



Research article

Recycling of nutrients from landfill leachate: A case study

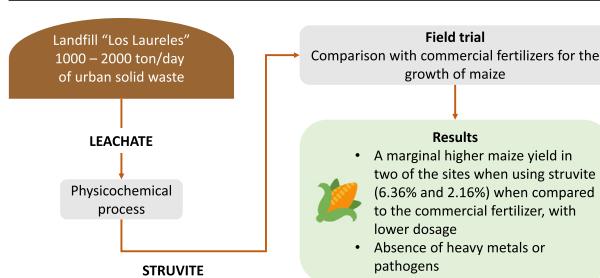
Deborah Lucero-Sorbazo^a, Margarita Beltrán-Villavicencio^b, Abelardo González-Aragón^b, Alethia Vázquez-Morillas^{b,*}

^a CAABSA Eagle, Morelos 2151, Col. Ladrón de Guevara, Guadalajara 44600, Mexico

^b Universidad Autónoma Metropolitana – Unidad Azcapotzalco, San Pablo 180, Col. Reynosa Tamaulipas, Azcapotzalco, Mexico City 02200, Mexico

HIGHLIGHTS

- Landfill leachate can be used as a source of nutrients for the growth of maize by precipitation of struvite.
- A field trial in real scale was performed.
- A marginal higher maize yield was achieved in two of the sites (6.36% and 2.16%) when compared to the commercial fertilizer.
- Struvite did not cause presence of pathogens or heavy metals in the crops.
- It offers an alternative to conventional leachate treatment options, aligned with the principles of the circular economy.

GRAPHICAL ABSTRACT**ARTICLE INFO**

Keywords:
Phosphorous
Nitrogen
Struvite
Toxicity

ABSTRACT

The continuous increase in the consumption of natural resources requires different solutions directed to the recovery and recycling of different materials and products, including the nutrients used as fertilizers for food production. In this context, this research assessed the feasibility of using landfill leachate as a source of nutrients for the growth of maize. Leachate was treated to precipitate struvite, a rich magnesium, phosphate, and ammonium mineral that can be applied directly as fertilizer. It was used for the growth of maize, which was sowed in three different parcels. A commercial DAP + urea mixture was used to compare, and non-fertilized parcels were used as controls. Struvite was successfully obtained and applied in the fields. A marginal higher maize yield was achieved in two sites when using struvite (6.36% and 2.16%) compared to the commercial fertilizer, even if it was applied in a lower dose to weather conditions. An increase in N and Mg in soil could be observed, which allowed for the assimilation of nutrients in the plants. Concerning safety, the use of struvite did not produce the transfer of heavy metals or pathogens to the soil or plants. This research shows a promising way of dealing with leachate, which could be attractive in countries where organic waste is buried in landfills.

1. Introduction

The possibility of nutrient recovery and recycling is considered one of the key elements to move towards the circular economy. It would reduce the depletion of natural resources, the impact of their extraction and

manufacture (Krishnamoorthy et al., 2021; Robles et al., 2020). However, when nutrients such as nitrogen and phosphorous are not recovered, they can become an environmental problem, affecting the quality of water bodies and land. This pollution could happen due to fertilizers runoff (Cui et al., 2020; Wang et al., 2019), discharges from livestock (Li

* Corresponding author.

E-mail address: alethia@azc.uam.mx (A. Vázquez-Morillas).

et al., 2020; Rothwell et al., 2020), and the production of organic solid waste (Luo et al., 2020a,b; Wainaina et al., 2020).

In many developing countries, organic waste is frequently disposed of in landfills and dumpsites (World Bank Group, 2018), producing global warming gases and leachates. Landfill leachate can be heavily polluted and must be treated by biological and physical-chemical processes, frequently combined, to reduce its impact on the environment (Luo et al., 2020a,b). Treatment can be challenging due to the presence of both organic and inorganic pollutants, including ammonia nitrogen. Treatment processes that can remove nitrogen include heating (Schwarzwälder Sprovieri et al., 2020), filtration, oxidation (Wang et al., 2020), adsorption (Halim et al., 2010), and osmosis (Iskander et al., 2018), among other techniques. One of the alternatives to removing nutrients in nitrogen-enriched wastewater and landfill leachate is through precipitation of struvite, which is obtained in the presence of magnesium and phosphorus (Tansel et al., 2018). The production of struvite also has been successful to remove phosphate on industrial wastewater (Nandre et al., 2021).

Struvite is a hydrated compound with equimolar concentrations of Mg, ammonium, and phosphate ($MgNH_4PO_4 \cdot 6H_2O$). It is characterized by orthorhombic crystals, which can be pyramidal or tubular, among other shapes, with white-yellow color (Anthony et al., 2001). It has a molecular weight of 245.43 g/mol, it has low solubility in water in neutral and basic pH (<5%), but is easily soluble in acid media (Chirmuley, 1994; Gu et al., 2021). It can be used as a slow-release fertilizer due to its low content of heavy metals (Hu et al., 2020; Sánchez et al., 2011). It has been shown that the use of struvite as a multi-element fertilizer prevents water pollution due to its low solubility, preventing washing by rain and irrigation (Li et al., 2019; Mikula et al., 2020). It can help reduce the extraction of resources as phosphorus, which faces the risk of depletion (Nesme et al., 2018). It also diminishes the high energy consumption of fertilizers containing nitrogen (Land and Water Division, 2002), and can be used to supply phosphorous in hydroponic production (Arcas-Pilz et al., 2021). The possibility of recycling nutrients is desirable for countries like Mexico, where agriculture is a relevant economic activity; in 2019 Mexico was the 7th producer of maize worldwide, based mainly on small farmers, which account for 60% of the production (SADR, 2019).

In this context, this research evaluates the feasibility of recycling nutrients extracted through the precipitation of struvite from landfill leachate and its use as a fertilizer in the growth of white maize in a full-scale field test. As urban solid waste in Mexico contains 46.62% of organics, and is mainly buried in landfills without segregation (SEMARNAT, 2020), the production of leachate with high nutrients content can be expected. For this research the leachate was obtained from the treatment plant of the landfill "Los Laureles." Struvite was applied in plots located in the agricultural area close to the landfill in the state of Jalisco, which traditionally has been the second producer of maize in the country, with 14% of the total production (ASCDMA, 2018). This study aimed to assess the comparative yield of maize produced in struvite-fertilized plots and evaluate possible risks due to the dissemination of pathogens and heavy metals. Obtained knowledge will contribute to a better understanding of the possibility of closing the cycle of nutrients in the region.

2. Materials and methods

Struvite was produced by precipitation of the leachate produced in the landfill Los Laureles, operated by CAABSA in the municipality of El Salto, Jalisco, México. After its characterization, it was used as fertilizer in a field trial in the growth of maize.

