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Exploration of Affordable Adsorbents for Removal Heavy Metals From Tannery Effluent

Anower Hossain Shawon[1]; Md. Mahmudul Hosain[1]; Muntahin Araf [1]; Md. Yasin[2]

Department of Civil Engineering
Ahsanullah University of Science and Technology
Dhaka, Bangladesh

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Abstract: Heavy metal contamination from tannery industries in the Savar Hemayetpur area of Dhaka, Bangladesh, poses serious environmental and public health risks. This study evaluated the heavy metal load of raw tannery wastewater and the performance of three low-cost adsorbents peanut husk charcoal, natural zeolite, and fly ash in removing chromium, copper, zinc, and lead from tannery wastewater collected from the BSCIC Tannery Industrial Estate, Hemayetpur, Savar, Bangladesh. Under controlled laboratory conditions, batch adsorption experiments were executed, and the concentrations of heavy metals were quantified using a DR6000 UV-Vis spectrophotometer. The analysis showed that the levels of heavy metals in the untreated wastewater exceeded the permissible thresholds set by the Bangladesh Department of Environment and the World Health Organization. After the adsorption treatment, all tested adsorbents exhibited notable reductions in metal concentrations. Among them, natural zeolite achieved the highest removal efficiency, especially for chromium. Peanut husk and fly ash also performed well in adsorption. These results indicate that low-cost adsorbents present an effective, sustainable, and economically viable solution for treating heavy metal contaminated tannery wastewater, with significant potential for large scale industrial wastewater treatment applications.

Keywords: *Affordable, Adsorption, Fly Ash, Heavy Metals, Natural Zeolite, Peanut Husk Charcoal, Tannery Wastewater.*

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I. INTRODUCTION

Heavy metal contamination of industrial effluent is among the most pressing environmental challenges facing rapidly industrialising economies, and the leather-processing sector of Bangladesh illustrates the problem acutely. The tanneries clustered in the Savar Hemayetpur area of Dhaka discharge large volumes of effluent bearing elevated concentrations of chromium (Cr), lead (Pb), copper (Cu), zinc (Zn), cadmium (Cd), and nickel (Ni), much of which reaches the Dhaleshwari River either directly or through an inadequately performing Central Effluent Treatment Plant (CETP) [1,2]. Chromium, introduced mainly through the basic chrome sulfate used in the tanning process, is consistently reported as the dominant contaminant, with concentrations in the Hemayetpur drainage network reported to exceed 100 mg/L against a Bangladesh Department of Environment (DoE) discharge limit of 0.5 mg/L [1]. Lead and cadmium enter the effluent stream from dyeing and finishing operations and from the corrosion of processing equipment [2]. Because the contaminated water is widely reused for irrigation in the surrounding agricultural belt, metals such as chromium accumulate progressively in adjoining soils, with concentrations near discharge drains reported as high as 250 mg/kg, impairing soil fertility and root development [3].

Unlike organic pollutants, heavy metals do not biodegrade; they persist indefinitely, bioaccumulate in aquatic organisms, and biomagnify along the food chain until they reach human consumers through fish, crops, and drinking water [4]. Chronic low-level exposure is associated with renal and hepatic dysfunction, neurodevelopmental impairment, and carcinogenesis, effects that are reflected in the stringent Maximum Contaminant Levels set by regulatory bodies such as the United States Environmental Protection Agency. The combination of high pollutant loading, limited treatment infrastructure, and downstream agricultural reuse makes the Savar Hemayetpur corridor a priority site for wastewater remediation research.

Conventional heavy-metal removal technologies chemical precipitation, ion exchange, membrane filtration,

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and electrochemical treatment can achieve high removal efficiencies but are frequently constrained by high capital and operating costs, complex equipment requirements, and the generation of secondary sludge, limitations that are especially restrictive for small- and medium-scale tanneries in developing countries [5]. Adsorption has therefore emerged as a comparatively simple, low-cost, and environmentally benign alternative, particularly when the adsorbent itself is an inexpensive agricultural or industrial by-product. Peanut husk charcoal (PHC), an agricultural residue rich in cellulosic and lignocellulosic surface functional groups, has demonstrated strong affinity for several heavy metal cations through ion exchange and surface complexation [6,7]. Natural zeolite, a crystalline aluminosilicate with substantial cation exchange capacity, is widely reported as an effective sorbent for divalent metal ions [8]. Fly ash, a coal-combustion by-product composed predominantly of silica and alumina, offers comparatively lower adsorption capacity but negligible cost and near-universal local availability [9].

