]. For example, olive residues with a revegetation would be beneficial providing it is composted, thus with lower phytotoxic organic compounds [27]. And yet, mature compost is preferred to immature one because the latter could still contain toxic compounds and highly degradable OM, increasing DOC. Moreover, the decomposition of OM in raw, or immature organic amendments would be in process until stabilization, decreasing oxygen concentration and hence ORP in soil and create a reducing environment in soil [51]. Furthermore, immature organic amendments could contain high amounts of organic acids, such as fulvic acids which could act as chelating agents, entailing metals liberation in pore water [29, 36]. Nonetheless, the high dissolved OM in immature organic amendments, particularly composts, would be an advantage for microbial activity [60], but plant- soil-microbe interactions deserve further studies. Stable organic materials contain recalcitrant organic-C but should be combined with other organic amendments source of labile organic compounds in order to promote activation of microbial populations [13].

Biosolids would better provide nutrients followed by manures, compost, olive processing solid waste, green wastes, peat and, finally, biochar. Higher OM could be found in peat, and then biosolids, manure, olive processing olive solid waste, compost. Organic amendments with high DOC and P should be carefully selected when reclaiming As-contaminated tailings. Balancing the mixture of C-rich (e.g., grape residues, olive residues) and N-rich organic amendments (e.g., manure, biosolids) should be performed in order to ensure the optimal C/N ratio for an effective reclamation [27, 61]. The recommended value to enhance soil fertility and plant persistence was a C/N ratio of 20-30 [27].

Selection of organic amendments doses must be done according to the tailings characteristics (acidic/neutral, highly/slightly contaminated, fresh/oxidized) for an efficient reclamation [27, 33]. Around $10\%_{dw}$ of biochar could be optimal and should be lower than the accompanying organic amendments dose to ensure plant development. The dose of alkaline materials could be calculated according to the equivalent amount of CaCO₃ required for the neutralization of acidity during the reclamation of extremely acidic tailings by using the method of Sobek et *al.* [62]. Addition of limestone at a dose of 55-75 t/ha would increase the pH at around 4 for tailings having initial pH of 2 while a dose >100 t/ha would be necessary to increase the pH \approx 7 depending on the used alkaline materials (marble waste, biochar, hydrated lime) [25, 32, 63].

Vegetation planted for tailings reclamation should be native and metallophytes (metal tolerant plants not accumulating metals to root tissues) [8]. Among the plant species frequently selected for phytostabilization are the type grass, such as *Festuca rubra* L. which has the capacity to translocate low metals concentration from root to shoot [19]. Grass species should be used only for low contaminated tailings or as a nurse crop [64]. For example, rye grass would not be a good candidate for acidic and extremely contaminated tailings [29]. Woody species could be used for slight and moderately contaminated tailings, but also to reclaim highly contaminated tailings; for example, *Salix Alba* L. would be suitable vegetation for Pb stabilization [31]. Coniferous species (jack pine, black spruce) would better survive to moderately saline condition (compost) than broadleaved trees [16]. Plant-tailings-microbes interactions should be considered in order to ensure long-term stabilization of metal-contaminated tailings. Indeed, a better follow up of the reclamation performance would be warranted with the evaluation of the enzymes, indicators for soil activity that could be released not only by plant roots but also by microorganisms [25].

Conclusion

In summary, the preliminary inventory from this study showed the following: 1) mixing organic amendments with inorganic materials, along with revegetation, was found successful for the reclamation of slightly contaminated tailings. Using mature and composted organic amendments is suggested (e.g., a combination of mature and composted animal manures with inorganic materials might be the most promising option) ; 2) anaerobic and high quality biosolids (low concentration of contaminants) mixed with inorganic materials could be applied only for low Fe and Al contaminated tailings; 3) multi-element contaminated tailings could be remediated with blended industrial (e.g., biochar) and raw organic amendments but phytostabilization is still a challenge; and 4) mixtures of compost and biosolids could mobilize mostly metalloids and might increase shoot metal concentrations in plants; 5) almost all organic amendments increased leaching of metalloids (particularly As) in tailings pore water. Future studies should then address the following research needs: 1) the evaluation of the stability of organic amendments, 2) the combined effects of plant and amendment doses on metal speciation and microorganisms communities in rhizosphere, and 3) the long-term (>10-20 y) monitoring of reclaimed tailings performance.