2.1. Production and characterization of struvite

The production was performed in the leachate treatment plant of the landfill, according to the method proposed before (Di Iaconi et al., 2010) by the addition of MgO (98%), H_3PO_4 (85%, pH < 0.05) and NaOH (50%,

pH = 14), to reach a 2:1:1:1 M ratio (Mg:NH₄:PO₄) and a final pH of 9.2. Mature leachate (2000 l) was added to a reactor and mixed with MgO for 10 min at 500 rpm. The mixture was poured into a settling tank, which allowed the separation of struvite and the treated effluent. Struvite was dried and stored. The obtained mineral was characterized in the Electronic Microscopy Laboratory at Universidad de Guadalajara, in a MIRA Tescan SEM. Qualitative chemical analysis was done by dispersive X-ray, in a SUPRA 55VP Zeiss equipped with an Bruker—S3 energy-dispersive X-ray spectroscopy (EDS) for elemental maps, using an accelerating voltage of 15 kV and 129 nm resolution. Quantitative analysis was done for the concentration of nitrogen (micro-Kjeldahl), phosphorous (Bray-I technique), and potassium (atomic absorption spectrophotometry).

2.2. Field test

The goal of the field test was to assess the viability of the use of struvite as a fertilizer in real conditions. The test was organized in three stages: a) project design; b) sowing and addition of fertilizers and pesticides; and c) assessment of the process.

2.2.1. Project design

The yield of maize was assessed in three 0.25 ha plots, located in sites called Hierbabuena, La Mesa, and La Tarjea, in the municipality of Zapotlanejo. These places are located near to the landfill, in the center of Jalisco state. Each parcel was divided into three 833 m² sections to test different experimental conditions: a) negative control, without fertilizer (CTRL), b) use of commercial fertilizers DAP and urea (DAP); and c) use of struvite as fertilizer (STRV).

Hybrid white maize P3055W for human consumption (Pioneer®) was used, given its prevalence among local farmers. These seeds have a minimal seeding of 85%. They are previously treated with Fluidioxonil, Metalaxyl-M, Thiamethoxam, Azoxystrobin, Clorantraniliprole, and Thiabendazole, to protect them against plagues during storage and in the first 20 days after sowing. As this variety has a six to eight months growth cycle, the field test began in June 2018 and finished in December 2018, when the plants were harvested.

2.2.2. Sowing, addition of fertilizers and pesticides

The sowing was performed according to local practices regarding the use of machinery, irrigation, and the application of pesticides to prevent plagues. Regarding the addition of fertilizers, the basic idea was to provide the required dosage of nitrogen. The Mexican national authority for agriculture, SAGARPA, recommends a dose of 240 kg of N for ha to produce eight tons of maize (SAGARPA & INIFAP, 2015). A mass balance, shown in Table 1, was performed to define the required dosage of fertilizers, based on their composition (De Vries et al., 2017; Lépiz et al., 2015).

Local farmers apply 250 kg/ha of DAP (45 kg N/ha) and 500 kg/ha of urea (200 kg N/ha), i.e., a total N load of 245 kg/ha. While DAP is applied during sowing, 200 kg of urea are added between days 24–30 after sowing, and 300 kg at tasselling. The mass balance resulted in a struvite requirement of 8,166.92 kg/ha, applied in different stages. However, only the first dosage of 2,144.16 kg/ha was applied due to the flooding of adjacent lands, limiting access to the plots during the test. Due to this situation, only 26.25% of the theoretical requirement of struvite was applied, as shown in the supplementary material.

Table 1. Composition of fertilizers.

Fertilizer	N–P–K (mass ratio)
Struvite	3-10-0.9*
Urea	40-0-0
Commercial fertilizer (DAP)	18-46-0

* Lab results of struvite produced in Los Laureles.

Irrigation was performed according to local practices (seasonal rain-water irrigation), guaranteeing similar conditions for plots located in each area. The most common plagues for maize in the area are *Spodoptera frugiperda*, *Helicoverpa zea*, and *Atta Mexicana*. According to local practices, the pesticides Flash Ultra, Calibre 90, Anaclor and Glifosato were added to prevent their attack.

2.2.3. Assessment of the process

The growth of maize was monitored by inspection of the size and conditions of the plants. The harvest was done manually to allow the separation of the product in the different experimental treatments. The assessment of the process for each combination of soil and fertilizer had two main objectives: to assess the efficiency of struvite as fertilizer and verify that it does not transfer heavy metals, pathogens, or other undesirable characteristics to the soil or the grain. For this purpose, the yield of the field (for grain and maize crop biomass) was calculated, and a linear regression analysis was performed to identify possible relationships between Mg concentration and yield of maize. The characterization of soil and grain was performed according to the methods listed in the Supplementary material.

The samples of soil were sundried, mixed, and sieved (<2 mm). They were digested following the method EPA 3051^a (EPA, 2007), with an acid solution containing 25% of HCl and 75% of HNO₃ (vol/vol) in a microwave (CEM, MARSX press) for 25 min. Grain samples were also dried, mixed, and sieved, and then they were digested in a solution of HClO₄-HNO₃ (Armienta et al., 2008). Pb, Cd, and Mg concentration were measured by atomic absorption spectrophotometry in a PerkinElmer PinAAcle 900 F (detection limit 0.05 mg/L) following the method SW-846 7000 (EPA, 2003). Arsenic was measured by hydride generation in an external certified lab, with a detection limit of 0.005 mg/L. All the analyses were done in duplicates.

The presence of total coliforms and *Salmonella* spp was assessed, adapting locally developed protocols (Castañeda, 2004; PPTAR, 2014). Sieved soil and whole grains were suspended in sterilized casein peptone solution, and they were then mixed in a vortex. Tenfold dilutions were filtered and added to Petri dishes containing BBL (*Salmonella*) or Difco m FC (coliforms) agar, to be incubated for 24 h at 35 ± 2 °C (coliforms) and 48 h at 35 ± 2 °C (*Salmonella*). Deionized sterilized water and a solution containing both pathogens were used as negative and positive controls. After incubation emergence of colonies was assessed by direct inspection. All microbial tests were done in triplicates.

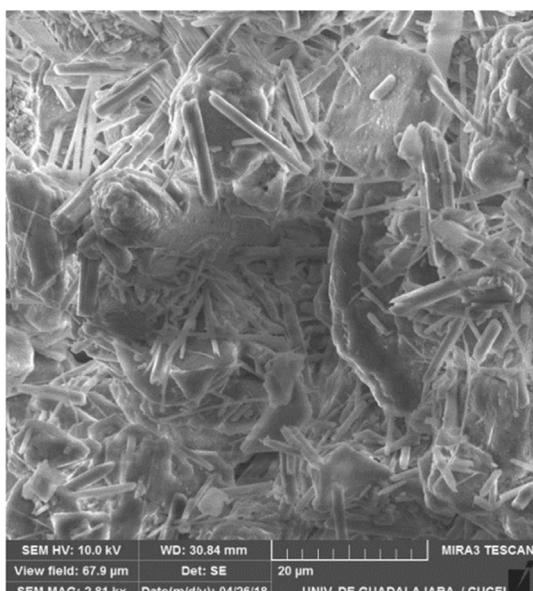


Figure 1. SEM images from struvite obtained in this research.