Agricultural and industrial waste materials, owing to their abundance, low cost, and favorable surface chemistry, have attracted particular interest as low-cost adsorbents for heavy metal removal [18,19,20]. Peanut husk charcoal has shown strong adsorption capacity for heavy metal ions due to its high surface area and the carboxyl, phenolic hydroxyl, carbonyl, and lactone functional groups present on its surface, which support metal binding through ion exchange and surface complexation [9,10]; comparative studies have found the adsorption capacity of peanut hull carbon for copper to substantially exceed that of commercial granular activated carbon [10]. Tea waste, sawdust, and other agricultural residues have likewise demonstrated useful removal capacities for copper, lead, and chromium [11,12]. Fly ash, an industrial by-product of coal combustion, offers a porous, reactive silica–alumina matrix that has been shown to remove several heavy metal cations from aqueous solution, performing comparably to activated carbon at sufficiently high dosages despite its lower cost [13], while natural zeolites hydrated aluminosilicate minerals formed by the alteration of volcanic glass are particularly effective for divalent heavy metal cations owing to their three-dimensional aluminosilicate framework and exchangeable sodium, calcium, and potassium ions, which are relatively innocuous and make zeolites well suited to treating industrial effluent [14,15]. Other studies have examined high-quality limestone [16] and goethite [17] as further examples of natural and mineral-based low-cost adsorbents, underscoring the breadth of materials that have been investigated as economical alternatives to activated carbon.

For an adsorbent material to be commercially viable, it should combine high selectivity for the target metal ions with favorable transport and kinetic properties, thermal and chemical stability, adequate mechanical strength, resistance to fouling, and, ideally, the capacity for regeneration and reuse. Low-cost adsorbents derived from agricultural and industrial waste can offer most of these properties at a fraction of the cost of commercial activated carbon, while simultaneously providing a productive use for materials that would otherwise present a disposal burden.

Despite this substantial body of work, most published adsorption studies evaluate synthetic single-metal solutions rather than authentic industrial effluent, and few studies directly compare multiple low-cost adsorbents under identical experimental conditions using real tannery wastewater from the Savar Hemayetpur zone. This represents a meaningful gap, since the presence of competing ions, organic matter, and variable pH in actual effluent can materially alter adsorption behaviour relative to synthetic systems. The present study addresses this gap by (i) quantifying the initial concentrations of Cr, Cu, Zn, and Pb in raw wastewater collected from tannery units within the BSCIC Tannery Industrial Estate, Dhaka (TIED), and (ii) evaluating and comparing the adsorption performance of peanut husk charcoal, fly ash, and natural zeolite for the removal of chromium, copper, zinc, and lead from this effluent under a standardized batch methodology, with the aim of identifying practical, low-cost treatment options suited to the Bangladeshi tannery industry.

II. MATERIALS AND METHODS

- *Study Area and Sample Collection*

Samples of wastewater were obtained from the BSCIC Tannery Industrial Estate in Dhaka (TIED), also referred to as Chamra Shilpa Nagari, located in Savar Upazila (23.8583°N, 90.2667°E). Effluent samples were collected from the primary discharge channels of three tannery facilities following the CETP outlet. This was done using 500 mL plastic bottles that had been pre-cleaned and covered with black tape to prevent photodegradation. To avoid cross-contamination, gloves were worn during sampling, and each bottle was labeled with the sampling location, date, and sample number to maintain traceability throughout the subsequent analysis and treatment stages.

- *Instrumental Determination of Initial Metal*

- *Concentrations*

The gathered tannery effluent samples were used to determine the initial amounts of chromium, copper, zinc, and lead. A DR6000 UV–VIS spectrophotometer, which measures each metal at its characteristic absorbance wavelength in accordance with the manufacturer-specified reagent protocol for the relevant metal test, was used to analyze each sample as received.

