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Extended use and optimization of struvite in hydroponic cultivation systems

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A B S T R A C T

Hydroponic systems are an attractive form of urban agriculture due to their low weight load, inert substrate conditions, and overall better control of plant nutrition and growth. However, gaining urban food sovereignty cannot be at the cost of increasing environmental impacts, such as eutrophication and nonrenewable resource depletion, associated with phosphorus fertilizer use. Struvite, a wastewater byproduct, is a potential slow-releasing P source that can serve as a substitute for mineral P fertilizer. In this study, we explored the adequacy struvite in hydroponic systems, testing different quantities (5 g, 10 g and 20 g per plant) compared with monopotassium phosphate for pepper and lettuce hydroponic production. The results show competitive productions for both crops with the use of struvite, especially during the first lettuce harvest (225.5 g, 249.9 g, 272.6 g, and 250 g for 5 g, 10 g, 20 g and control, respectively) where a greater struvite dissolution was seen. Although all struvite treatments in pepper show low phosphorous content in the biomass, yields do not deviate greatly from the control (3.6 kg, 4.3 kg, 7.5 kg and 5.3 kg for 5 g, 10 g, 20 g and control, respectively). The environmental performance of all lettuce treatments showed a reduction in all impact categories, especially freshwater eutrophication and mineral resource scarcity, except for marine eutrophication. All impact categories were reduced for all pepper treatments with 10 g and 20 g of struvite. When the results are extrapolated to a full year of production, we find that the slow dissolution of struvite can sustain competitive production with an initial 20 g, with less impact in all categories except marine eutrophication

1. Introduction

Urban agriculture (UA) has the potential to significantly increase food security in cities (Toboso-Chavero et al., 2019). Increasing green areas in urban landscapes have been gaining popularity, and with new technologies, greening and food production have been taken to building roofs, facades and even indoors (Despommier, 2013; Specht et al., 2014; Appolloni et al., 2021). In particular, soilless agriculture is highly attractive in urban settings because of the reduced weight load on building structures, inert substrate conditions, and overall control of plant nutrition and health (Walters and Stoelzle Midden, 2018; Vinci and Rapa, 2019), as well as because it provides an alternative to contaminated soils. Soilless production can also be a beneficial system to improve water savings since a more controlled environment can be ensured with more accurate irrigation systems as well as water recirculation depending on the installation (Parada et al., 2021). As shown by Appolloni et al. (2021) among 92 cases of urban agriculture identified from 2011 to 2019 a 46% produced with a soilless system. In addition to increasing food sovereignty, UA can promote biodiversity, CO_2 capture and pollination (Baró et al., 2014; Camps-Calvet et al., 2016; Ayers and Rehan, 2021) but can also have negative effects, such as the extensive use of mineral and synthetic fertilizers (Sanjuan-Delmás et al., 2018). Soilless agriculture does not contemplate the addition of nutrients through the substrate but through a nutrient solution given with the irrigation system (EI-Kazzaz, 2017; Sambo et al., 2019). Previous work on life cycle assessment of hydroponic production systems shows that,

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while these fertilizers secure direct nutrient uptake by the plant, their production, extraction, use and disposal are known to have adverse consequences for the surrounding natural ecosystems (Sanjuan-Delmás et al., 2018; (Rufí-Salís and Calvo, 2020; Rufí-Salís and Petit-Boix, 2020). Alternatives for the fertilization in soilless agriculture are gaining interest being aquaponic systems most know for the efficient use of fish debris as nutrient for crop production reducing the potential impact of the production system (Graber and Junge, 2009; Chen et al., 2020). However, aquaponic installations can entail a great initial investment and call for an additional production fish and therefore a greater need for maintenance and skill (Baganz et al., 2020).

The extraction of phosphate rock, the main source of phosphorus (P) for fertilizer use, has become a necessity for modern agriculture and is an indispensable nutrient for plant growth and animal feed (Rahman et al., 2014). However, phosphate rock deposits are limited due to the slow regeneration rate of their cycle compared to carbon or nitrogen, already generating supply shortages due to increasing prices and unequal distribution (Alewell et al., 2020). In recent decades, estimations have been made regarding imminent depletion if extraction continues at the present rate (Cordell et al., 2009), which can be drastically shortened by soil erosion caused by unsustainable production practices (Alewell et al., 2020).