3. Results and discussion

3.1. Characterization of struvite

A yield of 27.92 kg struvite (dry basis) for each cubic meter of leachate was achieved. It had a specific gravity of 0.54 t/m³ on dry basis. The 86% of ammonia nitrogen and 89% of phosphorous was removed from leachate, achieving a final suspended solids concentration of 20.5 mg/L. The struvite obtained by precipitation of the leachate showed two crystalline forms: regular round shapes and long prisms, as shown in the SEM image (Figure 1). The observed shapes are similar to those obtained by treating the leachate of the Hong Kong landfill (Li and Zhao, 2003).

The identity of the struvite was confirmed by energy-dispersive X-ray spectroscopy analysis (Figure 2), which showed a typical profile of the mineral. This test also allowed us to identify the presence of potassium in regular crystals of struvite, which could improve the efficiency of struvite as fertilizer. In addition, the presence of other elements in the mineral can be related to the raw material used to prepare the mineral; previous research has shown that besides Mg, other nutrients such as Ca, K, Na, and Fe can be present in struvite (Uysal et al., 2014).

The concentration of heavy metals found in the obtained mineral is similar to the one reported, showing a higher level of potassium and calcium. The molar relation obtained was 2Mg:1NH₄:1.2PO₄, similar to the one reported previously (Di Iaconi et al., 2010). However, the molar ratio of the obtained mineral differs from the stoichiometry of struvite (Mg:N:P = 1:1:1), possibly due to the coprecipitation of struvite along with other minerals such as magnesium phosphate, calcium phosphate, and magnesium hydroxide. The elemental composition and its comparison with reported values are presented in the Supplementary material.

The NH₃:PO₄³⁻ ratio obtained in struvite in this research (0.34:1) is lower than previously reported values of 0.43:1 (Di Iaconi et al., 2010) and 0.59:1 (Szymanska et al., 2019), while similar to the obtained by others (Uysal et al., 2014; Warmadewanthi et al., 2021). This ratio is the result of factors such as the crystallization process, the proportion of Mg, the pH, the aeration rate, the retention time, and the temperature (Rahman et al., 2014). It is also related to the initial molar ratio Mg:NH₄:PO₄, which was 2:1:1.2, different from previously used values of 2:1:1 (Di Iaconi et al., 2010) and 1.5:1:1 (Uysal et al., 2014).

The concentration of metals was lower when compared to phosphate fertilizers, which can include 2–1,200 mg As/kg, 7–225 mg Pb/kg, 7–179 mg Cd/kg, 1–12 mg Co/kg and 0.1–0.12 mg Hg/kg (Kabata-Pendias, 2011). These low metal content has been reported previously for struvite produced from the effluent of the yeast industry (Uysal et al., 2014), and landfill leachate (Li and Zhao, 2003). The metal content also complies with the maximum levels for the national regulations for biosolids (SEMARNAT, 2002) and soils intended for agriculture (SEMARNAT, 2004).

However, it must be noticed that presence of ionic species may hinder the efficient of struvite recovery, product purity, morphology, and reaction speed. High concentrations of Ca in particular are known to precipitate simultaneously as phosphate salts along struvite (Krishnamoorthy et al., 2021; Pastor, 2008), mainly at pH > 10. Soluble ions could also precipitate due to chelation into struvite (Nandre et al., 2021; Warmadewanthi et al., 2021). Coprecipitation of other minerals, such as hydroxyapatite, sulfohalite, and trisodium dipotassium triphosphidolite has been reported before (Warmadewanthi et al., 2021), affecting the yield of struvite formation. While no further analysis was performed to identify specific minerals, it can be assumed that not all the obtained mineral was struvite, due to the presence of calcium.

3.2. Harvest of maize and assessment of yields

The maize was harvested in December, six months after sowing. The complete process, from land preparation to tillage of stubble, is shown in Figure 3.

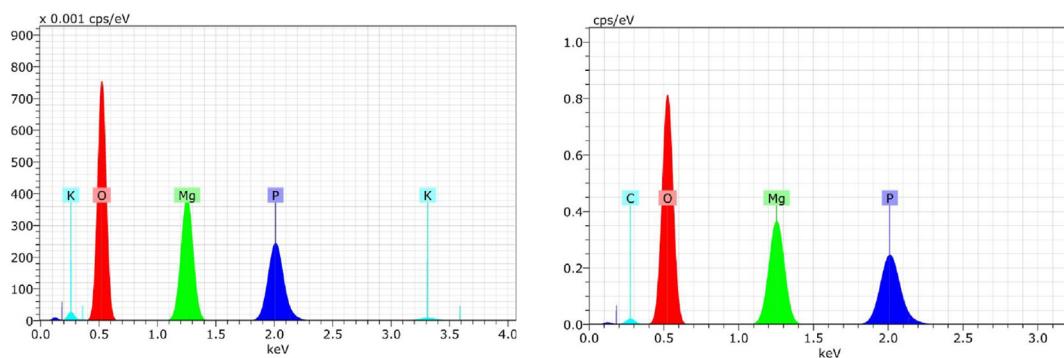


Figure 2. EDS spectrum from irregular crystals (left) and long prisms (right).

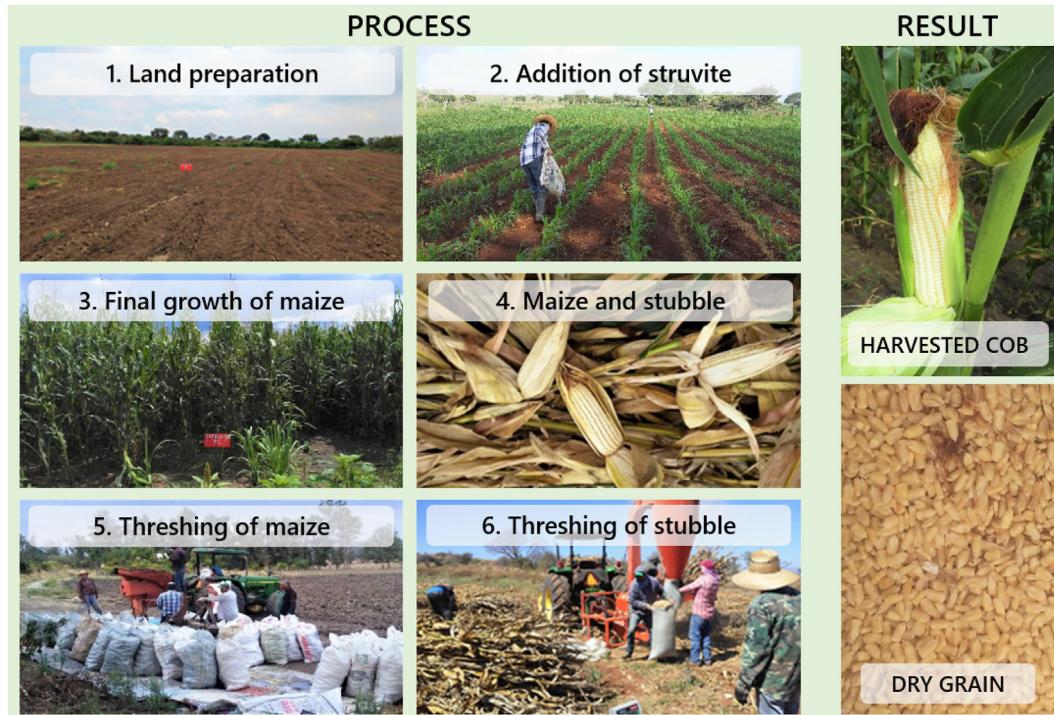


Figure 3. Sowing and harvesting of maize process.