Acknowledgements

This study was funded by the NSERC (Natural Sciences and Engineering Research Council of Canada) and the industrial partners of the RIME UQAT-Polytechnique Montreal, including Agnico Eagle, Canadian Malartic Mine, Iamgold, Raglan Mine-Glencore, and Rio Tinto.

References

- [1] Blowes, D.W., Ptacek, C.J., Jambor, J.L., Weisener, C.G.: The geochemistry of acid mine drainage. In: Treatise on Geochemistry, Holland, H.D., Turekian K.K. (Eds.) 9, 149-204 (2003)
- [2] Aubertin, M., Bussière, B., Pabst, T., James, M., Mbonimpa, M.: Review of the reclamation techniques for acid-generating mine wastes upon closure of disposal sites. In Proc. of the Geo-Chicago: Sustainability, Energy and the Geoenvironment, August 14-18, Chicago, 16p. (2016)
- [3] Genty, T., Bussière, B., Paradie, M., Neculita, C.M.: Passive biochemical treatment of ferriferous mine drainage: Lorraine mine site, Northern Quebec, Canada. In: Proc. of the International Mine Water Association (IMWA). July 11-15, Leipzig, Germany (2016)
- [4] Skousen, J., Zipper, C.E., Rose, A., Ziemkiewicz, P.F., Nairn, R., McDonald, L.M., Kleinmann, R.L.: Review of passive systems for acid mine drainage treatment. Mine Water Environ. 36 (1), 133-153 (2017)
- [5] Park, J.H., Lamb, D., Paneerselvam, P., Choppala, G., Bolan, N., Chung, J.-W.: Role of organic amendments on enhanced bioremediation of heavy metal (loids) contaminated soils. J. Hazard. Mater. 185, 549-574 (2011)
- [6] Bolan, N., Kunhikrishnan, A., Thangarajan, R., Kumpiene, J., Park, J., Makino, T., Kirkham, M.B., Scheckel, K.: Remediation of heavy metal (loids) contaminated soils- To immobilize or immobilize? J. Hazard. Mater. 266, 141-166 (2014)

