Paper mill sludge – application in geoenvironmental engineering

Lidiane da Silva Ibeiro Knop
Federal University of Rio Grande do Sul, Rio Grande do Sul, Brazil
Alexandre Knop[a], Nilo Cesar Consoli[b] & Karla Salvagni Heineck[b]
[a]*UMA Engineering Ltd, Winnipeg, Manitoba, Canada*
[b]*Department of Civil Engineering, Federal University of Rio Grande do Sul, Rio Grande do Sul, Brazil*

ABSTRACT
The objective of this research is to contribute to the development of new geotechnical materials, by evaluating geotechnical properties of a paper mill sludge from an industry located in Porto Alegre, southern Brazil. For the evaluation, hydraulic conductivity with different molding moisture contents, a compaction test, organic matter, specific gravity, Atterberg limits, electronic microscopy and x-ray fluorescence tests were complete. Results show that the paper mill sludge has low hydraulic conductivity coefficients when compacted under high moisture contents (between 160 and 190%), approximately 10^{-7} m/s.

RÉSUMÉ
L'objectif de cette recherche est de contribuer a le développement de nouveau matériel geotechnical, en évaluant les propriétés geotechnical du cambouis de moulin à papier, d'une industrie trouvée dans Porto Alegre, au sud du Brésil. Pour l'évaluaer, la conductivité hydraulique avec des différentes teneurs en humidité de moulures, un test de compaction, matériel organique, la densité, les limites d'Atterberg, la microscopie électronique et les épreuves de fluorescence de rayon-x étaient complets. Les résultats prouvent que le cambouis de moulin à papier a de bas coefficients hydrauliques de conductivité une fois compact sous la haute teneur en humidité (entre 160 et 190%), autour 10^{-7} m/s.

1 INTRODUCTION

During the last couple of decades, due to the large increase of the industrial production in all sectors, the generation of wastes and by-products of these production activities have also increased. Researchers around the world have intended to develop technologies to correctly treat these residues, or even better, to transform a waste material in a potential alternative material that can be used in geotechnical engineering.

Internationally, the re-use of waste materials is understood as being a correct way to minimize storage problems. It is a consensus between researchers that alternative materials such as production wastes should be re-used as alternative products in engineering in order to reduce the depletion of our natural resources. Contributing to this scenario, the paper industry is responsible by the generation of high amounts of sludge every day. The use and production of paper is increasing year by year around the world. In 2006, the per-capita consumption of paper in USA, Canada and Brazil was 300, 213 and 41 kg/habitant/year, respectively (Bracelpa, 2008).

The availability of paper mill sludge is very significant around the world. According to Cabral et al. (2000), in Quebec, Canada, the annual generation of paper mill sludge is estimated to be around 500,000 tons with high moisture content.

Due to these and other reasons, it is understood that paper mill sludges should receive more attention by the researchers in developing technologies for their use in geotechnical and geoenvironmental engineering.

Previous studies have already shown the technical potential of paper sludges when used as geotechnical material, mainly due to their important characteristics in terms of behavior: capacity of reaching very low hydraulic conductivity coefficients (Ibeiro, 2007) and the capacity to retain water. These characteristics is what make this material able to be used as a oxygen barrier in cover of tailings, in order to reduce the acid mine drainage generation (Cabral et al., 2002).

According to several authors (Moo-Young & Zimmie, 1996; Kraus et al, 1997; Cabral et al, 2002), paper mill sludges seem to have a satisfactory performance in terms of geotechnical parameters when used in cover liners, being an economic and attractive alternative in substitution of clayey liners.

A good water retention capacity of the material helps to keep the degree of saturation high, which in turn reduces the flux of oxygen (O_2) by diffusion (Cabral et al., 2000). Acid mine drainage can be curbed or reduced significantly by covering tailings sites with an O_2 barrier. Moreover, oxygen consumption by biodegradation leads to undetectable O_2 fluxes just 0.3 m below the surface.

A trial field experiment with paper mill sludge has been carried out in Manitoba, Canada. This research aims to determine the effects of paper mill sludge on the establishment and growth of vegetation on the top of tailings (Renault et al., 2007).

According to Calace et al. (2005), the use of paper mill sludge could be suggested as a good practice for environmental remediation. The paper mill sludge could be effective due to the organic matter, silicate and carbonate contents. The organic matter is able to form stable complexes with several metals; the silicates are materials of high cation exchange capacity (CEC) and the bicarbonate / carbonate system is able to increase the pH value of soil. These chemical features were able to reduce the harmful mobile metals in polluted soils when the paper mill sludge was added to them.

