

INFLUENCE OF PARTICLE SIZE DISTRIBUTION OF HAZARDOUS SOLID WASTE INCINERATION ASHES (HSWIA) ON LANDFILL LEACHATE FILTRATION

M. C. C. MOURA¹, G. O. SANTOS, M. L. SILVA, R. A. C. SILVA, N. D. LEITE, J. B. S. BASTOS
Instituto Federal de Educação, Ciência e Tecnologia do Ceará¹
ORCID ID: <https://orcid.org/0009-0008-6679-0996>¹
mcristina_cm@yahoo.com.br¹

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ABSTRACT

The study examines the reuse of hazardous waste incineration ashes in landfill leachate treatment. Its objective is to assess how the particle size distribution of the ashes affects the reduction of physicochemical parameters in leachate at the Caucaia Sanitary Landfill in Ceará, Brazil, thus valorizing them and minimizing their disposal in landfills. Two filtration columns with ashes of different particle sizes (F1 sieved and F2 raw) were set up. Leachate collected from the landfill was analyzed before and after filtration, demonstrating

effectiveness in removing parameters such as phosphorus, nitrite, turbidity, nitrate, etc. The characterization of the ashes revealed differences between the samples, including varying levels of oxides and particle sizes. Results suggest potential use in pre-treatment to reduce pollutant loads before the primary treatment of leachate. The study underscores the potential of incineration ashes to enhance leachate treatment and reduce the environmental impact of improper disposal.

KEYWORDS: Ash from Hazardous Solid Waste Incineration, Particle Size Distribution, Landfill Leachate.

INFLUÊNCIA DA GRANULOMETRIA DAS CINZAS DE INCINERAÇÃO DE RESÍDUOS SÓLIDOS PERIGOSOS (RSP) NA FILTRAÇÃO DE LIXIVIADO DE ATERRO SANITÁRIO

RESUMO

O estudo analisa a reutilização das cinzas de incineração de resíduos perigosos no tratamento de lixiviado de aterro sanitário. O objetivo é avaliar como a granulometria das cinzas afeta a redução dos parâmetros físico-químicos dos lixiviados no Aterro Sanitário de Caucaia, Ceará, valorizando-as e minimizando envio a aterros. Duas colunas de filtração com cinzas em diferentes granulometrias (F1 peneirada e F2 bruta) foram criadas. Lixiviado coletado no aterro foi analisado antes e após a filtração,

demonstrando eficácia na remoção de parâmetros como fósforo, nitrito, turbidez, nitrato etc. A caracterização das cinzas evidenciou diferenças entre as amostras, com variados teores de óxidos e granulometria. Resultados sugerem uso em pré-tratamento para reduzir a carga poluente antes do tratamento principal dos lixiviados. O estudo destaca o potencial das cinzas de incineração para melhorar o tratamento de lixiviado e reduzir o impacto ambiental do descarte inadequado.

Palavras-chave: Cinzas de incineração de resíduos perigosos, granulometria, lixiviados de aterro sanitário.

1 INTRODUCTION

The disposal of Hazardous Solid Waste (HSW) has become an increasing challenge, especially in large cities. In this regard, incineration has proven to be an alternative that significantly reduces both the weight (up to 70%) and volume (up to 90%) of waste through controlled combustion (PINTO, 2018).

In Brazil, a portion of HSW is sent to treatment facilities for incineration, autoclaving, microwave treatment, and industrial landfills (representing the more expensive alternatives). Another portion is blended and/or co-processed in cement kilns (utilizing them as inputs), while some ends up in Class IIA landfills or open dumps (inappropriate for HSW), posing various environmental and social risks.

In Fortaleza, Brazil, HSW generators (regardless of volume) are legally required to assume all costs for waste management services (segregation, packaging, storage, transportation, treatment, disposal) under Municipal Law 10,430/2015 (FORTALEZA, 2015). Licensed generators typically send their HSW for incineration to the Hazardous Waste Treatment Center (CTRP) in the Jangurussu neighborhood, through contracts with specialized transporters.

The incinerator in Fortaleza, inaugurated in 2001, employs German technology, operates with fluidized bed combustion, and has two chambers (operating at 900 and 1000°C) according to Viana (2013). Current information indicates it processes approximately 9 tons of HSW daily, ensuring sterilization and volume/weight reduction. Approximately 98% of the waste consists of Healthcare Waste (HCW), with the remaining 2% being industrial and other waste (both classified as HSW).

One of the byproducts of incineration is ash (fly ash and bottom ash), which requires special handling. In Fortaleza, this ash is sent to the Caucaia Sanitary Landfill in the Metropolitan Region.