- [7] Karna., R.R., Luxton, T., Bronstein, K.E., Redmon, J.H., Scheckel, K.G.: State of the science review: Potential for beneficial use of waste by products for *in situ* remediation of metalcontaminated soil and sediment. Crit. Rev. Env. Sci. Tec. 47 (2), 65-129 (2017)
- [8] Mendez, M.O. and Maier, R.: Phytostabilization of mine tailings in arid and semiarid environments-An emerging remediation technology. Environ. Health Persp. 116 (3), 278-283 (2008)
- [9] Kavamura, V.N. and Esposito, E.: Biotechnological strategies to the decontamination of soils polluted with heavy metals. Biotechnol. Adv. 28, 61-69 (2010)
- [10] Tordoff, G.M., Baker, A.J.M., willis, A.J.: Current approaches to the revegetation and reclamation of metalliferous mine wastes. Chemosphere 41, 219-228 (2000)
- [11] Larney, F.J. and Angers, D.A.: The role of organic amendments in soil reclamation: A review. Can. J. Soil. Sci. 92, 19-38 (2012)
- [12] Pardo, T., Bes, C., Bernal, M.P., Clemente, R.: Alleviation of environmental risks associated with severely contaminated mine tailings using amendments: modeling of trace elements speciation, solubility, and plant accumulation. Environ. Toxicol. Chem. 9999, 1-11 (2016)
- [13] Zornoza, R., Acosta, J.A., Fa, A., Bååth, E.: Microbial growth and community structure in acid mine soils after addition of different amendments for soil reclamation. Geoderma 272, 64-72 (2016)
- [14] Forsberg, L.S., Gustafsson, J., Kleja, D.B., Ledin, S.: Leaching of metals from oxidizing sulphide mine tailings with and without sewage sludge application. Water Air Soil Pollut. 194, 331-341 (2008)
- [15] Smart, D., Callery, S., Courtney, R., The potential for waste-derived materials to form soil covers for the restoration of mine tailings in Ireland. Land Degrad. Dev. 27, 542-549 (2016)
- [16] Larchevêque, M., Desrochers, A., Bussière, B., Cartier, H., David, J.S.: Revegetation of non acidgenerating, thickened tailings with boreal trees: a greenhouse study. J. Environ. Qual. 42 (2), 351-360 (2013)
- [17] Rodríguez-Vila, A., Asensio, V., Forján and Covelo, E.F.: Chemical fractionation of Cu, Ni, Pb, and Zn in a mine soil amended with compost and biochar and vegetated with *Brassia juncea* L., J. Geochem. Explor. 158, 74-81(2015)
- [18] Mingorance, M.D., Franco, I., Rossini-Oliva, S.: Application of different soil conditioners to restorate mine tailings with native (*Cistus ladanifer* L.) and non-native species (*Medicago sativa* L.). J. Geochem. Explor. 147, 35-45 (2017)
- [19] Galende, M.A., Becerril, J., Barrutia, O., Artetxe, U., Garbisu, C., Hernández, A.: Field assessment of the effectiveness of organic amendments for aided phytostabilization of a Pb-Zn contaminated soil. J. Geochem. Explor. 145, 181-189 (2014)
- [20] Shen, Z., Md Som, A., Wang, F., Jin, F., McMillan, O., Al-Tabbaa, A.: Long-term impact of biochar on the immobilization of nickel (II) and zinc (II) and the revegetation of a contaminated site. Sci. Total Environ. 542, 771-776 (2016)
- [21] Yang, S., Cao, J., Li, F., Peng, X., Peng, Q., Yang, Z., Chai, L.: Field evaluation of the effectiveness of three industrial by-products as organic amendments for phytostabilization of a Pb/Zn mine tailings. Environ. Sci: Processes Impacts 18, 95-103 (2016)
- [22] Pardo, T., Bernal, M.P., Clemente, R.: Phytostabilization of severely contaminated mine tailings using halophytes and field addition of organic and inorganic amendments. Chemosphere 178, 556-564 (2017)
- [23] Burger, J.A., and Zipper, C.E.: How to restore forests on surface-mined land: Reclamation guidelines for surface mined land in Southwest Virginia. Publ. 460-123. Virginia Coop. Ext., Arlington (2002)
- [24] Bussière, B.: Colloquium 2004: Hydrogeotechnical properties of hard rock tailings from metal mines and emerging geoenvironmental disposal approaches. Can. Geotech. J. 44 (9), 1019-1052 (2007)
- [25] Zornoza, R., Faz, A., Carmona, D.M., Kabas, S. Martínez-Martínez, S., Acosta, J.A.: Plant cover and soil biochemical properties in mine tailings pond five years after application of marble wastes and organic amendments. Pedosphere 22, 22-32 (2012)
- [26] Zanuzzi, A., Arocena, J. M., VanMourik, J.M., Faz Cano, A.: Amendments with organic and industrial wastes stimulate soil formation in mine tailings as revealed by micromorphology, Geoderma 154 (1-2), 69-75 (2009)
- [27] Santibãnez, C., De la Fuente, M.L., Bustamante, E., Silva, S., León-Lobos, P., Ginocchio, R.: Potential use of organic- and hard-rock mine wastes on aided phytostabilization of large-scale mine tailings under semiarid Mediterranean climatic conditions : Short term field study. Appl. Environ. Soil Sci. Article ID 895817, 15 p. (2012)