All the P extracted is mostly “lost” from agricultural lands and livestock management through surface and underground runoff (Carpenter and Bennett, 2011; Rahman et al., 2014). This one-way nutrient flow has increased fourfold since preindustrial times (Liu et al., 2008; Villalba et al., 2008; Carpenter and Bennett, 2011; Alewell et al., 2020) and contributes to great ecosystem damage, such as eutrophication, especially in freshwater environments (Cordell and White, 2014).

While this ever-growing thread demands better management of P sources, there is possible recovery from an ongoing loss of nutrients occurring daily in our wastewater treatment plants (WWTPs) (Cordell and White, 2013; Harder et al., 2019). These nutrients contained in wastewater sludge are disposed and managed mostly in complex processes due to the high content of heavy metals, pathogens and other compounds, making it a toxic residue (Panizza and Cerisola, 2001; Rahman et al., 2014). While direct application of sewage sludge to the soil is practiced in several countries, it’s application can entail a bad management of the soil, due to over application for P fertilization as well as the increase of pathogens and heavy metals into the soil, and the potential problematic of social acceptance of this practice due to the emitting odors (Pradel et al., 2020). Countries like Sweden have seen a reduction of the unwanted toxic metals since the 1970 and have started to regard sewage sludge as a potential nutrient provider and soil amendment, still only 20% of the sludge is applied in arable land (Kirchmann et al., 2017).

In recent decades, research has been conducted on the shift from a removal to a recovery approach in urban water cycles in terms of nutrients, not only for their further use in other production sectors but also to prevent their environmental damage in their disposal (Harder et al., 2019; Rufí-Salís and Brunnhofer, 2020).

One of the byproducts in these sewage treatment plants is magnesium ammonium phosphate ($MgNH_4PO_4 \bullet 6H_2O$), a crystal commonly called MAP struvite or just struvite. Struvite is not a new material; its precipitation was first documented in Los Angeles (Borgerding, 1972), but it was approached as a great problem since its precipitation occurs spontaneously at a 1:1:1 molar ratio of magnesium (Mg^{2+}), ammonium (NH_4^{+}) and phosphate (PO_4^{3-}) and under suitable pH conditions (8.5–9.5) (Buchanan et al., 1994; Bouropoulos and Koutsoukos, 2000; Le Corre et al., 2009). The purging of this uncontrolled struvite precipitation can be the cause of additional expenses due to damaged equipment that needs replacement or increased labor costs (Stratful et al., 2004). Since then, technologies aimed at struvite removal through induced precipitation in WWTPs have unveiled a product with great fertilizer potential.

The possibility of P recovery from wastewater in the form of the slow releasing fertilizer struvite has been deemed a solution not only for the supply of this nutrient in agriculture but also to avoid further phosphate rock extraction and an increase of P in wastewater streams and water cycles (Bradford-Hartke et al., 2015).

Struvite has already been tested in a variety of agricultural soils (Latifian et al., 2012; Li et al., 2019), obtaining a wide range of results in crop growth and yield for a diverse range of plants, as shown by Ahmed et al. (2018). Although results vary among different crops, a common perception is the slow solubility of granulated struvite; therefore, its most common application is in the form of pulverized struvite (Degryse et al., 2017). Struvite dissolution has been tested before in continuously stirred pots and at controlled temperature, showing a lower dissolution rate than triple superphosphate (TSP) (Ariyanto et al., 2017; Rech et al., 2018).

Further experiments testing struvite dissolution have demonstrated the importance of medium pH and plant root proximity (Massey et al., 2009; Achat et al., 2014; Talboys et al., 2016; Degryse et al., 2017). This proximity can provide greater access to dissolution mechanisms from the plant that can make the P available, such as the exudation of organic acids to lower the pH of the soil or substrate (Rech et al., 2018). This slow dissolution has been seen to hide crop development in early stages, corresponding to still early growth of the plant root.