In various parts of the world, efforts are growing to increase the valorization of ashes generated from incinerators and reduce their disposal in sanitary landfills. Some studies have explored their applicability in pavement layers (PATRA et al., 2017), cement production (CLAVIER et al., 2019), as aggregates in concrete (LI, et al., 2018), incorporation into normal and adhesive mortars (AGUIAR et al., 2002), red ceramics (COUTINHO; VIEIRA, 2016), asphalt mixtures, and the synthesis of alkali-activated lightweight materials (ZHU et al., 2018a). In Singapore, for example, there is significant interest in reusing these ashes due to land and resource scarcity (LIU et al., 2018). However, little is known about the behavior of these ashes as filter media for solutions or contaminated liquid samples, such as landfill leachate, which underscores the need for further research.

According to Silva et al. (2013), filtration is an operation that consists of solid-liquid separation, that is, solid particles are mechanically separated from a liquid suspension using a filter medium, and the clarified liquid obtained in this process is called filtrate. Thus, this process can be used to treat different types of effluents.

Generally, landfill leachate is treated using stabilization ponds or solar distillation (SÁ et al., 2012), discontinuous sequential biological reactors (CHÁVEZ-PORRAS et al., 2018), sand filters (ROEHRS, 2007), or electroflotation processes. Another bias is the treatment of these effluents through the bioremediation process using microorganisms (PESENTI et al., 2023), even so, the treatment of leachate in filtration columns composed of incinerated HSW ashes remains unexplored, motivating this research (which is unprecedented in Brazil). The aim of this study was to assess the influence of the particle size distribution of incinerated HSW ashes on the reduction of physical and chemical parameters in the leachate from the new Caucaia Sanitary Landfill.

2 METHODOLOGY

The experiment was conducted at the Soil Mechanics Laboratory, Department of Hydraulic and Environmental Engineering (DEHA), at the Federal University of Ceará (UFC), Pici campus. Two filtration columns were assembled using PVC pipes with a diameter of 10 cm and a height of 50 cm. At the bottom of each pipe, a properly perforated PVC cap was installed to allow the passage of leachate. In the tube corresponding to Filter 1 (F1), ashes from the first Characterization Campaign (CC1) were used, which had been previously dried and characterized, with a particle size distribution ranging from 2.0 mm to 1.2 mm. In Filter 2 (F2), raw ashes from the second Characterization Campaign (CC2) were used (not sieved). Three layers of 4 cm of ashes were placed inside each tube, and compaction was applied to each layer using the Proctor Normal energy, with 26 blows. Additionally, washed gravel with particle sizes ranging from 9.5 to 12.5 mm was placed as a drainage layer (below and above the compacted ash media), and a nylon screen (10 cm in diameter with an opening of 1.11 mm) was also used, as shown in Figure 1. Thus, the experiment consisted of two columns with 100% hazardous waste incineration ashes but with different particle size distributions.

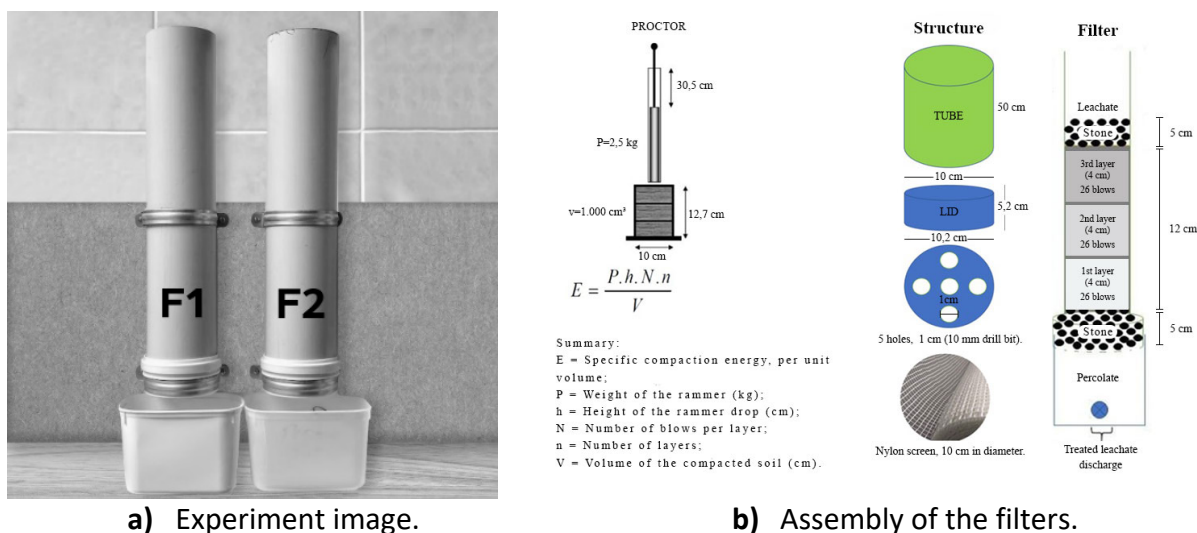


Figure 1: Filters of the pilot experiment.

