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Use of industrial pulp and paper waste as agricultural inputs: A systematic literature review[1]

Utilização de resíduos industriais de celulose e papel como insumos agrícolas: Revisão sistemática de literatura

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HIGHLIGHTS:

Several benefits to the soil were identified after the application of pulp and paper mill residue.
Organic and inorganic residue application increased soil fertility and promoted pH control.
Residue application in the field complied with environmental requirements.

ABSTRACT: Industrial waste disposal in landfills is a challenge for the pulp and paper industry, and its use as fertilizers and soil conditioners has exhibited potential as a new alternative destination. As such, the present study aimed to identify factors that affect this alternative use for these residues and validate its technical, legal and economic feasibility through a systematic literature review based on the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) protocol. In general, applying wastewater treatment plant sludge, green liquor dregs and biomass ashes in the field has been shown to increase soil fertility, control its acidity, and maintain toxic metal content within the recommended limits. This enables the environmental leveraging of the pulp and paper sector by providing an appropriate alternative destination for industrial waste and promotes circularity in forest and agricultural management. However, no conclusive studies on the economic aspects of this alternative application were found, demonstrating the importance of conducting comprehensive research to confirm its economic feasibility.

Key words: soil fertilization, pulp and paper mill waste, waste reuse for liming

RESUMO: A disposição de resíduos sólidos em aterros sanitários é um grande desafio para o setor de celulose e papel, e sua utilização como fertilizantes e corretivos de solo tem apresentado potencial como uma nova alternativa de destino. Assim, este trabalho objetivou identificar os fatores que influenciam esta alternativa de destinação dos resíduos e validar sua viabilidade técnica, legal e econômica através de uma revisão sistemática de literatura, de acordo com os principais itens para relatar revisões sistemáticas e meta-análises. De forma geral, foram identificados como benefícios o aumento da fertilidade e controle da acidez do solo através da aplicação de lodo de estações de tratamento de efluentes, resíduos da recuperação do licor verde e cinzas de biomassa, e os teores de metais tóxicos se mantiveram abaixo dos limites da legislação. Estes resultados, além de alavancarem ambientalmente o setor de celulose e papel pela destinação adequada de resíduos industriais, promovem a circularidade no manejo florestal e culturas agrícolas. Em relação aos aspectos econômicos, no entanto, não foram encontrados trabalhos conclusivos, sendo necessário o desenvolvimento de um estudo robusto para comprovação da viabilidade econômica desta aplicação.

Palavras-chave: fertilização de solo, resíduos de papel e celulose, reuso de resíduos para calagem

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Introduction

In 2022, planted forests in Brazil covered an area of 10 million hectares, 76% of which consisted of *Eucalyptus* intended largely for commercial purposes. In this scenario, the pulp and paper sector has grown significantly over the years, reaching 25 million tons of pulp in 2021 (22 million of which was short-fiber pulp), a 10% increase over 2020 (IBÁ, 2023).

In light of this expansion, growing demand from society, and the requirements of environmental agencies for more sustainable practices, the sector has invested in research and technology to optimize processes and reduce waste. As such, solid waste management has become a topic of discussion, since in addition to its environmental aspects, reusing these materials provides economic benefits.

For example, Foelkel (2008) discussed the Cleaner Production concept, which establishes that almost everything that becomes waste is a raw material discarded during the industrial process. Not only is a high-value raw material thrown away, the value added during the industrial process is also discarded, in addition to the costs incurred for waste handling and disposal.

The most common industrial residues produced by the pulp and paper industry are wastewater and water treatment plant sludge, which are primarily organic and contain high levels of nitrogen; inorganic residues from green liquor recovery (such as dregs, grits, and other waste generated in lime kilns), with high pH, calcium, and in some cases, containing sodium; and biomass boiler ash, which is inorganic and contains Wood's metal (Foelkel, 2008). Based on their physical and chemical characteristics, pulp and paper organic and inorganic residues show considerable agronomic potential, mostly associated with improved fertility and pH control.

However, determining the optimal application rate depends not only on their physical and chemical characteristics, but plant nutritional needs and regional soil and climate conditions. This enables residues to be treated and enriched to meet the required technical specifications for each location, preventing environmental damage and maintaining the productivity of forests and other crops.