According to Calace et al. (2005), paper mill sludge is generated by various processes in the production of pulp and in the manufacturing of paper, and increasing quantities produced make the disposal of this sludge a problem for the pulp and paper industry. Consequently, its utilization to “remediate” a contaminated soil could be suggested, once paper mill sludges do not have any toxic effect to the environment.

The main objective of this research is, from a geotechnical point of view, to investigate the paper mill sludge properties from a paper industry from Porto Alegre, Southern Brazil, in terms of hydraulic behavior when submitted to different conditions as well as physical properties of the sludge.

2 METHODOLOGY

The waste from the recycling paper industry has a format of sludge, high water content and consequent high compressibility. The moisture content is very high right after the production, around 256 %, and it is basically a mixture of cellulosic fibers, minerals and water.

The average sludge generation at the paper industry selected for this research is approximately 13 m³/day, and it is estimated that 1 m³ of sludge is generated for each ton of paper.

Generally, paper mill sludge generated in different industries has similar compositions and physical characteristics. However, different production procedures, sludge treatments and raw-materials used in the paper production can affect the final sludge properties.

Based on tests and regulations initially applied to soils, this research focused the physical-chemical characterization of the sludge as well as the investigation of the hydraulic behavior under different situations.

The following tests were carried out during the experimental program of this work: Specific gravity of solids, Atterberg Limits, organic matter content, compaction test (Standard Proctor), Scanning Electron Microscopy (SEM), x-ray fluorescence and hydraulic conductivity tests. This experimental program is part of a full set of tests and analyses completed in order to investigate mechanical and hydraulic characteristics of the paper mill sludge.

In order to keep the field conditions in terms of moisture content, the sludge was stored under low temperatures in a refrigerator. Previous to each test, the sludge was sieved through a 4.75 mm sieve in order to reproduce homogeneous samples (Figure 1).

In order to reach the specified moisture content for the tests, the sludge was allowed to dry at the ambient temperature until the aimed moisture content was reached, which means that no water was added to the samples.

Figure 1 – Wet sludge sieved through a 4.75 mm sieve.

When exposed to air without a previous sieving, the sludge tended to become heterogeneous conglomerates with different sizes, depending on the randomly distribution of the fibers, as shown in Figure 2.

Figure 2 – Paper mill sludge with natural moisture content (256%).

The specific gravity of solids and Atterberg limits tests were completed according to the following Brazilian standards: NBR 6508 (ABNT, 1984); NBR 6459 (ABNT, 1984) and NBR 7180 (ABNT, 1984).

The amount of organic matter was determined using a stove at temperatures around 440℃, according to NBR 13.600 (ABNT, 1996).

Standard Proctor compaction tests were carried out, according to the Brazilian standard NBR 7182 (ABNT, 1986). Differently from the usual procedure (from the dry side to the wet side of the compaction curve), the tests were completed starting with very wet samples, then decreasing the moisture content gradually until the dry-side was reached. This procedure was adopted not only due to the high natural moisture content of the paper sludge, but also because the material becomes heterogeneous (conglomerates) under low moisture contents. When dried, it does not return to its original condition, even under the same moisture content (by adding water), because these conglomerates become very strong when in dry conditions, and this new structure is not reversible just by increasing the moisture content.

The microstructure of the sludge was assessed by scanning electron microscopy (SEM) tests, and the identification of the elements present in the composition of the sludge was assessed by energy dispersive spectroscopy (EDS) analysis.

X-Ray fluorescence tests were completed for the evaluation of chemical compositions and concentration of elements in the sludge. Samples used for these tests and for Scanning Electron Microscopy (SEM) tests were obtained by burning the material at 440℃ (the burning was carried out for the organic matter content tests).

The hydraulic conductivity of the compacted material was determined during conventional triaxial tests (CIU), after the consolidation period, under a hydraulic gradient of 10 (according to ASTM D 5084, 1990). Results of triaxial tests are not covered by this article. For these tests, the samples were statically compacted in cylindrical molds 100 mm high and 50 mm in diameter, and submitted to an effective stress of 20 kPa during the consolidation phase. The molding moisture contents for the tests were 197 %, 163 % and 133%.