The ash samples were collected from the waste incinerator at CTRP in Fortaleza, Ceará, and, in the absence of specific standards for characterizing anthropogenic materials (in this case, waste),

methods from Technical Standards applied to soils were used, as also observed by Dijkstra et al. (2019), as shown in Table 1.

Table 1: Summary of HSW ash analyses.

Analysis	Reference or Method	Location
Particle Size Distribution (05 samples)	NBR 7181/2018	Laboratory of Soil Mechanics, Department of Hydraulic and Environmental Engineering, UFC
Normal Proctor Compaction (05 samples)	NBR 7182/2020	
Natural Moisture Content of the Sample (05 samples)	Oven at 60-65°C for 24 hours	
Grain-Specific Gravity Determination (05 samples)	NBR 6458/2017	
Permeability with Variable Load (02 samples)	NBR 14545/2021	
Permeability with Constant Load (02 samples)	NBR 13292/2021	
Elemental Chemical Composition (05 samples)	Espectrometria de Fluorescência de Raios-X	
Shape Properties in AIMS (02 samples)	Norma DNIT 432/2020 – ME	Geology Laboratory, Department of Geology, UFC

The raw leachate was collected from the second stabilization pond at the Sanitary Landfill and, like the treated leachate, was sent for characterization to the laboratory of the Núcleo de Tecnologia e Qualidade Industrial do Ceará (NUTEC) for chloride, apparent color, Chemical Oxygen Demand - COD, phosphorus, nitrite, nitrate, pH, dissolved solids, and turbidity analysis, following the Standard Methods for the Examination of Water and Wastewater.

Each filter received 2 liters of raw leachate (analyzed in the laboratory on the same day as collection at the Sanitary Landfill). The treated leachate was analyzed after percolating through the filtration columns, which means 24 hours later. Two campaigns were carried out: one in April and another in July 2022.

3 RESULTS AND DISCUSSIONS

The samples used in F1 exhibited compaction with a Maximum Dry Bulk Density (MEAS_{max}) of 1.610 g/cm³ and 12.42% for the mean Optimum Moisture Content (Hótima), natural moisture content of 2.14%, grain specific gravity of 2.714, and an average elemental chemical composition of Calcium Oxide (57.41%), Iron Oxide (16.43%), and Titanium Dioxide (7.88%), among others. The samples used in F2 exhibited compaction with MEAS_{max} of 1.506 g/cm³ and 22.76% for the mean Hótima, natural moisture content of 22.16%, grain specific gravity of 2.367, and an average

elemental chemical composition of Calcium Oxide (58.20%), Aluminum Oxide (8.28%), and Silicon Dioxide (7.84%), among others.

Permeability and shape property analyses were only performed on CC2 samples (due to equipment issues), achieving a permeability of $K_{20} = 3.2 \times 10^{-5}$ cm/s and shape properties, according to the classification proposed by Ibiapina et al. (2018), for coarse ashes: sphericity = low sphericity, angularity = subrounded, texture = soft; and for fine ashes: angularity = subrounded and 2D shape = semicircular.

The particle size distribution of the two analyzed samples exhibited dimensions similar to sand (63.0%), suggesting the formation of macropores and channels that enable good infiltration capacity. For CC1, 22% gravel, 22% coarse sand, 27% medium sand, 12% fine sand, and 16% silt/clay were observed; and for CC2, 13% gravel, 26% coarse sand, 24% medium sand, 15% fine sand, and 23% silt/clay were observed.

F1 (treated ashes) had the capacity to reduce the concentration of 6 leachate parameters in the first campaign (66.7% of the total analyzed parameters) and 5 parameters (55.6%) in the second campaign (qualitative observation). In both campaigns, a reduction in concentration was coincident for 4 parameters (apparent color, phosphorus, COD, and turbidity). The highest reduction rates occurred for phosphorus and nitrite in the first campaign (100.0%), as indicated in Table 2.

Table 2: Results of leachate filtration in filter 1.

Parameters	Gross 1 st C	Gross 2 nd C	Filtered 1 st C	%	Filtered 2 nd C	%
pH at 25°C	8,10	8,12	9,32	15,1	9,01	11,0
Chlorides (mg/L)	3.180,00	2.968,94	4.740,00	49,1	3.403,20	14,6
Apparent color (uH)	3.550,00	3.150,00	152,00	-95,7	1.170,00	-62,9
Phosphorus (mg/L)	4,57	3,85	**< LQ	-100,0	0,14	-96,4
COD (mg/L)	3.484,50	1.904,00	2.382,00	-31,6	463,40	-75,7
Dissolved solids (mg/L)	9.382,00	8.816,00	9.404,00	0,2	7.906,00	-10,3
Nitrate (mg/L)	6,19	*< LQ	1,03	-83,4	*< LQ	
Nitrite (mg/L)	0,08	*< LQ	*< LQ	-100,0	*< LQ	
Turbidity (mg/L)	370,00	195,00	14,32	-96,1	69,40	-64,4
			1 ^a C	%	2 ^a C	%
Summary	Elevation		3	33,3	2	22,2
	Reduction		6	66,7	5	55,6
	Maintenance		-	-	2	22,2