In addition to technical aspects, the direct costs involved in waste treatment, acquiring inputs to enrich fertilizers and soil conditioners, transportation, and field application could hinder the use of organic waste as fertilizer and should be carefully evaluated. Furthermore, it is important to understand the environmental parameters of each residue and the formulated product, as well as the indirect costs related to obtaining licenses from environmental agencies to install and operate the treatment station.

As such, the present study aimed to identify factors that affect the use of organic solid waste from the pulp and paper industry as an organic fertilizer and soil conditioner, based on published scientific articles. The most relevant technical, legal and economic aspects of this alternative disposal are highlighted, in addition to its strengths and challenges.

Material and Methods

This is a systematic literature review (SLR), conducted in accordance with Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines. According to Page et al. (2021), a systematic review must be comprehensive, transparent, and provide details on its methodology and findings to ensure it can be replicated by other researchers.

The PRISMA method for systematic literature reviews is rigorous, ensures source reliability, and provides guidance for reporting systematic reviews. It was designed to help authors transparently report the justification for their systematic review, the methodology adopted and the results obtained (PRISMA, 2024). Based on this research and a wide range of published studies, the aim was to assess the feasibility of applying raw or processed industrial residues from the pulp and paper industry as fertilizers and soil correctives.

The research question formulated as the first step of the review was "What are the challenges and opportunities of using industrial residues from pulp and paper production as organic fertilizers and soil conditioners?", followed by the selection of reliable databases, namely Scopus and Web of Science.

The following expression was used to search the databases: ALL = (pulp* OR eucalyptus OR kraft) AND (solid residue* OR residue* OR waste* OR dregs OR grits OR sludge OR ash OR lime mud) AND (fertilizer* OR soil* OR agricultur*). This enabled all materials of interest to be considered, including sludge from wastewater treatment plants, green liquor recovery residues (such as dregs, grits, lime mud and sludge, and different types of ash), and biomass boiler ash. Eligibility criteria were studies published in English or Portuguese between 2003 and 2023, from any region, since the feasibility of applying industrial residue-based products to soil does not depend on location. The search was then refined to limit it to fields related to the topic, excluding those such as medicine and microbiology, which are not associated with managing industrial residues from the pulp and paper sector.

In the Web of Science, articles were selected based on the following micro-topic filters: Cellulose; Heavy Metals; Anaerobic Digestion; Composting; Fly Ash; Forest Management; Soil Erosion; Wood; Life Cycle Assessment; and Paper Properties. In Scopus, the selected areas were: Environmental Science; Agricultural and Biological Sciences; and Economics, Econometrics, and Finance. In Scopus, another filter was applied to select articles based on Wastewater Treatment; Industrial Waste; Wastewater; Sludge; Biomass; Heavy Metals; Soil; Heavy Metal; Composting; Waste Management; Fly Ash; Cellulose; Waste Treatment; Waste Water Treatment; Water Treatment; Solid Waste; Soils; Lime; Waste Disposal; Waste; Ash; Vermicompost; Sustainable Development; and Paper and Pulp Mills. The search was conducted in July 2023.

After the database searches, a text file (.txt) containing the metadata of the articles (title, abstract, authors, year of publication, journal and keywords) was exported for inclusion in VOSviewer software, which is used to construct and visualize bibliometric networks (VOSVIEWER, 2023). At this stage, the main keywords in the pre-selected articles were checked

to establish whether there were any other relevant terms to include in the search.

Finally, the text files were uploaded to the Rayyan platform to screen the articles and select those potentially capable of answering the research question. This platform helps remove duplicate articles, and organize and visualize titles, authors, and abstracts to improve selection (RAYYAN, 2022). Through Rayyan, the titles and abstracts of each article were read in order to determine their relevance to the research question. Articles containing information on the soil application of at least one type of organic waste from the pulp sector, even if combined with solid waste from other industries, were pre-selected. Those focusing on materials or solid waste from other sectors or that addressed the waste of interest without focusing on soil application, were excluded.

Finally, the articles selected during the screening stage were read in full to assess and consolidate the available information to answer the research question.

Results and Discussion

The initial search using the established keywords, language, time periods, and filters produced 685 articles from the Web of Science platform and 533 from Scopus, totaling 1,218 articles. Figure 1 presents the four main clusters.