- *Preparation of Adsorbents*

Peanut husks were sourced from the local market and thoroughly washed with distilled water to eliminate dust. They were then oven-dried at 100 °C for 24 hours, ground with a laboratory mill, and sieved to achieve a particle size between 0.3 and 0.6 mm. The milled husks were rinsed with 0.1 N HCl, and their pH was adjusted to a range of 6 to 7 using the same solution. The resulting peanut husk charcoal was dried and stored at 80 °C until it reached a stable weight and moisture level, ensuring uniformity across batches for future experiments. Fly ash, also procured from the local market in Dhaka, was dried at 110 °C for 3 hours to eliminate any remaining moisture and sieved to a particle size of 0.0 to 0.6 mm. Locally obtained natural zeolite was crushed, ground, sieved to a size of 0.3 to 0.6 mm, and oven-dried at 100 ± 5 °C for 24 hours.

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- *Characterization of Adsorbents*

The surface area of PHC has been found to equal to 485 m^2g^{-1}. This value is very high in comparison with other carbons, which have a surface area about of 10-100 m^2g^{-1}. The adsorption capacity of carbon is strongly influenced by the chemical structure of its surface, which are of carbon oxygen. Functional groups suggested most often are carboxyl groups, phenolic hydroxyl groups, carbonyl groups (e.g. quinone type), and lactone groups [15]. The chemical composition of PHC is shown in Table 1, and the values are expressed in w/w.

XRD was used to measure the bulk fly ash's chemical composition; the results are listed in Table 2. SiO_2, Al_2O_3, and Fe_2O_3 were the primary components discovered; other elements were present in smaller amounts. The three-dimensional frameworks of $SiO4^+$ and $AlO4^+$ tetrahedra make up the structural components of the zeolites. Chemical analysis and X-ray diffraction (XRD) were used to characterize the structures [15]. Titrimetric techniques were used to analyze $Al2O_3$, Fe_2O_3, CaO, and MgO, whereas gravimetric techniques were used to analyze SiO_2. Flame photometry was used to identify Na_2O and K_2O.

- *Batch Adsorption Procedure*

Individual batch adsorption experiments were performed with each of the three adsorbents to treat tannery wastewater containing Cr, Cu, Zn, and Pb ions. Each solution received a constant adsorbent dose of 2.5 g, which was determined to be the optimal amount beyond which no further significant removal was observed. The pH of the solutions was adjusted to the specific optimal level for each adsorbent-metal combination, ranging from pH 5 to 8, using 0.1 N HCl or 0.1 N NaOH, and maintained throughout the experiment. The mixtures were stirred mechanically at a stable temperature of 27 ± 2 °C for 1 hour to reach near equilibrium. After stirring, the suspension was filtered using Whatman No. 42 filter paper, and the filtrate was analyzed with the DR6000 spectrophotometer to measure the remaining metal concentration.

Table 1 Chemical Composition of Peanut Husk Charcoal

Element	**C**	**H**	**O**	**N**	**Ca**	**Na**	**K**	**Al**	**Fe**	**Si**
% w/w	55	1	15.9	0.5	1.2	2.8	2.6	1	1	19

Table 2 Chemical Composition of Fly Ash and Natural Zeolite

Oxide	**Fly Ash**	**Natural Zeolite**
SiO_2	89.56	45.09
Al_2O_3	4.74	14.43
Fe_2O_3	4.24	10.59
CaO	0.01	5.76
MgO	0.13	4.49
L.O.I.	0.8	14.49
Others	0.52	5.15

- *Data Analysis*

For every metal-adsorbent combination, removal efficiency was computed:

$$\text{Removal Efficiency (\%)} = [(C_0 - C_e) / C_0] \times 100 \qquad (1)$$

where C_e is the residual concentration (mg/L) in the filtrate following treatment and C_0 is the initial metal content (mg/L) in the raw wastewater. To evaluate regulatory compliance, the results for each metal-adsorbent pair were compared to the discharge limitations set by the World Health Organization (WHO, 2022) and the Bangladesh Department of Environment (DoE, ECR 1997).