*< LQ= less than the quantifiable limit

F2 (raw ashes) had the capacity to reduce the concentration of 6 parameters in each of the campaigns (66.7% of the total analyzed parameters). In both campaigns, there was a reduction in concentration, coincident, for at least 5 parameters (apparent color, COD, nitrate, nitrite, and



turbidity). The highest reduction rate occurred with nitrite (88.0% in the first campaign and 89.4% in the second campaign), as shown in Table 3.

Table 3: Results of leachate filtration in filter 2.

Parameters	Gross	1 st C	%	2 nd C	%
pH at 25°C	8,31	8,81	6,0	8,93	7,5
Chlorides (mg/L)	4.475,56	5.202,29	16,2	5.033,90	12,5
Apparent color (uH)	2.370,00	1.900,00	-19,8	1.770,00	-25,3
Phosphorus (mg/L)	0,74	0,33	-55,4	1,10	48,6
COD (mg/L)	594,8	587	-1,3	310,00	-47,9
Dissolved solids (mg/L)	11.834,00	17.738,00	49,9	11.398,00	-3,7
Nitrate (mg/L)	0,33	0,26	-21,2	0,30	-9,1
Nitrite (mg/L)	2,84	0,34	-88,0	0,30	-89,4
Turbidity (mg/L)	264	173,7	-34,2	143,4	-45,7
		1st Campaign	%	2nd Campaign	%
Summary	Elevation	3	33,3	3	33,3
	Reduction	6	66,7	6	66,7
	Maintenance	-	-	-	-

*< LQ= less than the quantifiable limit

It is observed that the highest reduction rates, by parameter, occurred in F1 in both campaigns, highlighting the granulometric influence of the ashes used to achieve the best results, which, even after the time interval between the campaigns, continued to show high reduction percentages.

A more detailed analysis by parameter revealed that, despite the raw leachates being nearly constant (pH between 8.1 and 8.3), there was an increase of up to 15.1% in the pH of the leachate after filtration, making it more alkaline. The leachates obtained in both F1 campaigns were outside the relevant CONAMA and COEMA limits (5 to 9), but the results presented by F2 were within the permissible range, as shown in Figure 3.

The behavior of pH in leachate treatment processes does not follow a single trend, as follows: (i) Carard (2018), after applying photocatalytic ozonation treatment with titanium dioxide, achieved a reduction in pH in all conducted tests; (ii) Godoi (2019), using paper filters as the filter media, observed an increase in pH after filtration for most samples; (iii) Silva et al. (2021) used sands with various particle sizes, clays, and activated carbon as filter media, and the pH of the leachate did not vary considerably after filtration compared to raw leachate.

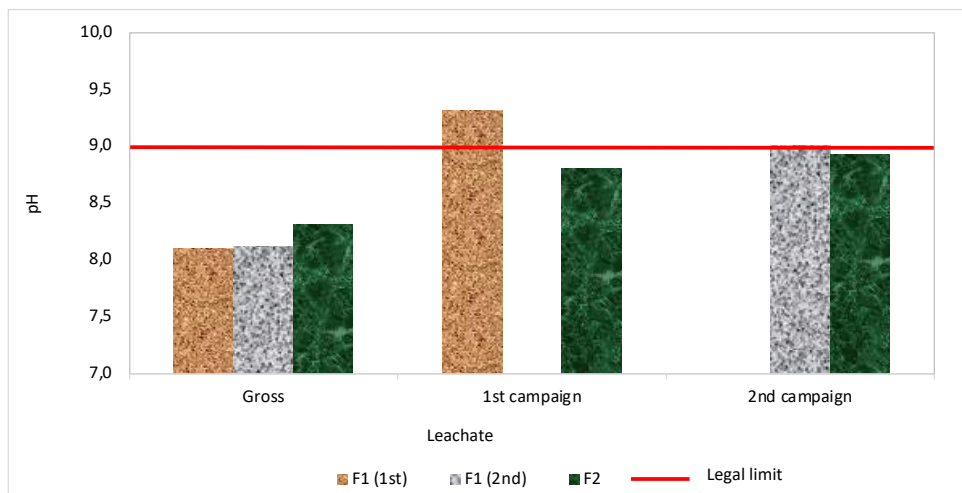


Figure 2: pH parameter behavior.