3 RESULTS AND COMMENTS

3.1 Index Properties

Table 1 introduces results of characterization tests completed on the paper mill sludge.

Table 1. Paper Mill Sludge – Properties.

Specific Gravity of solids	1.862
Liquid Limit (%)	210
Plastic Limit (%)	124
Moisture Content (%)	256
Organic Content (%)	39 – 48

It is observed that the sludge has high coefficients (LL, PL, Moisture Content and Organic matter content) if compared to a soil. An important factor is that the natural moisture content of the paper sludge (256%) is higher than the Liquid Limit (210%). These results indicate that Atterberg Limit tests represent a behavior different from the expected from a test on a conventional material (such as clay). Even under the natural moisture content (which is higher than the Liquid Limit), the material does not have a liquid behavior. It is understood that this distinct result took place due to the random arrangement of the fibers of the material.

The experimental program of this research was considered very relevant in trying to make comparisons of this new material to other materials previously studied in the literature (such as soils and other waste materials).

According to data shown in Table 1, organic matter contents of different samples sampled in different dates range from 39 % to 48 %, and these high contents are due to the presence of cellulosic fibers (which are a source of carbon). The small range of values shows that the industrial process is uniform, which generates sludge with very similar conditions over the time.

3.2 Compaction Test

The paper sludge was compacted under different moisture contents, but it was not possible to compact the sample with the natural moisture content due to the high water content of the material under this condition.

According to Figure 3, the optimum moisture content and maximum dry unit weight of sludge mass (γ_{dmax}) obtained from the compaction test (standard Proctor) for the paper sludge is 110% and 5.3 kN/m^3 respectively. However, these parameters were not used for molding the samples because under this condition (γ_{dmax} and optimum moisture content) there is no workability of the material. Also, samples molded with moisture contents close to the optimum do not have a satisfactory homogeneity.

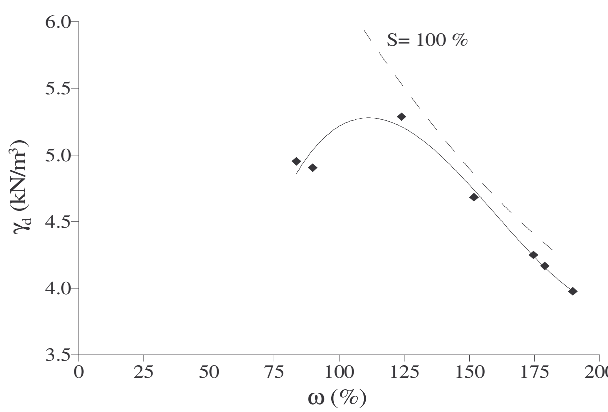
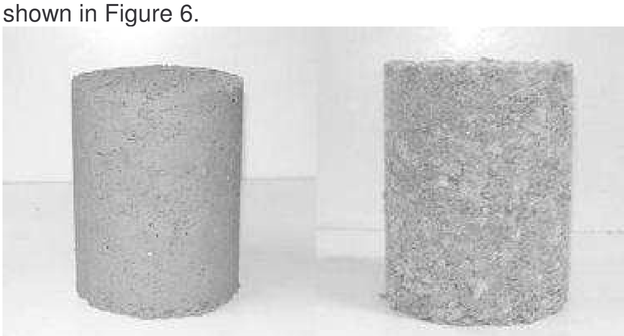


Figure 3. Compaction curve of the paper sludge

It is possible to observe a sample with 150% of moisture content in Figure 4 (a), and a sample with moisture content of 125% (close to the optimum) in Figure 4 (b). Samples molded with moisture content lower than 125% had no consistence as soon as they were removed from the mold.

3.3 Scanning Electron Microscopy (SEM)

Figure 5 show the image of a sample analyzed by a scanning electron microscopy with 160 times magnification. In Figure 5, it is possible to observe 2 areas called area 1 and area 2. These areas are the locations where the quantitative evaluation by means of Energy Dispersive Spectroscopy was completed, as shown in Figure 6.



a) Sample with w = 150 % b) Sample with w = 125 %

Figure 4. Samples compacted with different moisture contents

It is possible to observe in Figure 5 three-dimensional elements formatted by fibers and grains with aspect of a matrix. Fibers randomly present in the sample are up to 600 μm long. Fibers shorter than 100 μm tend to arrange themselves around a nucleus. It is also observed the presence of a mineral plate (area 2). During the paper manufacturing, minerals are added to the fibers mass in order to improve the density and mechanical strength of the mixture.