III. RESULTS

- *Initial Heavy Metal Concentrations*

Examination of untreated tannery wastewater from Hemayetpur, Savar, revealed significant levels of contamination by all four specified heavy metals. Chromium was found at an initial concentration of 5.73 mg/L, greatly surpassing the Bangladesh Department of Environment's (DoE, 1997) limit of 0.50 mg/L and the World Health Organization's (WHO, 2022) guideline of 0.05 mg/L, which aligns with the prevalent use of chromium sulfate in chrome tanning processes. Copper levels were recorded at 1.17 mg/L, which is above the DoE's limit of 0.50 mg/L but still within the WHO's acceptable range of 2.00 mg/L. Lead was present at 0.14 mg/L, which is exceed the DoE's limit of 0.01 mg/L, yet it is nearly fourteen times higher than the WHO's much stricter guideline of 0.01 mg/L, highlighting that even relatively low concentrations of lead pose a significant health hazard. Zinc, at 0.98 mg/L, was the only metal found within both the DoE 5.00 mg/L and WHO 3.00 mg/L permissible limits in the raw effluent.

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Table 3 Initial Concentrations of Heavy Metals in Raw Tannery Wastewater and Corresponding Permissible Limits

Heavy Metal	Initial Conc. (mg/L)	DoE 1997 (mg/L)	WHO 2022 (mg/L)
Chromium	5.73	0.50	0.05
Copper	1.17	0.50	2.00
Lead	0.14	0.01	0.01
Zinc	0.98	5.00	3.00

➢ *Residual Concentrations after Batch Adsorption*

Table 4 presents the residual concentrations of each metal following treatment with PHC, natural zeolite, and fly ash. For Cr, PHC produced the lowest residual concentration 0.85 mg/L, followed by fly ash 1.69 mg/L and natural zeolite 2.43 mg/L; none of these reduced Cr to within the regulatory limits. For copper, natural zeolite 0.21 mg/L and PHC 0.35 mg/L both met the DoE and WHO limits, whereas fly ash 0.65 mg/L satisfied the WHO but not the DoE threshold. All three adsorbents reduced zinc well below both limits, with natural zeolite performing the best 0.16 mg/L. Lead is the most difficult metal to remediate. Although natural zeolite achieved the lowest residual concentration 0.014 mg/L, followed by fly ash 0.036 mg/L and PHC 0.044 mg/L, all three values remained above the strict 0.01 mg/L limit set by the DoE and WHO.

➢ *Removal Efficiency and Adsorption Mechanisms*

Peanut husk, natural zeolite, and fly ash, three economical adsorbents, were assessed for their effectiveness in extracting chromium, copper, zinc, and lead from tannery wastewater. The following outlines their relative performance. Each adsorbent possesses unique characteristics in terms of surface chemistry, porosity, and ion exchange capacity, which significantly influence its ability to bind specific metal ions. By comparing these materials directly, it becomes possible to determine the most suitable low-cost option for each type of contaminant.

Among the adsorbents tested for chromium removal, peanut husk emerged as the most effective due to its cellulosic surface, which is rich in functional groups that facilitate ion exchange with chromium species. Fly ash showed a moderate capacity for adsorbing this metal, likely attributed to its porous aluminosilicate composition. In contrast, natural zeolite was less efficient, possibly due to limited affinity or competitive adsorption under the experimental conditions.

Table 4 Final Concentrations of Heavy Metals After Batch Adsorption Treatment

Metal	Adsorbent	Final Conc. (mg/L)	DoE (mg/L)	WHO (mg/L)
Cr	PHC	0.85	0.50	0.05
	Natural Zeolite	2.43		
	Fly Ash	1.69		
Cu	PHC	0.35	0.50	2.00
	Natural Zeolite	0.21		
	Fly Ash	0.65		
Zn	PHC	0.36	5.00	3.00
	Natural Zeolite	0.16		
	Fly Ash	0.50		
Pb	PHC	0.044	0.01	0.01
	Natural Zeolite	0.014		
	Fly Ash	0.036		

Nevertheless, all three materials significantly reduced chromium levels, with peanut husk standing out as the best option for this specific metal. This indicates that agricultural by-products like peanut husk can compete with, and even outperform, mineral-based adsorbents when chromium is the main pollutant. Its affordability and widespread availability locally enhance its suitability for practical, large-scale wastewater treatment.

