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Hazardous solid waste confined in closed dump sites: an urgent environmental liability to attend

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Abstract: The soil and water contamination by metals from hazardous waste confined with urban solid wastes, highlights the importance of enhance the monitoring of disposal sites once closed. It is common to fail to comply with the regulations on their location, operation and post-closure, and located in areas that affect the environment and the health of the population. In the closed dump of Morelia, contamination of the soil and groundwater by leachates with heavy metals in the water from supply wells has been reported. The objective of this study was to determine the presence of heavy metals and arsenic in the confined wastes of the Morelia closed dump, in order to diagnose the affectation from the contaminants. Composition, degradation status and the presence of heavy metals were analyzed in samples of confined solid wastes from eight wells with different age of confinement. The results of this study ratify the contamination of the leachates of the site and are associated with the contamination of the water for human consumption in the area. The actual regulation does not apply in the case of urban solid waste, so it is crucial to regulate monitoring and management for correct decision-making during post-closure management.

Keywords: metals; arsenic; leachates; pollution; Mexico.

1. Introduction

The environmental pollution is a consequence of the irrational use of natural resources. In general in developing countries, contamination of the soil and groundwater is mainly due to leachates generated at the sites of final disposal of urban solid waste (USW) [1]. This contamination results from the deficiency in the design and operation of the sanitary landfills in which the USW are deposited, as well as the lack of monitoring of environmental legislation [2].

Typically, closed sites are considered environmental liabilities, and as such, geographic sites polluted by the release of materials. Some of the most common landfills contaminants are heavy metals (HM) dissolved in leachates. The impact of these leachates on the environment occurs when their concentrations exceed the maximum permissible limits (MPL), and their bioaccumulation increases the risks for living beings [3]. This effect is increased when the landfill is closed, since the use of closed dump sites as farmlands is a common practice in urban and sub-urban centers in developing countries [4,5].

The concentration and toxicity of the HM depends on: (i) their mobility, associated with their oxidation state. The oxidation states +2, +3 and +4 are the most important since metals form octahedral or plane-square complex ions in solution; (ii) the amount of waste containing HM, although theoretically there should not be heavy metals in the USW, in the reality, hazardous waste that does contain them are thrown away; and (iii) the type of soil. The soil capacity to retain HM is a function of the cation exchange capacity (CEC).

However, its mobilization inside the soil matrix is related to the biological activity and the solid-liquid and water interactions present in the soil [6].

Previous work at the study site [7] reported high concentrations of cadmium, nickel, arsenic, lead, hexavalent chromium, and total chromium in leachates, which exceeded the MPL established in the Mexican legislation, Norma Oficial Mexicana NOM-127-66A1-1994 [8]. In later studies, high concentrations of heavy metals were also reported in USW samples confined at the same site [9,10], which was related to the disposal of hazardous waste in USW landfills, prohibited by the Mexican legislation [11,12] and in any landfill in the world [2].

In general, the so-called landfills of developing countries, and which more closely resemble the category expressed in [2] as open dumps or controlled dumps, are located in areas of high marginalization, characterized by low per capita income [1,13] and high population density; in addition to not complying with environmental legislation operating as dump sites. These characteristics coincide with the northwest of Morelia, in the State of Michoacán, Mexico, where the closed landfill is located. In parallel, this site is located in a fracture zone, with highly porous soils and in a groundwater recharge area. The aforementioned facilitates the production and percolation of leachates from the closed site, since it does not have a geomembrane coating to prevent their escape and the contamination of the groundwater.

Water pollution is a crucial problem that closed USW sites face. Many researchers, as [14] have found high levels of heavy metals in the leachate as Fe, Pb, and Cd and a relatively small proportion of Zn, Cr, and Cu. In the same sense, [15] reported that the study area has significant risk factors regarding the groundwater quality, related to the percolation of agrochemicals in agricultural areas and the inadequate management of wastewater discharged into channels. In addition, in the valley of Morelia-Capula, where the study site is located, the intermediate and local flows of the aquifer system are predominant, which indicates that the water extracted for human consumption is young and that the area functions more as a transit and discharge site than as an infiltration one. The above indicates that the water has a rapid circulation, consequently the infiltrated water is quickly extracted due to the increasing demand for human consumption; being this one of the reasons why exposure to arsenic through drinking water sources is an important problem in Mexico [16].