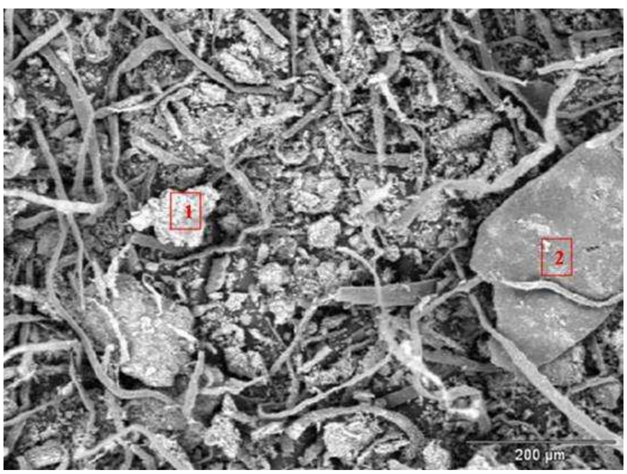
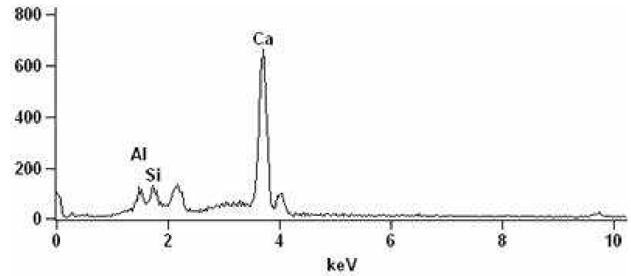


Figure 5. Result of Scanning Electron Microscopy (SEM)

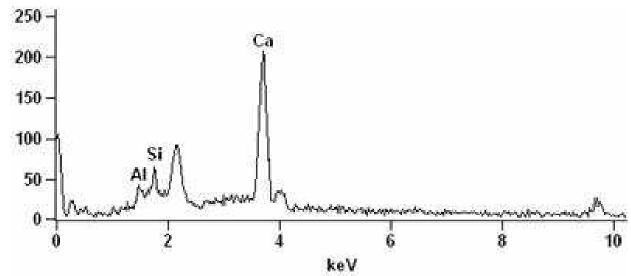
In Table 2, elements contents in the sludge according to results of Energy Dispersive Spectroscopy EDS for area 1 and 2 (Figure 5) are shown.

The silicon and aluminum contents (as shown in Table 2) suggest the presence of clay-mineral kaolinite in the sludge; also, the peaks of calcium in the spectrums (Figure 6) suggest the presence of calcium carbonate.

In Figure 6, Energy Dispersive Spectroscopy (EDS) results corresponding to the areas 1 and 2 (Figure 5) are presented.



EDS – Area 1



EDS – Area 2

Figure 6. Energy Dispersive Spectroscopy (EDS) – Areas 1 and 2 shown in Figure 5.

Table 2. : Elements contents according to EDS test (%)

Area	Element		
	Al	Si	Ca
1	6.69 %	8.50 %	84.81 %
2	5.33 %	9.54 %	85.13 %

3.4 X-Ray Fluorescence Tests

In Table 3 the results of chemical analysis of inorganic elements for samples submitted to the temperature of 440°C are shown.

According to Table 3, it is possible to observe that the five samples have similar results, showing that the material has a uniform composition. SiO_2 and Al_2O_3 oxides indicated the presence of clay-minerals such as kaolinite. Low K_2O contents indicate the presence of illitic clay-minerals. The presence of high contents of CaO and fire loss percentage around 30% corresponds to values characteristic of carbonate minerals. In this case, MgO contents smaller than 2% suggest that the carbonate material is basically formatted by kaolinite.

Analytical results from x-ray fluorescence tests confirm the results obtained from scanning electronic microscopy (Figure 6).

Table 3: Chemical composition (%) of paper mill sludge samples.