Four clusters were obtained, centering on soils; industrial waste management and metals present in this waste; water and wastewater treatment and physicochemical properties; biomass, cellulose and thermal and microbiological processes.

The results obtained from the preliminary VOSviewer keyword analysis and screening identified a wide range of published studies that analyze the environmental effects of different destinations for cellulose production waste. However, around half of the articles did not investigate agricultural application, such as biogas and biomethane production, and focused on solid waste from agricultural/forestry activities (such as bark and other by-products) and/or other industrial sectors. In the screening stage, 18 duplicate articles were removed and 1099 of the remaining 1,200 were excluded after reading the abstracts, leaving 101 to be read in full with the potential to answer the research question.

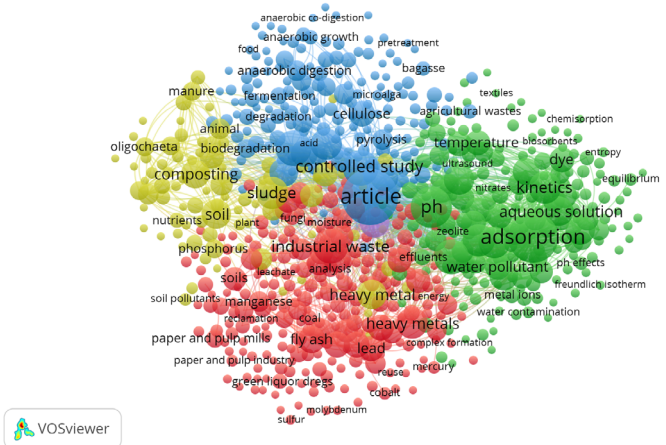

Figure 1. Clusters of keywords identified in the metadata of articles obtained from the Web of Science and Scopus platforms using VOSviewer software

During reading in full, 60 articles were excluded for focusing heavily on composting alternatives for waste using different bacteria without assessing its subsequent application to the soil; investigating combinations of cellulose waste with materials from other sectors, making it impossible to separate the specific effects of the targeted solid residue; incorporating other chemicals into the waste, which is not applicable to the reality studied; or failing to specify which solid waste was being investigated (using only a generic description of "solid waste from cellulose and paper production"). Thus, 41 articles were selected for the present study (Figure 2).

Temporal assessment of the 41 articles indicated that they were homogeneously distributed over the last two decades, as shown in Figure 3.

In 2023, only one relevant article had been published by the onset of the study, which was not included in the temporal publication evolution graph because the year had not yet concluded. However, it was included and considered in the review.

The 41 articles selected were published in 29 journals, indicating that no single periodical concentrated a large portion of the articles and the topic is well distributed across different sources. Journal quality was assessed by consulting the Sucupira Platform, a journal classification system developed by the Coordination for the Improvement of Higher Education Personnel (CAPES) from 2017 to 2020 that applies the so-called Qualis CAPES rating, with A1 denoting the highest quality.

Table 1 shows the main journals resulting from the search, their Qualis rating, and the number of articles from this review published in each.

The selected articles read in full predominantly addressed sludge from pulp and paper mill wastewater treatment stations and fly (from electrostatic precipitator) and heavy ashes from biomass boilers, due to their characteristics and high generation rates.

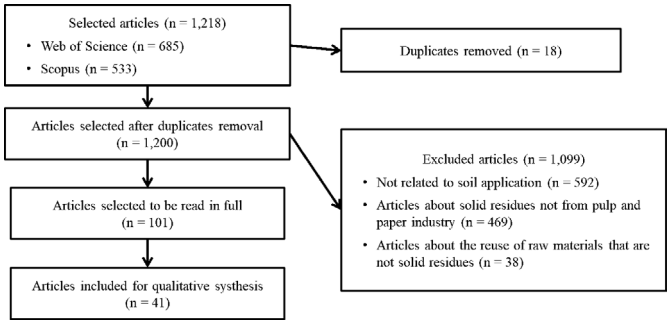

Figure 2. PRISMA Protocol Flowchart for this systematic literature review

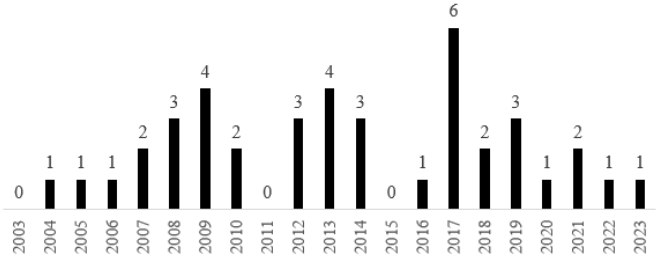

Figure 3. Distribution of the publication years of articles selected during full-text reading