Accelerated urbanization in developing countries, which has affected the urban metabolism of human settlements, has generated a problem that is reflected in urban and environmental functionality [17]. Such is the case of the study area, which is no longer a predominantly rural area and has become a densely populated human settlement with low economic income, mixed with agricultural and forested areas. The conurbation of this area with the closed dump site and the landfill results in greater exposure and vulnerability to highly polluting and dangerous particles for the population health, and it also poses serious threat to groundwater resources and soil.

The aim of this work is to demonstrate the presence of heavy metals in the USW, clandestinely confined together with hazardous waste due to non-compliance with current environmental legislation, and their probable escape through leachates. In this way, our study provides current data on the increasing environmental and public health risk represented by closed dump sites or controlled dumps sites in developing countries.

2. Materials and Methods

2.1. Physical description of the study area

The study area (16 km long and 8 km wide) (Figure 1) is located in central Mexico, in the Trans-Mexican Volcanic Belt, within the Michoacán-Guanajuato volcanic complex. The area lies in a recharge site of an overexploited aquifer that provides water to more than 120,000 people [18].

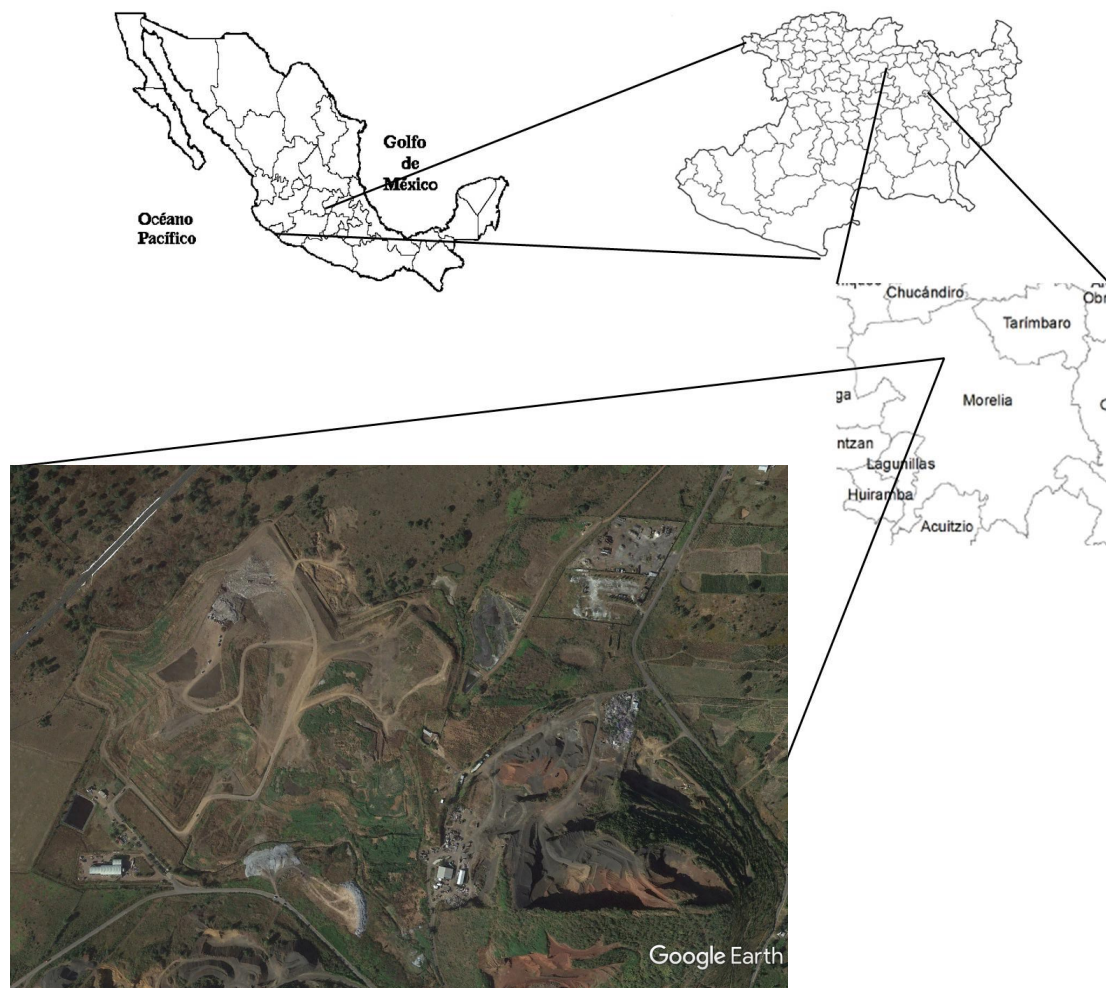


Figure 1. Location of the study area (Modified from Google Earth 2017®; INEGI).