Element	Sample 1	Sample 2	Sample 3	Sample 4	Sample 4
SiO_2	16.66	18.77	19.93	15.79	16.01
Al_2O_3	15.64	15.38	16.45	13.72	13.65
TiO_2	0.34	0.47	0.32	0.38	0.28
Fe_2O_3 (total)	0.73	0.67	0.85	0.58	0.6
MnO	0.01	0.01	0.02	0.01	0.01
MgO	1.64	1.94	1.87	1.78	1.8
CaO	35.53	34.16	33.3	36.87	37.36
K_2O	0.2	0.19	0.21	0.15	0.15
P_2O_5	0.14	0.1	0.14	0.11	0.11
Fire Loss	29.8	28.87	28.61	30.85	30.85

3.5 Hydraulic Conductivity tests

Table 4 shows the molding moisture content (ω) of samples submitted to hydraulic conductivity tests, as well as voids ratio (e), dry unit weight (γ_d) at the beginning of the tests and after the consolidation phase, and the volumetric strain (ΔV) during the consolidation phase. The measurements of the hydraulic conductivity were completed during triaxial tests, after the consolidation period. Results of triaxial tests are not analyzed in this article. It is observed that a considerable variation of the void ratio during the consolidation phase exists, which can be related to the high moisture contents.

Table 4. Variation of the physical indexes previously and after consolidation.

ω(%)	γ_d (kN/m^3) Initial	e Initial	γ_d (kN/m^3) Final	e Final	ΔV (%)
197	3.42	4.45	3.84	3.84	11.0
163	4.06	3.58	4.41	3.22	7.8
133	4.62	3.03	4.92	2.78	6.1

The variation of hydraulic conductivity coefficient (k) in samples molded under three different molding moisture contents and effective stress of 20 kPa is shown in Figure 7. Hydraulic conductivity coefficients were measured after 4 hours after the consolidation phase, time required for total stabilization of variations in k.

According to Figure 7, it is possible to observe that samples tested with higher moisture contents had lower hydraulic conductivities, under an effective confining stress of 20 kPa.

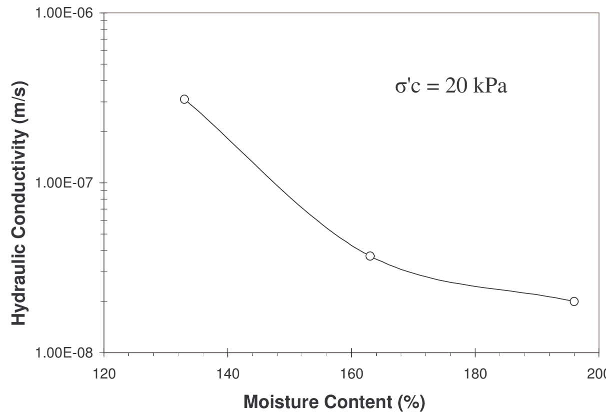


Figure 7. Hydraulic conductivity of the paper sludge versus molding moisture content.

The measured hydraulic conductivity of a sample with 197% of molding moisture content was around $2.5\text{x}10^{-8}$ m/s. Hydraulic conductivity at the same order of magnitude was also observed for the sample with 163% of moisture content. With 133% of moisture content, the hydraulic conductivity was one order of magnitude higher than previous samples, around $2.3\text{x}10^{-7}$ m/s.

It is believed that the variation of hydraulic conductivity values between the tree samples occurs due to fabric arrangements in which the particles are submitted by compaction under different moisture contents.

It was observed that under the lowest moisture content evaluated in this research, the paper mill sludge has a granular aspect, where the fabric is relatively more uniform and compact than under higher moisture contents.

Usually, clayey barriers are used to protect landfills from rainfalls and snow melting, reducing the leachate volume. In tailings, dry covers protect the wastes from oxidation by reducing the amount of oxygen and/or the presence of water. Clayey barriers are used because they have low hydraulic conductivity coefficients. However, in some situations, clayey soils are not available and the use of alternative materials should be considered as an option.

There are requirements that materials considered for cover systems must meet, in order to guarantee a satisfactory field performance and remediation efficacy. Usually these requirements are specified by standards or government regulations, and they can change according the hazard potential of the contamination to the environment.

For cover liners of domestic landfills, the maximum hydraulic conductivity of the material (usually clayey soils) is $1\text{x}10^{-7}$ m/s, according to La Grega et al. (2001). The values of hydraulic conductivity of all the samples of paper mill sludge studied in this research, under 3 different moisture contents, reached coefficients lower than 10^{-7} m/s.