Although information on the use of struvite as fertilizer is already available, its use in soilless agriculture is still scarce. Since hydroponic systems enable better control of plant nutrition but are designed to use chemical fertilizers, the use of struvite in exchange for the mineral phosphorous used in soilless agriculture has the potential to reduce its environmental burdens. First approaches have been made to identify the suitability of struvite in hydroponic production as well as it’s combination with biological amendments like rhizobium showing promising results (Arcas-pilz, 2021a). The P emissions to water seam to decrease significantly with the use of struvite compared to mineral derived P fertilizers while other studies reveal even greater productions with struvite (Arcas-pilz, 2021b; Carreras-Sempere et al., 2021). Previous work identifies the low solubility of struvite as a potential burden for plant uptake while it could also ensure reduced P emissions and longer productions over time.

With this knowledge on struvite the question arises if urban agriculture can directly profit from the nutrients generated in their immediate surroundings and strive for expansion without shifting the environmental damage to the generation of greater water and air emissions in urban settings. For this purpose, the production of crops for longer periods was proposed to understand the struvite nutrient discharge in time for short and long cycle crops.

The following experiment was performed to analyze the potential of struvite in providing P in hydroponic production systems by testing struvite solubility and uptake in granular form for two different crops: pepper plants (a highly P-demanding crop with a long growth cycle) and lettuce (shorter cycle). In addition to the dissolution and uptake analysis, this work also focuses on the nutrient discharge into water, covering the potential reduction of P loss into water bodies. To express the environmental burden of the struvite fertilizer and the mineral phosphate fertilizer, the collected information was used to perform an environmental analysis for the produced vegetables using the life cycle analysis assessment (LCA) to determine the environmental footprint during the timeframe of this experiment. The assessment was extrapolated to a one-year production period to simulate these fertilization techniques for a longer time to maximize the P available in the struvite placed in the substrate.

2. Materials and methods

2.1. ICTA-UAB greenhouse

The following experiment was performed in the ICTA-UAB integrated rooftop greenhouse in the Universitat Autònoma in Barcelona from June to September (2020). The production system was hydroponic using individual pots and perlite as the substrate (see picture in supporting information). Nutrients were given through fertigation, mixing concentrated nutrient solutions (NS) with harvested and filtered rainwater (RW) in a proportion of 1:100 (NS:RW) through a drip irrigation system with a 2 L h^{-1} flow. To ensure sufficient irrigation, the amount of drained water was determined daily and maintained ca. ~30% of the incoming irrigation with increasing or decreasing irrigation time. The growing frame consisted of 4 m x 0.5 m wide tables with the capacity to grow two crop lines. Between tables, a distance of 1 m was given from plant to plant.

2.2. *Crop growth and treatments*

In this experiment, the determination of struvite dissolution and uptake was carried out on two different crops, Lettuce (Lactuca sativa L.) and Pepper (Capsicum annum L.). The lettuce and pepper plants were obtained from a nearby nursery in early growth stage with the first growth of the true leaf's (about 7 cm tall for lettuce and about 10 cm tall for pepper), which were then planted in the greenhouse inside perlite filled pots. The perlite was previously wettened with water to ensure a better handling and provide moisture to the plants during the transplanting process. The treatments were arranged in rows to facilitate irrigation and leachate sampling as well as drainage measurement. Each row represents a treatment with a different struvite quantity ranging from 5 g (named 5LE for lettuce, 5P for pepper) and 10 g (named 10LE for lettuce, 10P for pepper) to 20 g (named 20LE for lettuce, 20P for pepper), including a control treatment (named CLE for lettuce, CP for pepper). All crops fertilized with struvite received P-deficient NS, while the control treatment was irrigated with conventional NS (NS specified in supplementary information Table 1). To maximize the contact between struvite and the plant, the granules were placed close to the root once the seedlings were transplanted into the pots.

In the case of the lettuce crop, each treatment consisted of 28 plants arranged in two rows, making two replicates of 14 lettuce plants distributed randomly for each treatment, while for the pepper crop, eight plants were arranged in simple rows (Fig. 1).