The area presents two important hydrogeological conditions since it is located on permeable geological materials and is affected by a regional fault system. Two semi-shield volcanic bodies dominate the landscape: “El Quinceo-Las Tetillas” in the northeast and “El Águila” in the southwest. The volcanism and the faults determine the relief, with dominant E–W structures, associated with the Morelia-Acambay fault system [19], in which two faults are notorious because of their size, “El Cerrito” and “Cointzio”.

The closed dump site of Morelia, Michoacán, was in operation as an open-air dump until 2007 in which an average of 900 tons of USW per day were deposited, including industrial solid waste, among which the most noteworthy are the matchmaking, oil, car batteries, and resin industries; paint, hospital, slaughterhouse, and papermaking wastes and wastewater treatment solids. The latter two were deposited even when the site was closed (Table 1) and the site closure work involved basically stabilizing slopes and covering the USW with soil.

Table 1. Generation sources identified in the disposal of hazardous waste in the Morelia closed landfill. Source: [20].

SOURCE		Waste
Agricultural equipment stores		Pesticides
		Herbicides
		Fertilizers
		Plaguicide
Hospitals / clinics / doctor's offices / Clinical and radiological analysis laboratories		Sharp objects
		Human waste
		Healing material residues
		Syringes
		Developing material / worn plates
Research laboratories		Reagent containers
		Organisms remains
Veterinary clinics		Dead animals
		Animals waste
		Healing material
Photographic developing workshops		Photographic film waste
		Film waste
		Containers of developing products
Computer, photocopier and printer maintenance workshops		Ink cartridges
		Accessories
		Toner cartridges
		Photocopier oil waste
Beauty salons Slaughterhouses, butchers, chicken shops, guts dispensing		Beauty products
		Dye residues
		Animal waste
Housing		Household cleaners
		Medicines and drugs
		Cosmetics
		Pests, garden and batteries
Sand mines / coal sale		Sand waste
		Coal waste
Electrical workshops		Incandescent lamps
		Light bulbs
		Batteries

Paint stores	Paint containers
	Paintbrushes
	Containers of varnishes, thinner, turpentine, gasoline
	Rag with solvents traces
Gas stations	Oil residues
	Cleaning material residues
Hardware stores	Solvent containers
Car parts stores	Oil containers
	Used car parts
	Antifreeze containers
Auto body shops	Paint containers
	Car parts
	Polisher containers
Garages	Oil containers
	Tires
	Used car parts
	Inner tubes
Construction companies	Cement waste
	Lime residues
	Tile glue residues

2.2. Sampling

The closed dump site area was divided into four quadrants oriented from southwest to northeast. Eight sites were randomly selected, wells were dug to a depth of three meters with a backhoe loader with extension (Case 2002®), and approximately three kilograms of USW sample were taken. Inside the well, the temperature was measured with a digital thermometer. USW samples were placed in black polythene bags, labeled and placed in a cooler for their transfer to the laboratory.

2.3. Sample characterization

The samples were characterized according to the Mexican Official Norm NMX-AA-022-1985 [21], all by-products were manually separated and subsequently grouped into two fractions: organic and inorganic. Then the organic fraction was grouped into categories of degradability according to the classification proposed in [22].

Physicochemical analyzes were performed according to the Mexican Official Norm NMX-AA-052-1985 [23]. The components of the sample were crushed with scissors and ground with an analytical mill (MF 10®) (with a one-millimeter sieve), deposited in plastic jars and frozen at -4°C. Moisture (NOM NMX-AA- 016-1984) [24], pH (NOM NMX-AA-25-1984) [25], total dissolved solids (TDS) NMX-AA-016-1984 [26], and volatile solids (VS) were determined using the 2540G technique from Standard Methods [27].