- [28] Hwang, T. and Neculita, C.M.: In situ immobilization of heavy metals in severely weathered tailings amended with food waste-based compost and zeolite. Water Air Soil Pollut. 224, 1388 (2013)
- [29] Pardo, J.H., Bernal, M.P., Clemente, R.: Efficiency of soil organic and inorganic amendments on the remediation of a contaminated mine soils: I. Effects on the trace elements and nutrients solubility and leaching risk. Chemosphere 107, 121-128 (2014)
- [30] Lindsay, B.J., Wakeman, K.D., Rowe, O.F., Grail, B.M., Ptacek, C.J., Blowes, D.W., Johnson, D.B.: Microbiology and geochemistry of mine tailings amended with organic carbon for passive treatment of pore water. Geomicrobiol. J. 28: 229-241(2011)
- [31] Lebrun, M., Macri, C., Miard, F., Hattab-Hambli, N., Heino-Motelica, M., Morabito, D., Bourgerie, S.: Effect of biochar amendments on As, Pb mobility and phytoavailability in contaminated mine technosols pytoremediated by *Salix*. J. Geochem. Explor. doi.org/10.1016/j.gexplo.2016.11.016 (2016)
- [32] Santos, E.S., Magalhães, M.C.F., Abreu, M.M., Macías, F.: Effects of organic/inorganic amendments on trace metals dispersion by leachates from sulfide-containing tailings of the São Domingos mine, Portugal. Time evaluation. Geoderma 226-227, 188-2013 (2014)
- [33] Valentín-Varga, A., Root, R.A., Neilson, J.W., Chorover, J., Maier, R. M.: Environmental factors influencing the structural dynamics of soil microbial communities during assisted phytostabilization of acid-generating mine tailings: A mesocosm experiment. Sci. Total. Environ. 500-501, 314-324 (2014)
- [34] Abad-Valle, P., Iglesias-Jiménez, E., Álvarez-Ayuso, E.: A comparative study on the influence of the difference organic amendments on trace elements mobility and microbial functionality of a polluted mine soil. J. of Environ. Manage. 188, 287-296 (2017)
- [35] Fellet, G., Marchiol, L., Vedove, G.D., Peressotti, A.: Application of biochar on mine tailings: Effects and perspectives for land application. Chemosphere 83, 1262-1267 (2014)
- [36] Fellet, G., Marmiroli, M., Marchiol, L.: Elements uptake by metal accumulator species grown on mine tailings amended with three types of biochar. Sci. Total Environ. 468-469, 598-608 (2014)
- [37] Singh, B., Singh, B.P., Cowie, A.L.: Characterisation and evaluation of biochars for their application as a soil amendment. Aust. J. Soil. Res. 48, 516-525 (2010)
- [38] Méndez, A., Gómez, A., Paz-Ferreiro, J., Gascó, G.: Effects of sewage sludge biochar on plant metal availability after application to a Mediterranean soil. Chemosphere 89, 1354-9 (2012)
- [39] Gil-Lioza, J., White, S.A., Root, R.A., Solís-Dominguez, F.A., Hammond, C.M., Chorover, J., Maier, R.M.: Phytostabilization of mine tailings using compost-assisted direct planting: Translating greenhouse results to the field. Sci. Tot. Environ. 565, 451-461 (2016)
- [40] Renault, S., Markham, J., Davis, L., Sabra, A., Szczerski, C.: revegetation of tailings at the Gunmar mine site, Manitoba (NTS52L14): plant growth in tailings amended with paper-mill sludge. Report of activity, Manitoba Geological Survey, p. 161-165 (2007)
- [41] Barajas-Aceves, M and Rodríguez-Vásquez, R.: Effects of organic amendments on the mobility of Pb and Zn from mine tailings added to semi-arid soils. J. Environ. Sci. Heal. B 48, 226-236 (2013)
- [42] Farrell, M. and jones, D.L.: Use of compost in the remediation of heavy metal contaminated soil. J. Hazard. Mater. 175, 575-582 (2010)
- [43] Schwab, P., Zhu, D., Banks, M.K.: Heavy metals leaching from mine tailings as affected by organic amendments. Biores. Technol. 98, 2935-2941 (2007)
- [44] Li, M., Mohamed, I., Raleve, D., Chen, W., Huang, Q.: Field evaluation of intensive compost application on Cd fractionation and phytoavailability in a mining-contaminated soil. Environ. Geochem. Health 36 (5), 1193-1201 (2016)
- [45] Santibáñez, C., Verdugo, C., Ginocchio, R.: Phytostabilization of copper mine tailings with biosolids: Implications for metal uptake and productivity of Lolium perenne. Sci.Total Environ., 395, 1-10 (2008)
- [46] Zheng, R.-L., Cai, C., Liang, J.-H., Huang, Q., Chen, Z., Huang, Y.-Z., Arp, H.P.H., Sun, G.-X.: The effects of biochars from rice residue on the formation of iron plaque and the accumulation of Cd, Zn, Pb, As in rice (Oryza sativa L.) seedlings. Chemosphere 89, 856-62 (2012)
- [47] Beesley, L., Marmiroli, M., Pagano, L., Pigoni, V, Fellet, G., Fresno, T., Vamerali, T., Bandiera, M., Marmirolu, N.: Biochar addition to an arsenic contaminated soil increases arsenic concentrations in the pore water but reduces uptake to tomato plants (*Solanum lycopersicum* L.). Sci. Total Environ. 454-455, 598-603 (2013)
- [48] Guittonny-Larchevêque, M. and Pednault, C. Substrate comparison for short-term success of a multispecies tree plantation in thickened tailings of a boreal gold mine. New Forests 47(5), 763-781 (2016)