During the experiment, several sensors were used to record the climatic conditions inside the greenhouse. Humidity and temperature were recorded with a CS215 Campbell Scientific, and radiation was recorded with a pyranometer (L202 by Hukseflux).

All water flows were measured daily throughout the experiment. The irrigated water was quantified through water meters installed in the irrigation system, while the volume of drained water was measured on buckets at the end of each crop line. Samples of incoming and outgoing water were taken three times a week for each treatment. To ensure good irrigation conditions, the pH and EC for these water samples were measured immediately after collecting daily samples (Supplementary information Figs. 2 and 3).

Table 1
Average yield (g/ per plant) expressed as fresh weight (FW) and dry weight (DW) for all three harvests at 27 DAT, 54 DAT and 81 DAT. Significant differences ($p < 0.05$) between treatments marked with different letter (a,b,c).

Average Yield (g/per plant)	1st Harvest		2nd Harvest		3rd Harvest	
	FW	DW	FW	DW	FW	DW
5LE	225.5^{c} ± 43.2	9.1^{c} ±1.5	224.9^{b} ± 52.1	9.8^{b}±2.2	133.3^{a} ± 28.1	5.5^{a}±1.6
10LE	249.9^{b} ± 35.2	10.1^{b} ±1.4	251.7^{a} ± 56.9	10.9^{a}±2.4	139.8^{a} ± 31.2	5.8^{a}±1.6
20LE	272.6^{a} ± 32.1	10.9^{a} ±1.3	261.4^{a} ± 59.2	11.4^{a}±2.6	149.6^{a} ± 56.4	5.8^{a}±3.0
CLE	250.0^{b} ± 26.6	10.1^{b} ±1.1	279.0^{a} ± 33.5	12.1^{a}±1.5	137.8^{a} ± 35.7	5.4^{a}±2.0

The short cycles of lettuce lasted a total of 27 days after transplanting (DAT). Once the plants were harvested, a new seedling was planted in the same pot (14∅ I 13 cm high for lettuce and 25∅ I 20 cm high for pepper). For each treatment, two pods were removed after each cycle to take substrate samples. Pepper plants were planted parallel to the first lettuce crop until the harvest of the third lettuce cycle (81 DAT), as shown in Fig. 1. Pepper fruit harvests were made weekly once production started, accounting for a total of four harvests before finalizing the experiment. On the other hand, lettuce yields were weighed after each cycle, generating three harvests.

To obtain a more accurate understanding of the possible yield variations among lettuce samplings, 15 pots of each lettuce treatment were labeled with a reference letter (from A to O) maintained throughout the experiment. Relative yields obtained could then be traced back to the corresponding pot, therefore allowing precise appreciation of possible production changes.

The yield produced by the pepper plants was obtained in four harvests. The total fruit weight recorded for each treatment was obtained as the sum of all four harvests. The number of fruits produced in each harvest was also accounted for to estimate the weight per fruit.

2.3. *Plant sampling methods*

For each lettuce cycle, the fresh and dry weights of each plant were measured. After harvest, a random sample of four plants for each treatment was dried at 60 °C until a constant weight was achieved (ca. 7 days).

At the end of the experiment, all pepper plants were harvested and weighed. The pepper plants were separated into leaves, stems and roots, removing all flowers. Additionally, we quantified the leaf weight, number, area index (LAI), stem weight and length. The latter was measured accounting for the main central stem without considering ramifications. However, these ramifications were considered when weighing the stem. The LAI was obtained using the scanned images of 25% of the leaf fresh weight and further processed with a Python script (as indicated in the Supplementary material) (relating the number of pixels per leaf area) to give the total area of each pepper plant (Garrido et al., 2020; Ribalta-Pizarro et al., 2021). A sample of five plants per treatment was also dried in an oven at 60 °C until reaching a constant dry weight (ca. 7 days). In the case of the fruit, a sample of pepper pods was taken from each treatment after every fruit harvesting. Fresh and dry weights were measured following the same procedure as for the plant biomass.