With a one gram aliquot, each sample was determined for metals by acid digestion of sediments according to the EPA method 3050B [28]. A flame atomic absorption spectrophotometer (FLAA) was used and arsenic was determined with the hydride generation method according to the Mexican Official Norm NMX-AA-051-SCFI-2016 [29]. The analyzes were performed in duplicate.

2.4. Statistical analysis

In order to analyze the presence of significant differences among the metals concentrations and in the content of the organic fraction according to the confinement time of the USW, the results were captured in a database and processed with descriptive statistics and analysis of variance (ANOVA) using JMP 8 software.

3. Results

This section may be divided by subheadings. It should provide a concise and precise description of the experimental results, their interpretation, as well as the experimental conclusions that can be drawn.

3.1. Types of closed dump site waste

In addition to USW, hazardous wastes were identified. Several of them come from 20 different sources, as is the case of chemical containers, which were reported by businesses such as garages, paint stores or hardware stores. The varied list shows from inert (sharp objects) to organisms waste (animals and humans).

The categorization of the USW organic fraction samples showed that 82% of the by-products are of very rapid degradation (Table 2) due to the fact that they mainly come from food residues. 13% of the samples showed moderately slow and slow degradation. This type of degradation is associated with the fraction of residues that, although organic, have a higher content of cellulose and lignin compared to those that come from food residues.

Table 2. Degradability of the organic fraction of the solid residues of the study site (% fresh weight).

Degradability of the organic fraction	%
Very rapid	82.0
Moderately rapid	4.0
Moderately slow	10.6
Slow	2.3

The physical-chemical characterization (Table 3) showed statistically significant temperature averages for both confinement periods. The predominantly basic pH values, together with the low moisture content, directly influence the rate of degradation of the organic matter, although most of it is easily degraded. Total solids (TS) values varied between 58 and 78%. These values are considered high regardless of the USW confinement time. On the other hand, the VS values showed a wide variation, between 17 and 79%. Likewise, the values of the ash, residue of the VS, reaffirm the previous results and corroborate a high variation in the degradation state of the organic fraction of solid waste within the study site.

Table 3. Average values of the physicochemical parameters of the USW with 5 and 10 years of confinement.

PARAMETER	AVERAGE \pm ERROR ESTANDAR	YEARS OF CONFINEMENT	SIGNIFICANCE
Temperature ($^{\circ}$ C)	26.5 \pm 0.44	5	*
	35.0 \pm 0.86	10	*
pH	8.35 \pm 0.03	5	NS
	8.14 \pm 0.14	10	NS
Moisture (%)	31.7 \pm 1.66	5	NS
	31.8 \pm 1.24	10	NS
Total solids (TS) (%)	68.2 \pm 1.66	5	NS
	68.1 \pm 1.24	10	NS
Volatile solids (VS) (%)	73.1 \pm 2.86	5	*
	52.1 \pm 3.65	10	*
Ash (%)	26.8 \pm 2.86	5	*
	49.4 \pm 3.41	10	*

*= Significant. NS=Non significant

The heavy metals in the solid residues of the closed dump site were lead, copper, nickel, zinc, chromium, iron and the arsenic metalloid (Table 4). The values could not be compared with reference values, since currently there is no Official Mexican Norm that specifies the maximum permissible limits of heavy metals in the USW. No significant differences ($p=0.8427$) were found regarding the heavy metal content among wells, despite the different confinement ages of the USW.

Table 4. Heavy metals present in urban solid waste from the dump site (mg/kg). Source: [30].

Well	Pb	Cu	Ni	Zn	Cr	Fe	As
1	47.17	4.17	42.00	209.33	383.00	24740.33	8.83
2	42.67	7.67	54.75	365.17	0.00	23264.83	20.00
3	89.00	744.17	79.00	217.33	13.67	18314.17	60.56
4	55.33	0.00	45.00	116.17	0.00	15012.17	18.55
5	56.00	92.17	53.50	173.17	127.5	18450.17	65.04
6	149.67	8.83	53.00	408.17	3.67	25814.17	22.76
7	108.67	141.00	48.00	165.00	0.00	28431.67	95.85
8	47.83	21.50	57.00	54.33	0.00	31101.67	20.75
Average	74.54	127.44	54.03	213.58	57.19	23141.15	39.04

4. Discussion

The degradation rate of the waste and its physical-chemical characterization allowed evaluating the dump site handling during its operation stage, as well as the degradation behavior of the confined USW and the effectiveness of the site closure measures.