Table 1. Main journals resulting from the search, their Qualis rating, and number of articles from this review published in each

Journal	Qualis	Published articles
Canadian Journal of Soil Science	A4	4
Fuel Processing Technology	A1	3
Chemosphere	A1	2
Environmental Science and Pollution Research	A2	2
Forest Science	A3	2
Journal of Cleaner Production	A1	2
Journal of Environmental Quality	A2	2
Science of the Total Environment	A1	2

Source: Sucupira Platform - CAPES (2023)

According to Dinel et al. (2004), Fraser et al. (2009), Gagnon et al. (2014), Abdi et al. (2017), and Lee et al. (2017), applying appropriate doses of sludge from pulp and paper mill wastewater treatment stations to the soil (Corbel et al., 2016), after composting and proper treatment, contributed to increasing carbon, nitrogen, and phosphorus levels, plant growth in different crops, and pH (Poykio et al., 2007). Gagnon & Ziadi (2012) reported that, in some cases, sludge alone did not provide the expected increase in macronutrients such as potassium and magnesium, which require prior enrichment before field application. Additionally, according to Alvarenga et al. (2019), Cruz et al. (2023), Morris et al. (2012), Ribeiro et al. (2018), and Undurraga et al. (2017), when combined with biomass ash and other alkaline residues, sludge also produces good results as a liming agent. However, although no relevant toxicity to flora, soil, or the environment has been reported, Gagnon et al. (2013), Hazarika et al. (2017), and Poykio et al. (2014) emphasize the need to monitor soil levels of toxic metals such as cadmium, mercury, lead, and molybdenum, which increased in some studies (albeit below the limits stipulated by local regulations). Moreover, it is important to note that these findings apply to composted sludge, which, when used as an organic fertilizer without prior treatment, can cause environmental damage, including introducing microorganisms such as E. coli, which have the potential to disrupt ecosystem dynamics (Oksanen et al., 2021).

On the other hand, biomass boiler ash increased pH due to the presence of carbonates, oxides, hydroxides, and calcium, magnesium, sodium, and potassium silicates, and improved soil fertility through higher phosphorus, potassium, and boron levels (Sharifi et al., 2013; Domes et al., 2018). In some cases, as reported by Gomes-Rey et al. (2013) and Hart et al. (2019), greater carbon, nitrogen and calcium availability (Nurmesniemi et al., 2008) and higher crop yields were observed. The use of biomass boiler ash in a Pinus plantation increased soil calcium and magnesium availability and pH, even in deeper layers and over longer time periods (Quadros et al., 2021), while another study showed increased trunk size with application of this residue (Poblete et al., 2022).

In the studies analyzed, application of fly (from electrostatic precipitators) and bottom ashes (from boiler bed purging), soil toxic metal levels remained below the limits established by environmental legislation in Finland (Dahl et al., 2009). Only Dahl et al. (2010) and Poykio et al. (2009) observed higher arsenic and lead levels, making the isolated use of ashes unfeasible and highlighting the need for further study of these metals in this residue. Finally, in one study, applying ashes in forests incurred 15 to 20% higher costs than landfill disposal. However, the authors identified many environmental gains and savings in terms of agricultural input purchases (Hope et al., 2017). In summary, the studies identified considerable agronomic potential in applying biomass ashes to the soil, either alone or combined with residues from other industries, or after enrichment (Poykio et al., 2005; Cabral et al., 2008; Nurmesniemi et al., 2008; Gagnon & Ziadi, 2012; Nurmesniemi et al., 2012; Gagnon et al., 2014; Undurraga et al., 2017; Alvarenga et al., 2019).