In sanitary landfills, chloride is not subject to significant chemical or biological transformations, allowing it to pass through older layers without significant attenuation (SOUTO, 2009). Therefore, the removal of this parameter, among others, is considered extremely important for effective leachate treatment (LEBRON et al., 2021). There was an increase in chloride in the treated leachate, possibly due to residual chlorine from the disinfection process, an operational step performed in the studied incinerator. This increase was more pronounced in F1 in the first campaign (49.1%). Rodrigues (2007), applying electrolytic treatment to landfill leachate, observed a 16% removal of chloride compared to raw leachate; however, when adding salt to the effluent before treatment, chloride values increased by more than 200%.

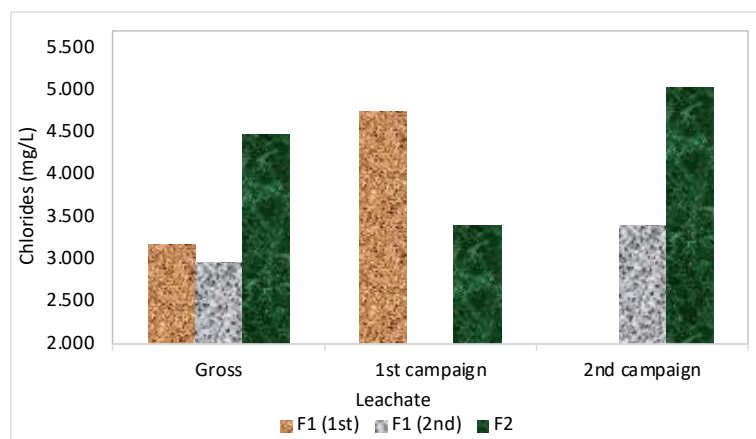


Figure 3: Chloride parameter behavior.

Apparent color is a biological process that only occurs when there is some means of transmitting sunlight through the effluent, making it relevant in the study of treatment efficiency (GODOI, 2019). Both filters removed apparent color, with F1 (first campaign) achieving a particularly high reduction of 95.7%. The removal of this physical (organoleptic) parameter is of great importance and is desired as an outcome. Regarding the legal requirement for this parameter, COEMA 02/2017 determines that non-sanitary effluents can only be discharged directly into water bodies, subject to other applicable requirements, when they do not contain colorants

and pigments. Therefore, despite the reduction achieved, this legal requirement was not met by the method used in this study.

Carard (2018), applying photocatalytic ozonation treatment with titanium dioxide, achieved the removal of apparent color in leachate of up to 61%. Fujii et al. (2019) achieved removal of up to 99.7% through coagulation followed by upflow filtration using gravel as post-treatment.

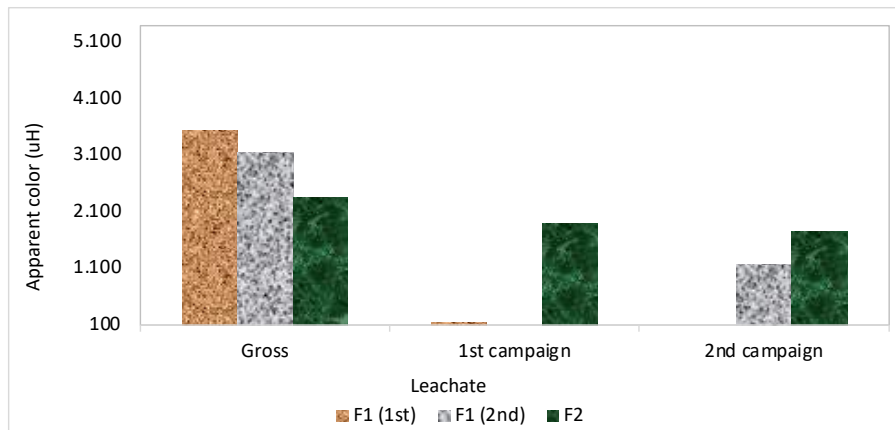


Figure 4: Apparent Color parameter behavior.

The quantity of the chemical element phosphorus released during the degradation of organic matter in effluents primarily has anthropogenic origins. It is an essential element for living organisms, as it forms cell membranes, nucleic acids, bones and teeth. However, it is a limiting nutrient in landfill environments, and when present in effluents discharged into receiving bodies of water, it can lead to eutrophication (LÔBO, 2006).

The removals for this parameter, especially in F1 (100% and 96.36%), represented one of the most promising results of the research, as phosphorus is a growth-limiting factor that is difficult to achieve for most treatment systems.

According to CONAMA Resolution No. 430/2011, for the parameter phosphorus, it is at the discretion of the competent environmental agency to define specific standards for the discharge of effluents into receiving bodies of water with a historical record of cyanobacterial blooms, in areas where water is used for public supply.