The production of maize when struvite was added as fertilizer was higher in two of the three sites (Figure 4). At La Mesa, 4,165.76 kg/ha were obtained, showing a yield of 6.36% higher than the commercial fertilizer and 8.38% than the control. A similar result was observed in La Tarjea, where the production with struvite (3,368.5 kg/ha) was 2.16%

and 33.8% higher than the yield observed for the commercial fertilizer and the control, respectively. On the other hand, in Hierbabuena, the higher production was achieved in the control (4,059.84 kg/ha), probably due to the poor drainage conditions that lead to flooding in the plots where struvite and the commercial fertilizer were used.

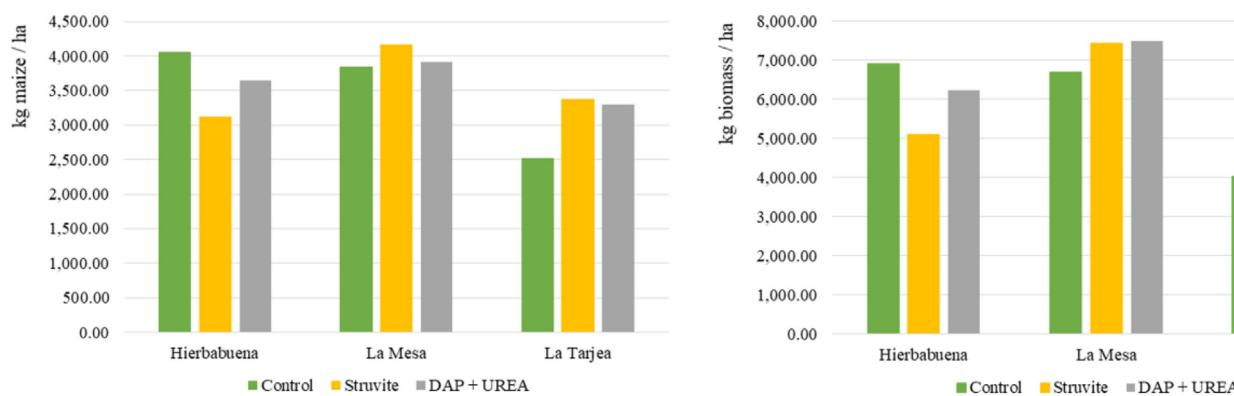


Figure 4. Production of maize (left) and biomass (right) for different locations and treatments.

In general, struvite showed a very competitive performance (Table 2), considering that it was applied in a lower dosage than the DAP + urea mixture (73.3% less than required). This result coincides with previous research, where struvite was compared to ammonium phosphate, showing similar or higher yields (Szymanska et al., 2019). Furthermore, the addition of fertilizers also increased the size of the grains. For example, the mass of 50 grains showed that in La Tarjea and La Mesa, fertilized soils increase up to 8.7% in the mass of the grains.

Production of biomass in La Mesa and La Tarjea was higher for the DAP + urea mixture than for struvite (6.22% and 0.83% for each site). This increase can be explained by a higher mass of stubble when the commercial product was used. In Hierbabuena, as it happened for maize, the yield was higher for the non-fertilized plot. The results of La Mesa and La Tarjea showed that while struvite increases maize production, the DAP + urea mixture increases the yield of stubble. The positive effect due to the addition of struvite coincides with previous research where maize, tomato, and grass showed higher biomass yield when compared to commercial formulae (Liu et al., 2011; Szymanska et al., 2019; Uysal et al., 2014). Li and Zhao (2003) found similar results for four fast-growing (*Brassica parachinensis*, *Brassica rapa* var. *chinensis*, *Ipomoea aquatica*, and *Ipomea aquatica*, *I. reptans*). They found that the overdosage of struvite (2–8 times) did not harm the growth of the plants due to their low solubility. When struvite was compared to mineral phosphate fertilizers in the growth of *Lactuca sativa L.*, the plants rendered higher fresh biomass and P absorption, explained by the authors by a synergistic effect produced by the presence of Mg (González et al., 2009).

3.3. Effects of struvite in nutrient assimilation

The characterization of grain is shown in Table 3. The use of struvite did not affect the organoleptic characteristics of the maize. The level of nitrogen (10.50 ± 0.42 to 13.25 ± 0.78) is close to previously reported values, which go from 14% in unfertilized tests to 15.1% in Zn-enriched soils in the interval commonly reported for maize (Puga et al., 2013). The lower levels obtained can be related to the initial quality of the soil (Kabata-Pendias, 2011). There was no clear tendency in the assimilation of N derived from the use of the fertilizers, as struvite showed better performance than the commercial mixture in one site and lower values in the others. This finding, different from previously reported results (Puga et al., 2013), can be attributed to the lower dosage of struvite applied when compared to the DAP + urea fertilizer. Despite the lower mass added, struvite also increased nitrogen content in the soil above the commercial mixture.

Content of phosphorous in maize has been reported in the intervals of 3.0 a 3.1 g/kg in Zn-enriched soils (Puga et al., 2013), and 2.9 g/kg in dry grain (Uhart and Echeverría, 1998). In this study, the grains obtained in plots where fertilizer was used showed a phosphorous concentration of 2.1 a 3.05 g/kg, coincident with previously reported results. The maize without fertilizers (CTRL) assimilated less phosphorous; however, it is within limits described as expected (2.1 mg/kg) (TA, 2019). Other authors have found that struvite increases the assimilation of nitrogen and phosphorous when compared against commercial fertilizers (González et al., 2009; Puga et al., 2013; Szymanska et al., 2019; Uysal et al., 2014), the variable results obtained in this research can be attributed to the lower dosage of struvite applied when compared to the DAP + urea

Table 2. Yield of maize in fertilized plots.

Site	Commercial formulae (DAP + urea) (ton/ha)	Struvite (ton/ha)	Increase of yield when using struvite
Hierbabuena	3,642.16	3,114.72	- 14.48 %
La Mesa	3,916.80	4,165.76	+ 6.36 %
La Tarjea	3,297.00	3,368.5	+ 2.17 %

Table 3. Final characterization of maize grains and soils for the different sites and fertilizers.