A high percentage of the samples reported a very fast degradation capacity, which, according to [31], is consistent with the high humidity values analyzed ($\pm 30\%$), despite the fact that the samples were collected in the dry season. A higher humidity in the samples allows a better degradation of the residues, since the optimal conditions for the establishment of the microbial consortia that degrade the solid residues exist. The influence of precipitation on the USW humidity has been widely reported [32]. According to the [33] humidity lower than 15% is unfavorable for the degradation of the USW. However, [34] reported a minimum of 10% humidity for USW efficient degradation.

Likewise, both humidity and precipitation play a determining role in the amount of leachates produced, favoring the solubility of the toxic components of USW, due to its role as a catalyst for the degradation processes of hydrolysis and dissolution of toxic components of organic and inorganic matter [35]. Although in this study only the humidity values are reported in the dry season, several visits to the dump site in the rainy season revealed a continuous infiltration of water and a wet consistency in the residues, which suggests an increase in humidity in this season, and consequently, despite the high values reported during the dry season, drastic changes in the moisture contents related to the seasonality of the rains.

Another determining factor in the samples moisture content and the leaching of the residues is related to the site closure conditions. In the case of the closed dump site it was carried out from the leveling and compaction of the solid waste, followed by a covering by a layer of approximately 10 cm of clay and tezontle (volcanic rock). Regarding this, [36] reported the importance of controlling the amount of water in the USW of the landfill once closed, limiting the entry of rainwater and groundwater, by waterproofing the bottom and the landfill cover. Therefore, the humidity values reported in this study confirm that the closure work of this site was not sufficient to adequately isolate the confined solid waste.

Basic pH values indicate that acetogenic anaerobic bacteria are not in their optimal environment, since according to [37], they develop optimally at a pH close to neutrality and are sensitive to pH variations, with an optimum of 7.2. Despite this, these bacteria are active since they are totally inhibited at a pH below 6.0, which would be reflected in an accumulation of organic acids. Notwithstanding the reported pH results, changes in pH can be naturally attenuated due to the regulatory effect of the medium (buffer effect), which depends essentially on the concentration of dissolved carbonates (CO_3), bicarbonates (HCO_3) and CO_2 , as well as on ammonia ions present in the medium. The latter are produced during the anaerobic degradation of proteins. Likewise, organic acids and salts formed during the phases of acidogenesis and acetogenesis contribute to the pH regulating effect.

The deficiency in the closure work does not ensure the achievement of optimal conditions for the establishment of organic matter degrading microorganisms, which delays the stabilization of the USW confined in the closed dump site. On the other hand, [31] also attribute the above to the holocellulose/lignin ratios of lignocellulosic compounds, which are found in waste with a high content of biodegradable organic matter, as is the case of the USW at the study site. However, these same authors report the influence of the confinement time of solid waste ($p=0.01$), which is why more studies are required in this regard, involving a greater number of samples. The wide range of variation in VS content indicates the variability in the state of degradation of solid waste, as well as the time of confinement, establishing four zones that represented 5 and 10 years of confinement at the sampling time. On the other hand, [38] reported that the presence of large concentrations of heavy metals can also retard the stability of the solid waste degradation process.

The results of this study confirm that the USW confined in the closed landfill contain several heavy metals and arsenic. These results, together with the registration of the hazardous wastes that were disposed during its operation, reveal the illegality of the operation of the closed sanitary landfill in Morelia.

In this study, of the contaminants analyzed, only arsenic exceeds the MPL according to the NOM-147-SEMARNAT/SSA1-2004 [39]. However, the MPL of contaminants recognized in Mexican regulations (Table 5) refer to samples of biosolids, water, soil and PECT extract leachate from which the toxic constituents of the residue and its concentration are determined in order to identify if it is dangerous due to its toxicity to the environment according to [40].

Table 5. Maximum permissible limits for metals and arsenic, established in the Official Mexican Norms.