Natural zeolite consistently outperformed other materials for copper and zinc, owing to its robust cation-exchange capacity and crystalline structure, which both enhance the absorption of divalent metal ions. Peanut husk showed a moderate effectiveness for these metals, whereas fly ash exhibited the least affinity, suggesting it is more effective for chromium removal than for copper or zinc. These findings highlight natural zeolite as a more adaptable adsorbent when divalent cations are the primary contaminants. The consistent performance across both metals indicates that the exchange mechanism in zeolite is broadly applicable to divalent species rather than being specific to a particular metal. This adaptability makes natural zeolite a promising option for treating wastewater containing a mix of copper, zinc, and similar cations, rather than a single predominant pollutant.

Natural zeolite demonstrated the highest effectiveness in removing lead, attributed to its strong electrostatic attraction and efficient ion exchange at active sites, with fly ash and peanut husk following in performance. Although this was the most effective removal among all metal–adsorbent combinations examined, the lead levels after treatment still exceeded the stringent discharge limits set by regulatory standards. This indicates that while adsorption significantly reduces lead concentration, a single treatment stage might not suffice for full compliance, suggesting the potential need for additional or combined treatment methods. The presence of residual lead, despite high removal rates, highlights the stringent nature of the safety threshold for lead compared to other metals. Consequently, practical treatment systems focusing on lead may need to emphasize polishing or secondary stages rather than solely relying on adsorption.

In general, fly ash lagged behind the other two materials in effectiveness for most metals. Natural zeolite excelled in removing the divalent cations copper, zinc, and lead, while peanut husk was particularly effective for chromium. Zinc was the easiest metal to reduce to safe levels, whereas lead was the most challenging to manage completely. These observations indicate that no single adsorbent is universally the best, and that combining peanut husk with natural zeolite, or using a staged treatment process, might provide a more comprehensive and dependable method for treating tannery wastewater with multiple heavy metal contaminants. Choosing adsorbents based on the specific metal composition of the effluent, rather than depending on one material for all pollutants, seems to be a more practical approach. Additional research into the combined or sequential use of these affordable materials could help apply these laboratory results to a practical field-scale treatment solution.

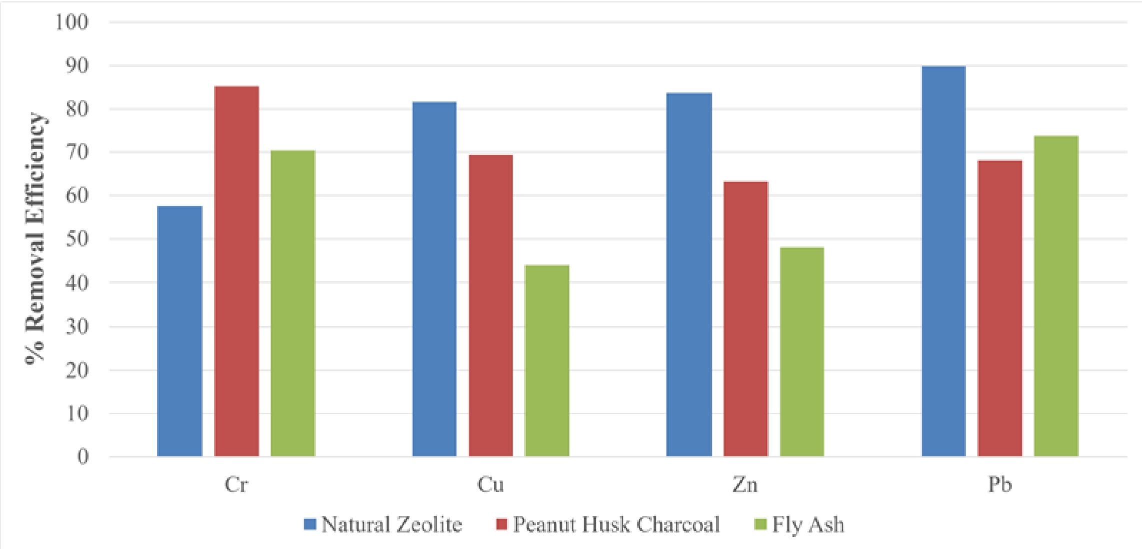

Fig 1 Comparative Removal Efficiency (%) of Cr, Cu, Zn, Pb by Peanut Husk Charcoal, Natural Zeolite, Fly Ash