GRAIN	Hierbabuena			La Tarjea		
	CTRL	DAP	STRV	CTRL	DAP	STRV
N (g/kg)	12.75 ± 0.049	13.25 ± 0.78	11.75 ± 0.35	10.50 ± 0.42	10.95 ± 0.71	11.45 ± 0.21
P (mg/100 g)	236.60 ± 8.54	286.65 ± 10.91	249.62 ± 11.4	249.20 ± 9.76	298.2 ± 9.93	279.58 ± 8.44
Mg (mg/100 g)	128.59 ± 3.12	132.05 ± 5.61	145.81 ± 2.91	137.58 ± 6.11	142.05 ± 4.18	138.91 ± 3.42
Mass of 50 grains (g)	17.82 ± 0.22	18.51 ± 0.19	18.31 ± 0.20	18.05 ± 0.32	17.70 ± 0.27	18.09 ± 0.16
SOIL						
N (%)	0.11 ± 0.01	0.08 ± 0.00	0.11 ± 0.00	0.06 ± 0.00	0.08 ± 0.00	0.11 ± 0.00
P (mg/kg)	16.84 ± 0.22	20.58 ± 0.35	25.17 ± 0.44	21.88 ± 0.72	30.80 ± 1.88	30.98 ± 0.24
Mg (mg/kg)	1390.1 ± 20.8	1413.94 ± 34.2	1714.3 ± 18.8	1991.5 ± 57.75	2013.3 ± 28.18	2080.2 ± 18.51

CTRL = Control; STRV = struvite; DAP = Fertilizante comercial DAP + urea.

Table 4. Presence of metals and pathogens in soil and maize grains.

	La Mesa			Hierbabuena			La Tarjea		
	CTRL	DAP	STRV	CTRL	DAP	STRV	CTRL	DAP	STRV
GRAIN									
Pb (mg/kg)	ND	0.29 ± 0.01	ND	0.1 ± 0.0	ND	ND	0.297 ± 0.011	ND	0.095 ± 0.004
As (mg/100 g)	ND	ND	ND	ND	ND	ND	ND	ND	ND
Cd (mg/kg)	ND	ND	ND	ND	ND	ND	ND	ND	ND
Fecal coliforms	+	+	+	-	-	-	-	-	-
Salmonella spp.	+	+	+	-	-	-	-	-	-
SOIL									
Pb (mg/kg)	13.25 ± 0.61	16.58 ± 0.50	23.83 ± 1.03	15.23 ± 0.27	17.45 ± 0.11	12.53 ± 0.31	24.54 ± 0.05	20.8 ± 0.04	23.66 ± 0.10
As (mg/kg)	1.09 ± 0.05	1.09 ± 0.05	0.98 ± 0.05	1.29 ± 0.07	1.2 ± 0.07	0.98 ± 0.06	1.2 ± 0.04	1.2 ± 0.04	1.2 ± 0.04
Cd (mg/kg)	0.04 ± 0.00	0.04 ± 0.00	0.038 ± 0.00	0.069 ± 0.003	0.07 ± 0.003	0.05 ± 0.003	0.44 ± 0.01	0.74 ± 0.04	0.77 ± 0.03
Fecal coliforms	+	+	+	-	-	-	-	-	-
Salmonella spp.	+	+	+	-	-	-	-	-	-

(+): Present (-): Absent ND: non detected.

CTRL = Control; STRV = struvite; DAP = commercial mixture DAP + urea.

fertilizer. The use of fertilizers increased phosphorous content in the soils, with higher values for struvite in La Mesa and Hierbabuena. This improvement in the content of this nutrient in soil when using struvite has been reported before (Puga et al., 2013).

All the sites had a high content of Mg, and a medium level of exchangeable Mg (SEMARNAT, 2000), which increased with the use of fertilizers. A more significant increase was observed when struvite was applied. The average concentration of magnesium in maize grains reported in the literature is 127 mg/100 g (TA, 2019), a comparable value to the one obtained in soils without fertilization. In La Mesa and La Tarjea, higher content of Mg was observed in the ones where struvite was applied, as reported by Uysal et al. (2014). The concentration of Mg obtained in our research when applying struvite is similar to previously reported results (Puga et al., 2013). It has been found that lower concentrations of this element could be related to low baseline concentrations in soil. On the other hand, a synergistic effect has been reported between the presence of Mg in struvite and the high absorption of P by plants (González et al., 2009). According to the obtained model, the statistical analysis, which involved the tree sites and three experimental conditions, showed a significant effect from the concentration of Mg in soil with the maize yield, according to the obtained model, shown in the supplementary material.

The soils of La Mesa and La Tarjea had acidic pH values, while Hierbabuena reached a moderately alkaline final pH (7.4–8.5). Maize grows well in all kinds of soils; however, the optimal pH for the plant is in the 6.0–7.0 pH interval (CONACYT, 2019). It has been found that pH in the soil does not affect the effectiveness of struvite as a fertilizer (Achat et al., 2014; Talboys et al., 2016); however, in extreme conditions, a better performance was observed under acidic conditions (De Vries et al., 2017). It also has been proved that acidic exudates from the rhizosphere increase the solubility of struvite (Vaneeckhaute et al., 2015).

3.4. Safety assessment of the use of struvite

One of the main concerns related to the use of landfill leachate to produce struvite is the possible transfer of pollutants, mainly heavy metals and pathogens, to the soil and plants. In Mexico, the legal regulation that fixes the sanitary limits for the production of grains, NOM-247-SSA1-2008 (SS, 2008), sets a maximum Pb content of 0.5 mg/kg. In this research, none of the experimental conditions exceeded that limit (Table 4). The presence of Pb in the soil only was detected in the traditionally fertilized plot in La Mesa, the control, and the struvite plots in La Tarjea. In all cases, this can be attributed to the baseline concentration in soil, which ranged from 12.5 to 24.54 mg/kg, into the allowed range established in the Mexican norm for soil intended for agriculture (400

mg/kg) (SEMARNAT, 2004). The obtained struvite has less than 0.500 mg/kg of Pb, and no correlation was observed related to its use. The neglectable contribution of Pb from struvite has been reported before (Uysal et al., 2014). The regulation mentioned above sets maximum levels of 22 mg/kg for As and 37 mg/kg for Cd. These metals were not found in the grains, and their level in soils, in all cases below the limits, can be considered as typical baseline concentrations (Kabata-Pendias, 2011).

Pathogens were found only in grains and soils of La Mesa. This finding is highly likely a consequence of the soil contamination, which could be caused by nearby cowsheds and the use of water from the river Santiago for irrigation. This river receives untreated wastewater discharges, as shown in the supplementary material, and has been assessed as highly polluted (IACHR, 2020).

3.5. Cost analysis

Production cost for struvite in this research was 629.8 USD/ton. This cost is high when compared to reported prices for struvite (Table 5). Compared with current alternatives in Mexico also shows a higher cost when using struvite; while struvite had a total cost, considering the applied dosage, of 1370 USD/ha, the cost of DAP + urea was 271 USD/ha. However, the additional benefits of using struvite must be taken into account: first, it allows recovering nutrients from solid and liquid wastes. Second, it is a more complex slow-release fertilizer, contributing to primary and secondary nutrients, such as N, P, K, Mg y Ca, while DAP is

Table 5. Production and sale costs reported for struvite.