Considerable reductions in phosphorus concentrations were achieved in treatments proposed by Carard (2018): 85.0% and Rodrigues (2007): from 50.6% to 90.1%. Among the leachate treatment experiences studied by Lebron et al. (2021), chemical precipitation was described as having the best results in reducing phosphorus.

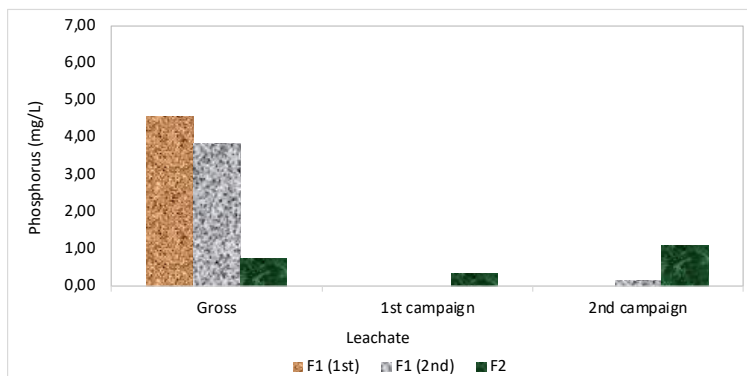


Figure 5: Phosphorus parameter behavior.

Chemical Oxygen Demand (COD) is an indicator of the degradation of waste over time; the lower the COD values found, the older the waste is. It measures the amount of organic matter that can be chemically oxidized (COSTA, 2021). Vicente (2021) emphasizes that COD and ammonia concentrations are considered the main indicators of acute toxicity in an effluent.

F1, in the second campaign, had the highest reduction (75.66%) for the parameter; however, it should be noted that in both filters the most significant reductions occurred in the second campaign. Probably, the results with the ashes were not better due to their own chemical composition.

Lebron et al. (2021) observed COD reduction of around 40% in stabilization pond treatments, 59% in activated sludge systems, 76% in biological filters, 60% to 81% in moving bed biofilm reactors, and 89.6% in membrane bioreactors. Fujii et al. (2019) claimed to have achieved COD removals of 82.4% through coagulation followed by filtration.

The regulations (CONAMA and COEMA) establish a limit of 200 mg/L for this parameter, which was not met by either of the filters.

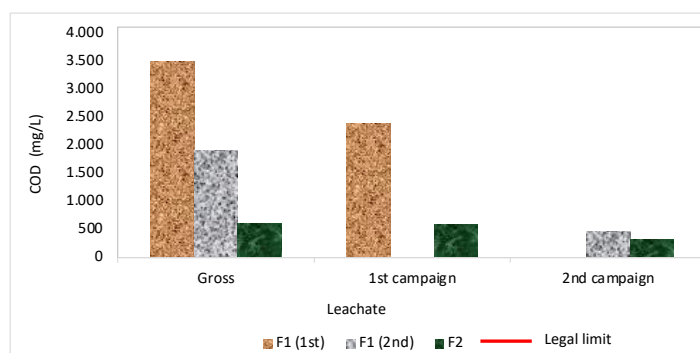


Figure 6: COD parameter behavior.

Dissolved solids are certain elements included in the water, such as bicarbonate and phosphorus, which do not settle. Although essential for life maintenance, when the concentration of these solids is high, it can negatively affect water density and directly influence the development of aquatic environments. In leachate, the action of dissolved solids directly influences the variability of ions present, thereby affecting conductivity (SILVA, 2017).

The best performance in the removal of this parameter was observed for F1, in the second campaign (10.32%). It should be noted that the possible increase in this parameter in the first campaigns in both filters may have been caused by the entrainment of elements present in the filter medium itself. Silva (2021) achieved a 78.2% removal of this parameter with ponds (anaerobic, facultative, and dry) at a landfill. Current legislation does not establish a specific discharge limit for this parameter.

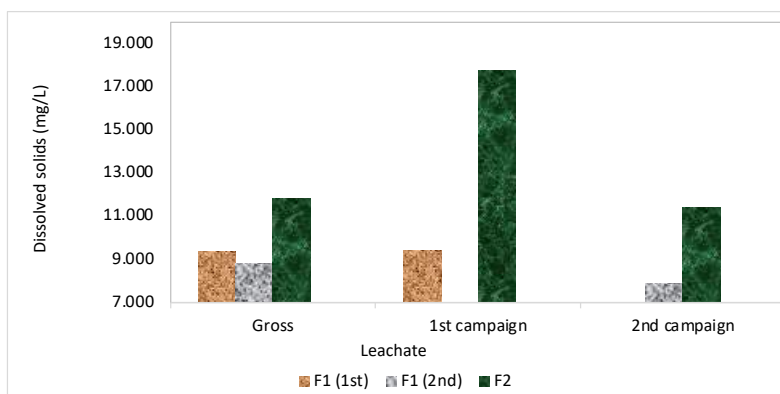


Figure 7: Behavior of the Dissolved Solids parameter.