- [49] Angers, D.A. and Caron, J.: Plant-induced change in soil structure: processes and feedbacks. Biogeochemistry 42 (1), 55-72 (1998)
- [50] Cao, R.X., Ma, L.Q., Chen, M., Singh, S.P., Harris, W.G.: Phosphate-induced metal immobilization in a contaminated site. Environ. Pollut. 122, 19-28 (2003)
- [51] Jiménez, E.I. and Garcia, V.P.: Evaluation of city refuse compost maturity: A review. Biol. Wastes 27, 115-142 (1989)
- [52] Garcia, C., Hernandez, T., Costa, C., Ayuso, M.: Evaluation of the maturity of municipal waste compost using simple chemical parameters. Commun. Soil Sci. Plan. 23 (13-14), 1501-1512 (1992)
- [53] Lindsay, M.B.J., Blowes, D.W., Condon, P.D., Lear, K.G., Ptacek, C.J.: Evaluation of organic carbon amendments for passive in situ management of tailings pore-water quality. In Proc. of the 8th International Conference on Acid Rock Drainage, June 23-26, Skellefteå, Sweden. 10 p. (2007)
- [54] Ahmad, M., Rajapaksha, A.U., Lim, J.E., Zhang, M., Bolan, N., Mohan, D., Vithanage, M., Lee, S.S., Ok, Y.S.: Biochar as a sorbent for contaminant management in soil and water: A review. Chemosphere 99, 19-33 (2014)
- [55] Forján, R., Asension, V., Rodríguez-Vila, A., Covelo, E. F.: Contribution of waste and biochar amendments to the sorption of metals in a copper mine tailings. Catena 137, 120-125 (2016)
- [56] Pardo, T., Clemente, R., Bernal, M. P.: Effects of compost, pig slurry and lime on trace element solubility and toxicity in two soils differently affected by mining activities. Chemosphere 84, 642-650 (2011)
- [57] Uchimiya, M., Chang, S., Klasson, K.T.: Screening biochars for heavy metal retention in soil: role of oxygen functional groups. J. Hazard. Mater. 190, 432-441 (2011)
- [58] Usman, A.R.A., Kuzyakov, Y., Stahr, K.: Dynamics of organic C mineralization and the mobile fraction of heavy metals in a calcareous soil incubated with organic wastes. Water Air Soil Pollut. 158, 401-418 (2004)
- [59] Bernal, M.P., Alburquerque, J.A., Moral, R.: Composting of animal manure and chemical criteria for compost maturity assessment. A review. Bioresource Technol. 100, 5444-5453 (2009)
- [60] Zmora-Nahom, Z., Markovitch, O., Tarchitzky, J., Chen, Y.: Dissolved organic carbon (DOC) as a parameter of compost maturity. Soil Biol. Biochem. 37, 2109-2116 (2005)
- [61] Brown, S.L., Henry, C.L., Chaney, R., Compton, H., DeVolder, P.S.: Using municipal biosolids in combination with other residuals to restore metal-contaminated mining areas. Plant Soil 249, 203-215 (2003)
- [62] Sobek, A.A., Schuller, W.A., Freeman, J.R., Smith, R.M.: Field and laboratory methods applicable to overburdens and mine soils. EPA-600/2-78-054 (1978)
- [63] Muñoz, M.A., Guzman, J.G., Zornoza, R., Moreno, F., Faz, A., Lal, R., Effects of biochar and marble mud on mine waste properties to reclaim tailings ponds. Land Degrad. Develop. 24, 1227-1235 (2016)
- [64] Yuanyuan, Z., Junhong, L. Yuanming, Z., Tingyun, G., Jing, W., Yinlin, G.: Enhanced phytoremediation of mixed heavy metal (mercury)–organic pollutants (trichloroethylene) with transgenic alfalfa co-expressing glutathione S-transferase and human P450 2E1. J. Hazard. Mater. 260, 1100-1107 (2013)