Country	Production cost (USD/ton)	Sale cost (USD/ton)
Japan (Ueno and Fujii, 2001)	NR	295 + transportation cost
Australia (Doyle and Parsons, 2002)	140	877
Japan (Doyle and Parsons, 2002)	460	1885
United Kingdom (Doyle and Parsons, 2002)	NR	283
Average for different countries (Molinos et al., 2011)	NR	902
Estimation of market price (Molinos et al., 2011)	NR	222–902
The Netherlands (De Vries et al., 2017)	NR	60 in the country 600 if exported to another continent
USA (Ishii and Boyer, 2015)	NR	570

mainly a nitrogen source. As struvite needs only one application, its use can lead to lower farming costs.

The potential production of struvite for landfill leachate in the state of Jalisco, based on a daily generation of 5000 t/day with 30% moisture, is 41.8 kg/day. However, yield and selectivity could be improved by the optimization of the operation parameters. Other alternatives that would decrease the cost would be the use of industrial waste as a source of phosphorous, decreasing the need to buy reactants. Potential sources of this element include waste bones ash from nearby farms (Darwish et al., 2017) and residual phosphoric acid. On the other hand, the use of cheaper Mg sources, such as MgO (Huang et al., 2014; Stolzenburg et al., 2015), MgSO₄, MgCl₂ (Di Iaconi et al., 2010) or even seawater could be assessed (Shin and Lee, 2010). Additional options to decrease the cost of the process include pretreatment to eliminate ions such as Ca and increasing the yield to struvite (Warmadewanthi et al., 2021), recycling of struvite in more than one cycle (Huang et al., 2014), or its application as the first stage on a treatment train. It should be noticed that the production of struvite would substitute current technologies used for leachate treatment, such as reverse osmosis, whose cost has been estimated as US\$ 8.58/m³ (Almeida et al., 2020). In the region, the cost of leachate treatment ranges from US\$ 15/m³ to US\$ 30/m³; the production of 1 t of struvite would eliminate the cost of treatment from approximately 35 m³ of leachate, which would cost US\$525- US\$1059. This avoided treatment cost improves the economic feasibility of the process.

4. Conclusions

This research shows the feasibility of the recovery of nutrients from landfill leachate for their use as fertilizers in the production of maize. Struvite, the obtained fertilizer, showed a similar performance to the commercial control, even if it was applied in lower soil:fertilizer proportions. The results are promising, as the process would contribute to the recycling and conservation of natural resources.

Leachate can include different pollutants coming from waste that could transfer to soil and maize through the use of struvite. Nevertheless, no adverse effects were observed regarding pollutants transfer in this field test. However, it must be considered that the composition of leachate is inherently variable, so continuous monitoring would be required to guarantee the safety of struvite in terms of possible migration of metals, pathogens, and other contaminants. Another issue to be solved is the reduction in the production cost of struvite, which currently could be seen as a limitation if the product is commercialized and compete with fertilizers currently in the market. However, the equilibrium point could be reached if those costs and the externalities related to the current leachate management are considered.

On the other hand, the process could render different environmental benefits, such as diminishing the need to extract nutrients to produce fertilizers, the decrease of the nutrients load in the leachate, and their reincorporation into the production cycles. This alternative would be especially attractive to countries like Mexico, where the burial of organic waste in landfills is still a common practice and where farming is also practiced in a non-industrialized way.

Declarations

Author contribution statement

Deborah Lucero-Sorbazo: Conceived and designed experiments; Performed the experiments; Contributed reagents, materials, analysis tools or data.

Margarita Beltrán-Villavicencio: Performed the experiments; Contributed reagents, materials, analysis tools or data; Analyzed and interpreted data; Wrote the paper.

Abelardo González-Aragón: Analyzed and interpreted data.

Alethia Vázquez-Morillas: Analyzed and interpreted data; Wrote the paper.

Funding statement

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Data availability statement

Data included in article/supp. material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

Supplementary content related to this article has been published online at <https://doi.org/10.1016/j.heliyon.2022.e09540>.

Acknowledgements

The authors want to acknowledge the help of Universidad de Guadalajara in the development of some analysis.

References

- Achat, D.L., Daumer, M., Sperandio, M., Santellani, A.C., Morell, C., 2014. Solubility and mobility of phosphorus recycled from dairy effluents and pig manures in incubated soils with different characteristics. *Nutrient Cycl. Agroecosyst.* 99, 1–15.
- Almeida, R. de, Bila, D.M., Quintaes, B.R., Campos, J.C., 2020. Cost estimation of landfill leachate treatment by reverse osmosis in a Brazilian landfill. *Waste Manag. Res.* 38 (10), 1087–1092.
- Anthony, J.W., Bideaux, R.A., Bladh, K.W., Nichols, M.C., 2001. *The Handbook of Mineralogy*. Mineral Data Publishing, Chantilly, VA.
- Arcas-Pilz, V., Rufi-Salís, M., Parada, F., Petit-Boix, A., Gabarrell, X., Villalba, G., 2021. Recovered phosphorus for a more resilient urban agriculture: assessment of the fertilizer potential of struvite in hydroponics. *Sci. Total Environ.* 799, 149424.
- Armitage, M.A., Ongley, L.K., Rodríguez, R., Cruz, O., Mango, H., Villaseñor, G., 2008. Arsenic distribution in mesquite (*Prosopislaevigata*) and huizache (*Acacia farnesiana*) in the Zimapán mining area, México. *Geochem. Explor. Environ. Anal.* 8, 191–197.
- Agencia de Servicios a la Comercialización y Desarrollo de Mercados Agropecuarios (ASCDMA), 2018. Maíz grano cultivo representativo de México [WWW Document].
- Castaneda, B.M.T., 2004. *Microbiología Aplicada. Manual de Laboratorio*. Universidad Autónoma Metropolitana Unidad Azcapotzalco. México, D. F.
- Chirmuley, D.G., 1994. Struvite precipitation in WWTPs causes and solution Water. *J. Aust. Water Assoc.* 21–23.
- Consejo Nacional de Ciencia y Tecnología (CONACYT), 2019. Maíz [WWW Document].
- Cui, N., Cai, M., Zhang, X., Abdelhafez, A.A., Zhou, L., Sun, H., Chen, G., Zou, G., Zhou, S., 2020. Runoff loss of nitrogen and phosphorus from a rice paddy field in the east of China: effects of long-term chemical N fertilizer and organic manure applications. *Glob. Ecol. Conserv.* 22, e01011.
- Darwish, M., Aris, A., Puteh, M.H., Jusoh, M.N.H., Abdul Kadir, A., 2017. Waste bones ash as an alternative source of P for struvite precipitation. *J. Environ. Manag.* 203, 861–866.
- De Vries, S., Postma, R., Scholl, L.V., Blom-Zandstra, G., Verhagen, J., Harms, I., 2017. Economic Feasibility and Climate Benefits of Using Struvite from the Netherlands as a Phosphate (P) Fertilizer in West Africa (No. WPR-673). Paises Bajos, Wageningen.
- Di Iaconi, C., Pagano, M., Ramadori, R., Lopez, A., 2010. Nitrogen recovery from a stabilized municipal landfill leachate. *Bioresour. Technol.* 101, 1732–1736.
- Doyle, J.D., Parsons, S.A., 2002. Struvite formation, control and recovery. *Water Res.* 36, 3925–3940.
- EPA, 2007. Method 3051 (SW-846). *Microwave Assisted Acid Digestion of Sediments, Sludges, Soils and Oils*.
- EPA, 2003. The SW-846 compendium. In: *Methods 7000A Index to EPA Tests Methods*.
- González, R.P., López, E.G. de S., Plaza, C., 2009. Lettuce response to phosphorus fertilization with struvite recovered from municipal wastewater. *Hortscience* 44, 426–430.
- Gu, C., Zhou, Q., Cusick, R.D., Margenot, A.J., 2021. Evaluating agronomic soil phosphorus tests for soils amended with struvite. *Geoderma* 399, 115093.
- Halim, A.A., Aziz, H.A., Johari, M.A.M., Ariffin, K.S., 2010. Comparison study of ammonia and COD adsorption on zeolite, activated carbon and composite materials in landfill leachate treatment. *Desalination* 262, 31–35.