Nitrate is formed from the oxidation of nitrite, possesses high solubility, and is carried into water bodies through leaching, resulting in their contamination (MOREIRA, 2019). The highest performance in removing this parameter was observed in F1, the first campaign (83.36%).

Silva (2021) achieved a removal rate of 78.2% using ponds (anaerobic, facultative, and dry) in a landfill. Fujii et al. (2019) achieved reduction of 98% by applying the ammonia stripping treatment followed by activated biological treatment. In the study on the efficiency of advanced processes in reducing the toxicity of landfill leachate using nanofiltration membranes, Reis (2014) obtained a 47% reduction in nitrate. Compliance with discharge standards allows for 10 mg/L (COEMA n° 02/2017), a result achieved by both filters in this study, in both campaigns.

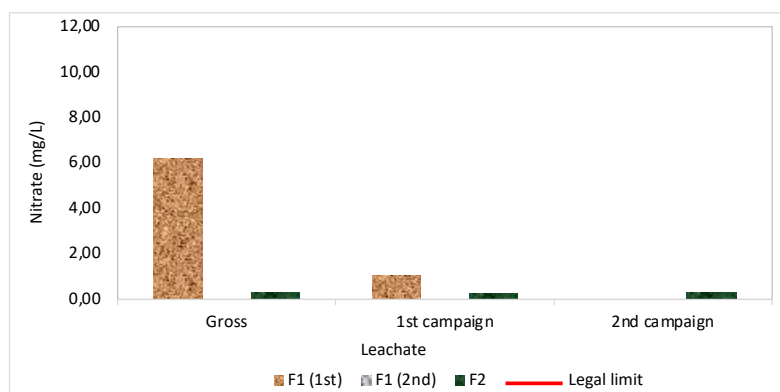


Figure 8: Nitrate parameter behavior.

Wang et al. (2018) warn that nitrite is a recognized carcinogen and should not be overlooked in treatments, as the detrimental effects on the environment can be substantial.

Consistently, the scientific literature points to nitrites as responsible for increased toxicity in leachates after their degradation (REIS, 2014).

The removal of this parameter showed high efficiency in both filters under study, with the highest achievement being observed in F1, the first campaign (100%).

The maximum allowable discharge value, as established by relevant regulations, is 1 mg/L, result achieved by both filters studied in the campaigns where verification was possible.

After ammonia stripping treatment followed by activated biological treatment, Fujii et al. (2019) achieved a 96% nitrite removal. Reis (2014) obtained an average reduction of 48% when applying nanofiltration after a yeast-inoculated membrane bioreactor.

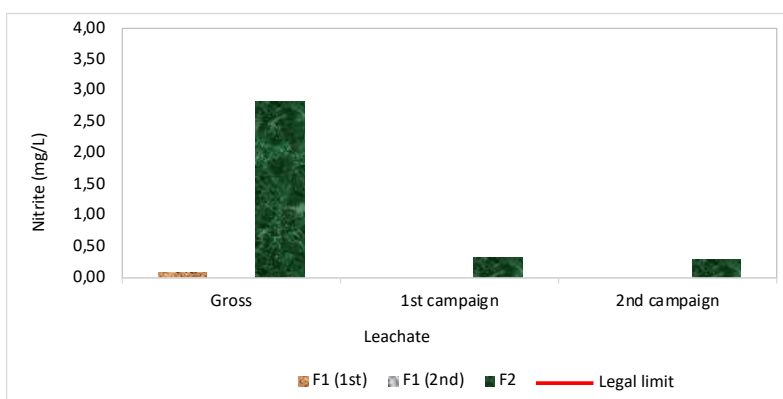


Figura 9: Nitrite parameter behavior.

Turbidity is a physical property of fluids that results in reduced transparency due to the presence of suspended materials, caused by the presence of suspended solids, both organic and inorganic in nature, and microorganisms (SILVA, 2020).

Both filters under study showed efficiency in reducing turbidity, with the highest achievement observed in F1, the first campaign (96.13%).

Regarding compliance with turbidity standards, the resolution CONAMA n° 357/2005 stipulates that discharges into freshwater of class 2 and 3 should have a maximum of 100 NTU (Nephelometric Turbidity Units). This result was achieved in both campaigns for F1.

In a study conducted by Cunha (2022), through the combination of specific doses of adsorbents and coagulants, a reduction of 99.99% in turbidity was achieved. Rocha, Lucena and Porto (2020), using the photo-Fenton process and harnessing solar light to enhance degradation reactions, achieved turbidity removal greater than 90%. Castilhos Junior, Dalsasso and Rohers (2010) noticed an increase in turbidity when studying upward direct filtration in activated carbon columns as a pretreatment for landfill leachates, which the authors attributed to the passage of flocs within the filter medium.