Chemical constituent	NOM-127 (mg/L)	NOM-001 (mg/L)	NOM-052 (mg/L)	NOM-004 (mg/Kg)	NOM-147 **(mg/kg)
Aluminium	0.2	NI*	NI*	NI*	NI*
Arsenic	0.05	0.2	5	41	22
Barium	0.7	NI*	100	NI*	NI*
Cadmium	0.005	0.2	1	39	NI*
Copper	2	6	NI*	1500	NI*
Total chromium	0.05	1	5	1200	NI*
Hexavalent chromium	NI*	NI*	NI*	NI*	280
Iron	0.3	NI*	NI*	NI*	NI*
Mercury	0.001	0.01	0.2	17	NI*
Nickel	NI*	4	NI*	420	1600
Silver	NI*	NI*	5	NI*	NI*
Lead	0.01	0.4	5	300	400
Selenium	NI*	NI*	1	NI*	NI*
Zinc	5	20	NI*	2800	NI*

** Total Reference Pollutant Concentrations (PR_T) by type of land use. NI*: Non included in the Mexican law.

Made from: NOM-052-SEMARNAT 2005 [40], NOM-127-SSA1-1994 [8], NOM-001- SEMARNAT-1996 [41], NOM-004-SEMARNAT-2002 [42], NOM-147-SEMARNAT/SSA1-2004 [39].

A worse scenario is reported for copper, zinc and iron. For these contaminants found in the leachates of the study site, there are no reference values with which to es-

establish whether the reported concentrations represent a danger to human and environmental health.

However, the heterogeneity in the values of the heavy metals and arsenic is due to the variability of the type of waste and the difference in the time of waste confinement; for which the Kruskal Wallis test indicated a significant difference ($p=0.01$), confirming the differences in the degradation state of the residues of the different wells.

The presence of heavy metals and arsenic in the confined residues indicates their presence also in the leachates that are generated. Given the physical characteristics of the location of the closed dump site, the results suggest a probable contamination of the groundwater. The aforementioned highlights the need for the implementation of monitoring to ensure the control of leachates produced at these sites during their operation and after closure. The situation is even more worrying in developing countries where research efforts towards monitoring the environment have not received the desired attention by stakeholders [1]. Furthermore, the retention of the contaminants in solution in the USW matrix is not assured due to the poor biodegradation of the organic matter in the waste, the lack of a geomembrane, the absence of a leachates collection system and the poor coverage of the site [43].

This study provides remarkable evidence on the importance of paying attention to closed dump sites and landfills, and of planning and carrying out the necessary actions for the monitoring of leachates and/or biogas [14], as well as carrying out post-closure maintenance work to avoid and/or decrease the leakage of potential contaminants. Despite the great environmental and health impact due to the poor management of sanitary landfills, in many developing countries they continue to be the main option for the treatment of USW [44], due to the low cost of construction and operation compared to other technologies such as incineration, pyrolysis and gasification. However, sanitary landfills are built without complying with national legislation for their construction and operation and near urban areas. Since soil is a crucial component of urban environments and its management is the key to its quality [45], it is essential to include in construction of sanitary landfills, catchment systems and leachate treatment. And most importantly, develop the necessary tools to strengthen the institutions in charge of complying with environmental legislation, and thus avoid the final disposal of hazardous waste in landfills.

5. Conclusions

The hazardous wastes clandestinely confined for 20 years in the landfill are a potential source of heavy metals and arsenic that are contaminating the groundwater, both due to the physical and social characteristics of the landfill location, as well as due to deficiencies in management during its operation and its later closure.

The concentration of heavy metals and arsenic in the organic and inorganic fractions confirm that the solid residues confined at the study site are the source of contamination for leachates escaping from the same site.

This work demonstrates the non-compliance with the Environmental Legislation regarding the USW disposal, the safe and adequate confinement, and consequently, the control of the HM in the leachates derived from these practices. This study also highlights the importance of including the implementation of operational practices to control and/or avoid those leachates escape, so that they do not reach the streams and/or water channels, affecting the quality of the water for human consumption.

The environmental and public health problems caused by the disposal of USW in the soil make it urgent to review the methodologies for the USW disposal and the operation of sanitary landfills in developing countries.

The results of this study and all referenced agree on the need to implement regulations related to HM MPL in USW and leachate in order to verify the correct operation of sanitary landfills.

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