- Hu, L., Yu, J., Luo, H., Wang, H., Xu, P., Zhang, Y., 2020. Simultaneous recovery of ammonium, potassium and magnesium from produced water by struvite precipitation. *Chem. Eng. J.* 382, 123001.
- Huang, H., Xiao, D., Pang, R., Han, C., Ding, L., 2014. Simultaneous removal of nutrients from simulated swine wastewater by adsorption of modified zeolite combined with struvite crystallization. *Chem. Eng. J.* 256, 431–438.
- IACHR (I.-A.C. on H.R.R. 7/2020), 2020. Inhabitants of the areas near the Santiago River regarding Mexico [WWW Document]. Precautory Meas. No. 708-19. URL. <http://www.oas.org/en/iachr/decisions/pdf/2020/7-20MC708-19-ME.pdf> (consultado 5.26.21).
- Ishii, S.K.L., Boyer, T.H., 2015. Life cycle comparison of centralized wastewater treatment and urine source separation with struvite precipitation: focus on urine nutrient management. *Water Res.* 79, 88–103.
- Iskander, S.M., Novak, J.T., He, Z., 2018. Enhancing forward osmosis water recovery from landfill leachate by desalinating brine and recovering ammonia in a microbial desalination cell. *Bioresour. Technol.* 255, 76–82.
- Kabata-Pendias, A., 2011. Trace Elements in Soils and Plants, fourth ed. CRC Press, Boca Raton, Florida.
- Krishnamoorthy, N., Dey, B., Unpaprom, Y., Ramaraj, R., Maniam, G.P., Govindan, N., Jayaraman, S., Arunachalam, T., Paramasivan, B., 2021. Engineering principles and process designs for phosphorus recovery as struvite: a comprehensive review. *J. Environ. Chem. Eng.* 9, 105579.
- Land and Water Division, 2002. Los fertilizantes y su uso, 4a ed.
- López, I.R., Sánchez, P.S., López, A.E., López, A.J.J., Chavarín, E.I.E., Meza, V.K.E., 2015. El cultivo de frijol en Jalisco. Tecnología para altos rendimientos, 2a ed. Universidad de Guadalajara, Zapopan, Jalisco, México.
- Li, B., Boiarkina, I., Yu, W., Huang, H.M., Munir, T., Wang, G.Q., Young, B.R., 2019. Phosphorous recovery through struvite crystallization: challenges for future design. *Sci. Total Environ.* 648, 1244–1256.
- Li, W., Lei, Q., Yen, H., Wollheim, W.M., Zhai, L., Hu, W., Zhang, L., Qiu, W., Luo, J., Wang, H., Ren, T., Liu, H., 2020. The overlooked role of diffuse household livestock production in nitrogen pollution at the watershed scale. *J. Clean. Prod.* 272, 122758.
- Li, X.Z., Zhao, Q.L., 2003. Recovery of ammonium-nitrogen from landfill leachate as a multi-nutrient fertilizer. *Ecol. Eng.* 20, 171–181.
- Liu, Y.H., Rahman, M.M., Kwag, J.H., Kim, J.H., Ra, C.S., 2011. Eco-friendly production of maize using struvite recovered from swine wastewater as a sustainable fertilizer source. *Asian-Australas. J. Anim. Sci.* 24, 1699–1705.
- Luo, H., Zeng, Y., Cheng, Y., He, D., Pan, X., 2020a. Recent advances in municipal landfill leachate: a review focusing on its characteristics, treatment, and toxicity assessment. *Sci. Total Environ.* 703, 135468.
- Luo, Z., Lam, S.K., Hu, S., Chen, D., 2020b. From generation to treatment: a systematic reactive nitrogen flow assessment of solid waste in China. *J. Clean. Prod.* 259, 121127.
- Mikula, K., Izidorczyk, G., Skrzypczak, D., Mironiuk, M., Moustakas, K., Witek-Krowiak, A., Chojnacka, K., 2020. Controlled release micronutrient fertilizers for precision agriculture – a review. *Sci. Total Environ.* 712, 136365.
- Molinos, S.M., Hernández, S.F., Sala, G.R., Garrido, B.M., 2011. Economic feasibility study for phosphorus recovery processes. *Ambio* 40, 408–416.
- Nandre, V., Kumbhar, N., Battu, S., Kale, Y., Bagade, A., Haram, S., Kodam, K., 2021. Siderophore mediated mineralization of struvite: a novel greener route of sustainable phosphate management. *Water Res.* 203, 117511.
- Nesme, T., Metson, G.S., Bennett, E.M., 2018. Global phosphorus flows through agricultural trade. *Global Environ. Change* 50, 133–141.
- Pastor, A.L., 2008. Estudio de la precipitación y recuperación del fósforo presente en las aguas residuales en forma de estruvita ($MgNH_4PO_4 \cdot 6H_2O$). Universidad Politécnica de Valencia.
- PPTAR (P.P. de T. de A.R. de la U.A.), 2014. Procedimiento para detectar y cuantificar bacterias coliformes totales y/o fecales, *Salmonella* y *Shigella* en aguas residuales y residuales tratadas de la PPTAR.
- Puga, A.P., de Mello, R.P., Mattiuz, B., Wylyam, D.V., Fonseca, I.M., 2013. Chemical composition of corn and sorghum grains cultivated in Oxisol with different application methods and doses of zinc. *Cien. Investig. Agrar.* 40, 97–108.
- Rahman, M., Mohd, S.M.A., Rashid, U., Ahsan, A., Mujaffar, H.M., Ix, R.C., 2014. Production of slow release crystal fertilizer from wastewaters through struvite crystallization – a review. *Arab. J. Chem.* 7, 139–155.
- Robles, Á., Aguado, D., Barat, R., Borrás, L., Bouzas, A., Bautista Giménez, J., Martí, N., Ribes, J., Ruano, M.V., Serralta, J., Ferrer, J., Seco, A., 2020. New frontiers from removal to recycling of nitrogen and phosphorus from wastewater in the Circular Economy. *Bioresour. Technol.* 300, 122673.
- Rothwell, S.A., Doody, D.G., Johnston, C., Forber, K.J., Cencic, O., Rechberger, H., Withers, P.J.A., 2020. Phosphorus stocks and flows in an intensive livestock dominated food system. *Resour. Conserv. Recycl.* X 163, 105065.