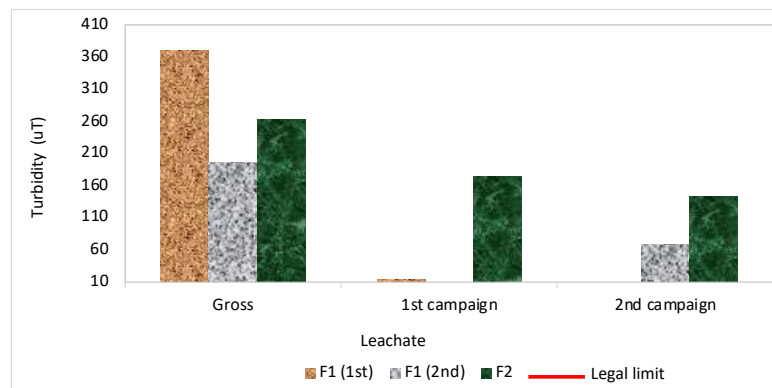


Figure 10: Turbidity parameter behavior.

4 CONCLUSIONS

This groundbreaking study in Brazil aimed to evaluate the influence of the particle size distribution of ashes from the incineration of Hazardous Solid Waste (HSW) as a filtration element in the treatment of landfill leachate. It demonstrated that Filter 1 (with particle sizes selected between 2.0 mm and 1.2 mm) was the most efficient in reducing the percentage concentration of various parameters. Specifically, in the first campaign, it achieved significant reductions: apparent color (95.7%), phosphorus (100%), COD (Chemical Oxygen Demand) (31.6%), nitrate (83.4%), nitrite (100%), and turbidity (96.1%). In the second campaign, it also showed substantial reductions: apparent color (62.9%), phosphorus (96.4%), COD (75.7%), and dissolved solids (10.3%).

Except for the behavior observed for phosphorus (F2, second campaign), both filters (F1 and F2) exhibited similar trends of reduction and elevation in the same parameters, differing only in the percentage achieved by each filter.

This research, part of a broader ongoing study, has contributed to understanding the physical and chemical characteristics of incinerator ashes and their performance as a filtration medium for leachate from the Caucaia Landfill. It opens possibilities for larger-scale experiments, reduces disposal on the ground, and initiates a line of investigation for sanitary and environmental engineering. Furthermore, comparing the results of leachate treated in the proposed system with those treated using other established technologies will help determine to what extent the ash columns align with discharge standards set by CONAMA and COEMA.

5 ACKNOWLEDGMENTS

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ABOUT THE AUTHORS

M. C. C. MOURA

Estudante do Mestrado em Tecnologia em Gestão Ambiental (PGTGA) do IFCE. Membro do Grupo de Estudos, Projetos e Pesquisas em Resíduos Sólidos do IFCE.

E-mail: mcristina_cm@yahoo.com.br

<https://orcid.org/0009-0008-6679-0996>

G. O. SANTOS

Doutor em Saneamento Ambiental pela Universidade Federal do Ceará (UFC). Professor Titular do IFCE e do PGTGA. Coordenador do Grupo de Estudos, Projetos e Pesquisas em Resíduos Sólidos do IFCE.

E-mail: gemmelle@gmail.com

<https://orcid.org/0000-0003-1852-8770>

M. L. SILVA

Mestre pelo PGTGA. Membro do Grupo de Estudos, Projetos e Pesquisas em Resíduos Sólidos do IFCE.

E-mail: contatomichaells@gmail.com

<https://orcid.org/0000-0002-9481-5854>

R. A. C. SILVA

Doutor em Geologia pela UFC. Professor Pleno I da Secretaria da Educação. Técnico do Laboratório de Mecânica dos Solos da UFC. Membro do Grupo de Estudos, Projetos e Pesquisas em Resíduos Sólidos do IFCE.

E-mail: roberto@det.ufc.br

<https://orcid.org/0000-0003-2214-4999>

N. D. LEITE

Mestre pelo PGTGA. Gerente de projetos do LABIFOR (Prefeitura Municipal de Fortaleza). Membro do Grupo de Estudos, Projetos e Pesquisas em Resíduos Sólidos do IFCE.

E-mail: nirlania.dl@gmail.com

<https://orcid.org/0000-0003-3219-4172>

J. B. S. BASTOS

Doutora em Engenharia de Transportes pela UFC. Professora do PGTGA. Membro do Grupo de Estudos, Projetos e Pesquisas em Resíduos Sólidos do IFCE. Atualmente pesquisa na área de Engenharia de Transportes, com ênfase em Infraestrutura de Transportes.

E-mail: juceline.santos@ifce.edu.br

<https://orcid.org/0000-0002-1125-2196>

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