The dregs and grits generated in green liquor recovery are also widely addressed in the studies evaluated. In general, Nurmesniemi et al. (2010), Sebogodi et al. (2020) and Żolnowski et al. (2019) found that these residues showed considerable potential as liming agents because of their alkaline properties (mainly due to the hydrolysis of calcium carbonates into oxides, increasing pH), with calcium and magnesium levels similar to those of commercial limestone (but providing greater soil improvement). Additionally, these residues increased levels of potassium, phosphorus, and micronutrients such as copper, zinc, and boron (Zambrano et al., 2007).

Furthermore, the authors reported that dregs and grits applied in the correct formulations and concentrations did not increase soil sodium content, despite being present in these residues due to the industrial process (Cabral et al., 2008). As observed for wastewater sludge, Nurmesniemi et al. (2010) found that toxic metal levels after dregs and grits application were also below the legally stipulated limit. However, it is important to carefully monitor and control their soil concentrations, at least at the onset of application. Furthermore, Jia et al. (2017) reported concerns regarding increased soil pH resulting from the combined application of dregs and biomass fly ash, also leading to the precipitation of metals such as zinc and nickel, which merit special attention in future research. However, the same authors found that application was satisfactory from a soil stability perspective.

For lime mud and sludge, also obtained in chemical recovery, the studies analyzed here identified significant potential for liming and higher soil pH levels. Additionally, in the United States, He et al. (2009) observed an up to 10% increase in yield with controlled lime mud application in the correct formulations when compared to conventional inputs. According to Poykio et al. (2006), lime sludge exhibited considerably higher calcium content than that typically found in soil, indicating potential for increasing levels of this metal when compared to conventional conditioners. It also contained magnesium, an essential metal to ensure eucalyptus productivity (Poykio et al., 2006). In this case, although toxic metal levels were below the legally recommended limits, they should be monitored and the best formulations with other residues or inputs studied to ensure the correct balance in soil and plants (Poykio et al., 2006; Zambrano et al., 2007; Poykio & Nurmesniemi, 2008; He et al., 2009; Gagnon & Ziadi, 2012; Gagnon et al., 2013; Gagnon et al., 2014).

Finally, as a relatively new technology in the pulp industry, gasification ashes were also identified in this review, which

is interesting because the use of gasifiers has increased as an option to remove fossil fuels from lime kilns. Kilpimaa et al. (2013) reported that gasification ashes showed potential for use as a fertilizer and soil conditioner. The authors underscore that the toxic metal content of this residue is below the legislated limit, indicating its environmental feasibility.

Overall, the 41 articles selected in this systematic literature review addressed, to some extent, the technical feasibility of using certain solid residues from the pulp and paper industry as fertilizers and soil conditioners. The results demonstrate that, under certain conditions, this destination is a promising alternative.

In terms of environmental feasibility, although toxic metal concentrations have remained below the stipulated limits and comply with environmental regulations, the increased levels in the soil are concerning. In this respect, many authors emphasize the importance of periodic monitoring to ensure that there is no significant negative environmental impact.

Only one study addressed economic feasibility (Hope et al., 2017), but was limited to comparing the operational costs of using ashes as agricultural inputs to landfill disposal costs. As such, despite the large number of scientific studies demonstrating the technical and environmental potential of using residues for soil application, there is still a gap in terms of investigating its economic benefits. Thus, no conclusive articles with a comprehensive economic analysis were found, demonstrating the importance of further research to confirm the economic feasibility of this alternative.

Conclusions

1. The disposal of industrial waste in landfills is a significant challenge for the pulp and paper sector, and research has shown that its use as soil conditioners and organic fertilizers in different countries and agricultural contexts may be an environmentally appropriate alternative.

2. The articles selected in the systematic literature review examined the use of sludge from wastewater treatment plants, green liquor recovery residues, and biomass boiler ash as fertilizers and soil conditioners, and the impacts of these materials when applied to the soil.

3. After adequate processing and treatment, along with appropriate formulations, organic and inorganic residues show considerable potential in controlling soil acidity, improving fertility due to micronutrient availability, and promoting higher crop yields, thereby confirming the technical feasibility of their use as inputs in the soil.

4. In terms of environmental viability, toxic metal levels in the soil and products comply with the regulations in the countries where the research was conducted.

5. Given that research addressing the economic dimension of using residues as organic fertilizers and soil conditioners is scarce, economic feasibility studies should be conducted to comprehensively evaluate this waste management alternative for the pulp and paper industry.

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