4 CONCLUSIONS

The main conclusions of this study are as follows:

- The paper mill sludge evaluated in this research is basically formatted by cellulosic fibers, minerals and water. The water content of the sludge immediately after the paper manufacturing process is 256%;
- Once dried, the paper mill sludge fibers randomly rearrange and the structure of the sludge becomes very strong, formatting very strong conglomerates with a granular aspect;
- In terms of compaction, the sludge has a high compressibility and the best workability occurred under the highest moisture content (around 190 %);
- Hydraulic conductivity coefficient (k) for samples molded with 197% and 163% of moisture content was around 10^{-8} m/s; For the lowest moisture content of this study (133%), k was around 10^{-7} m/s;
- The ability of reaching very low hydraulic conductivity values is an important characteristic of the behavior of the paper sludge, which indicates that this material can possibly be used as an alternative geotechnical material, especially as a substitute of usual materials used in cover liners.

ACKNOWLEDGEMENTS

The authors wish to express their gratitude to Pronex-Fapergs (Process 04/0841.0), MCT/CNPq and MEC/CAPES for the financial support, and UMA Engineering Ltd, Winnipeg, Manitoba, Canada for the technical support.

REFERENCES

ASTM D 5084. 1990. Standard test method for measurement of hydraulic conductivity of saturated porous materials using a flexible wall permeameter. American Society for Testing and Materials, Pennsylvania, USA, 9p.

Bracelpa – Brazilian Association of Cellullose and Paper. 2008. Internet acess on April 20, 2008. www.bracelpa.org.br

Cabral, A. R.; Racine, I.; Burnotte, F.; Lefebvre, G.; 2000. Difusion of oxygen through a pulp and paper residue barrier. Canadian Geotechnical Journal, 37: 201 - 217.

Cabral, A. R.; Burnotte, F.; Lefebvre, G.; Panarotto, C. T. 2002. Geotechnical characterization of a pulp and paper (deinking) residue used in cover systems. In: Environmental Geotechnics, 207-212.

Calace, N.; Campisi, T.; Lacondini, A.; Leoni. M.; Petronio, B.M.; Pietroletti, M. 2005. Metal-contaminated soil remediation by means of paper mill sludges addition: chemical and ecotoxicological evaluation. Environmental Pollution 136 (2005) p.485-492.

Ibeiro, Lidiane da Silva. 2007. Geotechnical characterization of a paper mill sludge from a Brazilian Industry. M.Sc. thesis, Civil Engineering, Federal University of Rio Grande do Sul, Brazil.

Kraus, J.; Benson, C.; Van Maltby, C.; Wang, X. 1997. Laboratory and field hydraulic conductivity of three compacted paper mill sludges. In: Journal of Geotechnical and Geoenvironmental Engineering, 123 (7), 654-662.

La Grega, M. D.; Buckingham, P. L.; Evans, J. C. 2001. Hazardous Waste Management, 2nd Ed., McGraw-Hill, 2001, 1202 pages.

Moo-Young, H. K.; Zimmie, T. F. 1996. Effects of organic decomposition on paper mill sludges used as landfill cover material. In: Environmental Geotechnics, 827-832.

NBR 6459. 1984. Soil – Determination of the Liquid Limit. Brazilian Association of Brazilian Standards. Brazil, 1984. In Portuguese.

NBR 7180. 1984. Soil – Determination of the Plastic Limit. Brazilian Association of Brazilian Standards. Brazil, 1984. In Portuguese.

NBR 6508. 1984. Soil – Determination of the Density of Solids – Grain Size Distribution – sieve 4.8mm. Brazilian Association of Brazilian Standards. Brazil, 1984. In Portuguese.

NBR 7182. 1986. Soil – Compaction Test. Brazilian Association of Brazilian Standards. Brazil, 1986. In Portuguese.

NBR 13600. 1996. Soil – Determination of the Organic Matter by Burning at 440°C. Brazilian Association of Brazilian Standards. Brazil, 1996. In Portuguese.

NBR 10004. 2004. Solid Wastes – Classification. Brazilian Association of Brazilian Standards. Brazil, 2004. In Portuguese.

Renault, S.; Markham, J.; Davis, L. Sabra, A. Szczerski, C. (2007) Revegetation of tailings at the Gunnar minesite, Manitoba (NTS 52L14): plant growth in tailings amended with paper-mill sludge. In: Report of Activities 2007, Manitoba Science, Technology, Energy and Mines, Manitoba Geological Survey, p. 161–165.