IV. DISCUSSION

The distinct behavior of chromium compared to the other three metals offers valuable insights. The exceptional ability of peanut husk charcoal to remove chromium aligns with its cellulosic surface chemistry, characterized by carboxyl, phenolic hydroxyl, carbonyl, and lactone groups on PHC surfaces. These groups facilitate various binding mechanisms, including ion exchange, surface complexation, and possibly partial reduction of Cr (VI) to the less mobile Cr (III), which are not as accessible in the ion-exchange-focused zeolite framework. This observation aligns with previous findings that lignocellulosic adsorbents are effective for chromium. On the other hand, the consistent superiority of natural zeolite for copper, zinc, and lead is attributed to its crystalline aluminosilicate structure and its cation-exchange capacity, where framework Na^{+}, Ca^{2+}, and K^{+} ions are easily replaced by the divalent target cations. A similar mechanism has been suggested in other studies to account for zeolite's strong performance with these metals.

Fly ash proved to be the least efficient adsorbent for all metals examined, which aligns with its simpler and less functionalized surface compared to PHC and zeolite. Despite this, it successfully reduced copper and zinc levels to within WHO standards. Considering its negligible cost and widespread availability as a by-product of coal combustion, fly ash could still be useful as an economical pretreatment or blending agent, even if it cannot function as a standalone solution.

From a regulatory perspective, the most significant discovery is that none of the three adsorbents, when used individually in a single batch process, managed to reduce chromium or lead to within DoE or WHO standards, although the reductions achieved were significant with natural zeolite reducing lead by nearly 90% of its initial concentration. The gap arises from the extremely low allowable concentrations for these metals, 0.01 mg/L for lead according to both standards and 0.05 mg/L for chromium as per WHO compared to their initial levels, rather than any shortcoming in adsorption efficiency itself. This indicates that relying solely on single-stage batch adsorption with any of these materials is insufficient. Instead, achieving full compliance will likely necessitate multi-stage treatment, the sequential application of complementary adsorbents (such as a PHC stage followed by a zeolite stage), or the incorporation of a polishing step like precipitation or membrane filtration.

These conclusions must be considered within the limitations of the experimental setup. The trials were performed at a bench scale with constant pH, dosage, contact

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time, and temperature, lacking isotherm or kinetic modeling, repeated trials, or the inclusion of cadmium and nickel, which are known to be present in this effluent. Additionally, the study did not account for the temporal changes in tannery discharge linked to production cycles. Future research should focus on adapting these batch results to a continuous, full-scale treatment system and verifying their reproducibility and long-term regeneration characteristics.

V. CONCLUSION

This study characterized heavy metal contamination in raw tannery wastewater from the BSCIC Tannery Industrial Estate, Hemayetpur, Savar, and evaluated the batch removal performance of three low-cost adsorbents. The raw effluent substantially exceeded the DoE and WHO limits for Cr, Cu, and Pb, with Cr being the most severely elevated (5.73 mg/L). Peanut husk charcoal was the most effective adsorbent for Cr (85.13% removal), whereas natural zeolite exhibited the best performance for Cu (81.64%), Zn (83.73%), and Pb (89.82%) removal. Although fly ash is less efficient overall, it offers practical value as a low-cost supplementary concrete material. Copper and zinc were reduced to within regulatory limits using a single adsorption stage, whereas Cr and Pb remained non-compliant regardless of the adsorbent used, indicating that multistage or hybrid treatment systems are necessary for these metals. Overall, the findings demonstrate that locally sourced, low-cost adsorbents are a technically viable and economically accessible option for the partial treatment of tannery wastewater in Bangladesh, particularly for small- and medium-scale operations with limited capital for conventional wastewater treatment systems. Future work should incorporate adsorption isotherm and kinetic modelling, continuous-flow (column) trials, an expanded panel of target metals including Cd and Ni, and pilot-scale evaluation of multi-stage or combined adsorbent systems to establish design parameters for full-scale implementation.

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