- SADR (S. de A. y D.R.), 2019. Maíz el cultivo de México [WWW Document]. URL. [https://www.gob.mx/agricultura/articulos/maiz-el-cultivo-de-mexico?idiom=es#:~:text=En el 2019%2C la superficie,millones 228 mil 242 toneladas \(consultado 4.26.21\).](https://www.gob.mx/agricultura/articulos/maiz-el-cultivo-de-mexico?idiom=es#:~:text=En el 2019%2C la superficie,millones 228 mil 242 toneladas (consultado 4.26.21).)
- SAGARPA (S. de A.G.D.R.P. y A., INIFAP, I.N. de I.F.A. y P.), 2015. Agenda Técnica Agrícola de Jalisco, 2a ed. Ciudad de México, México.
- Sánchez, H.E.P., Rodríguez, R.B., Heredero, R.R., García-Peña, N.R.M., 2011. Experiencias para la recuperación del fósforo de las aguas residuales en forma de estruvita en Canal de Isabel II, Cuadernos I+D+i. Biblioteca virtual, Madrid, España.
- Schwarzwalder Sprovieri, J.A., Octavio de Souza, T.S., Contrera, R.C., 2020. Ammonia removal and recovery from municipal landfill leachates by heating. *J. Environ. Manag.* 256, 109947.
- SEMARNAT, 2020. Diagnóstico Básico para la Gestión Integral de los residuos. Medio Ambiente 68–70.
- SEMARNAT (S. del M.A. y R.N.), 2004. NOM-147-SEMARNAT/SSA1-2004, Que establece criterios para determinar las concentraciones de remediación de suelos contaminados por arsénico, bario, berilio, cadmio, cromo hexavalente, mercurio, níquel, plata, plomo, selenio, talio y/o vanadio. D. Of. la Fed.
- SEMARNAT (S. del M.A. y R.N.), 2002. NOM-004-SEMARNAT-2002, Protección ambiental. Lodos y biosólidos. Especificaciones y límites máximos permisibles de contaminantes para su aprovechamiento y disposición final. D. Of. la Fed.
- SEMARNAT (S. del M.A. y R.N.), 2000. NOM-021-RECNAT-2000, Que establece las especificaciones de fertilidad, salinidad y clasificación de suelos. Estudios, muestreo y análisis. D. Of. la Fed.
- Shin, H.S., Lee, S.M., 2010. Removal of nutrients in wastewater by using magnesium salts. *Environ. Technol.* 19 (3), 283–290.
- SS (S. de S.), 2008. NOM-247-SSA1-2008, Productos y servicios. Cereales y sus productos. Cereales, harinas de cereales, sémolas o semolinillas. Alimentos a base de: cereales, semillas comestibles, de harinas, sémolas o semolinillas o sus mezclas. Productos de panificación. Disposit. D. Of. la Fed. 117.
- Stolzenburg, P., Capdevielle, A., Teychené, S., Biscans, B., 2015. Struvite precipitation with MgO as a precursor: application to wastewater treatment. *Chem. Eng. Sci.* 133, 9–15.
- Szymanska, M.S.E., Was, A., Sosulski, T., Van Pruisen, G.W.P., Cornelissen, R.L., 2019. Struvite—an innovative fertilizer from anaerobic digestate produced in a bio-refinery. *Energy* 12, 296.
- TA, T., 2019. Tabla Nutricional: Grano de maíz, blanco [WWW Document].
- Talboys, P.J., Heppell, J., Roose, T., Healey, J.R., Jones, D.L., Withers, P.J.A., 2016. Struvite: a slow-release fertiliser for sustainable phosphorus management? *Plant Soil* 401, 109–123.
- Tansel, B., Lunn, G., Monje, O., 2018. Struvite formation and decomposition characteristics for ammonia and phosphorus recovery: a review of magnesium-ammonia-phosphate interactions. *Chemosphere* 194, 504–514.
- Ueno, Y., Fujii, M., 2001. Three years experience of operating and selling recovered struvite from full-scale plant. *Environ. Technol.* 22, 1373–1381.
- Uhart, A.S., Echeverría, E.H., 1998. El rol del nitrógeno y del fósforo en la producción de maíz. In: Report Number: Boletín Técnico Morgan-Mycogen. Buenos Aires, Argentina.
- Uysal, A., Demir, S., Sayilgan, E., Eraslan, F., Kucukyumuk, Z., 2014. Optimization of struvite fertilizer formation from baker's yeast wastewater: growth and nutrition of maize and tomato plants. *Environ. Sci. Pollut. Res.* 21, 3264–3274.
- Vaneeculta, C., Janda, J., Meers, E., Tack, F., 2015. Efficiency of soil and fertilizer phosphorus use in time: a comparison between recovered struvite, FePO4-sludge, digestate, animal manure, and synthetic fertilizer. In: Rakshit, A., Singh HB, S.A. (Eds.), Nutrient Use Efficiency: from Basics to Advances. Springer, Nueva Delhi, pp. 73–85.
- Wainaina, S., Awasthi, M.K., Sarsaiya, S., Chen, H., Singh, E., Kumar, A., Ravindran, B., Awasthi, S.K., Liu, T., Duan, Y., Kumar, S., Zhang, Z., Taherzadeh, M.J., 2020. Resource recovery and circular economy from organic solid waste using aerobic and anaerobic digestion technologies. *Bioresour. Technol.* 301, 122778.
- Wang, R., Min, J., Kronzucker, H.J., Li, Y., Shi, W., 2019. N and P runoff losses in China's vegetable production systems: loss characteristics, impact, and management practices. *Sci. Total Environ.* 663, 971–979.
- Wang, Y., Meng, G., Shan, M., Wang, D., Bai, Z., Zhou, X., Lv, Y., Bai, J., 2020. Treatment of high-ammonia-nitrogen landfill leachate nanofiltration concentrate using an Fe-loaded Ni-foam-based electro-Fenton cathode. *J. Environ. Chem. Eng.* 8, 104243.
- Warmadewanthi, I.D.A.A., Zulkarnain, M.A., Ikhlas, N., Kurniawan, S.B., Abdullah, S.R.S., 2021. Struvite precipitation as pretreatment method of mature landfill leachate. *Bioresour. Technol. Reports* 15, 100792.
- World Bank Group, 2018. What a Waste 2.0. Washington, Estados Unidos.