Before drying all plant biomass was rinsed with Elix water and dried to avoid any potential external contamination.

Once the dry weights for lettuce, pepper fruit and biomass were quantified, the samples were ground for further analysis, consisting of digestion with concentrated HNO_3 in a single reaction chamber microwave to be then analyzed for total P concentration using optical spectrometry (ICP-OES).

Substrate samples were transferred to a polypropylene sampling pot after thoroughly mixing the perlite from the pot in a clean container. For each treatment, two samples were taken at the end of each of the three production cycles. After taking the perlite samples, they were placed on aluminum trays and dried at 60 °C for 72 h. Once dry, the samples were weighed and ground for total P determination using the method detailed before.

Irrigation and leached water samples were proportionally unified into weekly samples considering a volume ratio. These samples were filtered through a 0.22 mm filter and analyzed with ionic chromatography (ICS-200) for nitrite and nitrate contents. The Mg and P contents in the water samples were analyzed with ICP-OES (Perkin-Elmer Optima 4300DV).

To calculate the struvite dissolution rate, the amount of P found in the plant and leachate was calculated for each treatment (for lettuce, each cycle was taken separately). The obtained quantity was assumed to

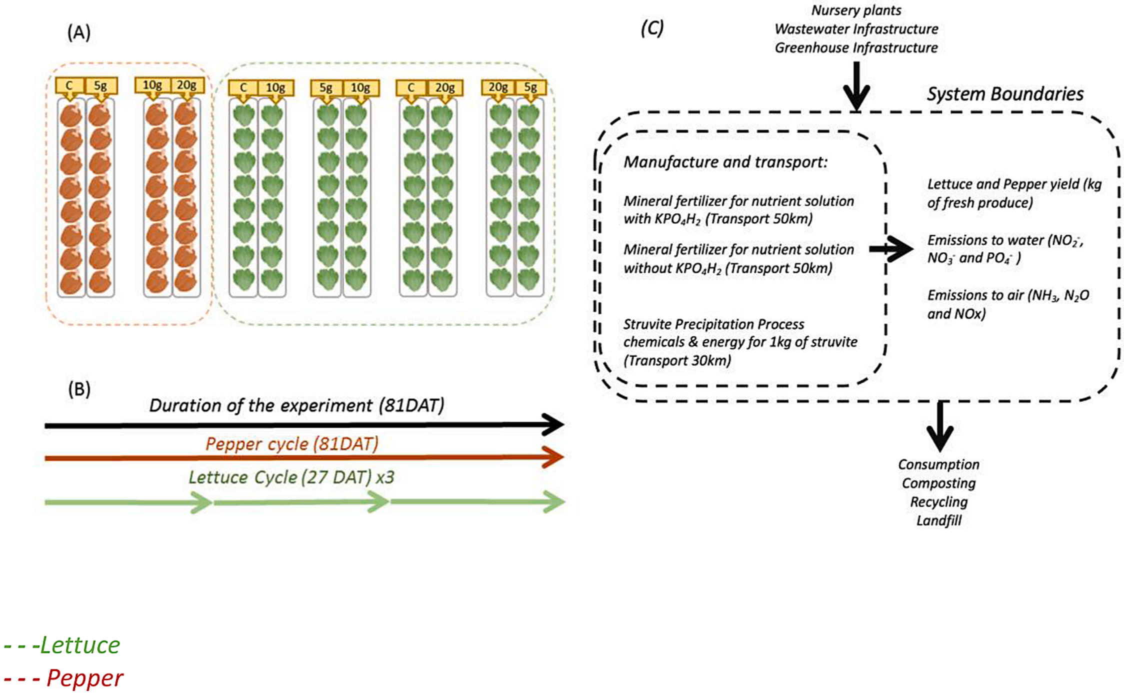


Fig. 1. Experimental outline (A) shows the distribution of the pepper and lettuce treatments along the laboratory greenhouse (C= control treatment). The experimental timeline (B) shows the total duration of the experiment and the duration of the pepper and lettuce cycles (DAT= days after transplanting). The System boundaries for the environmental analysis (C) show the scope of the analysis within the dotted line.

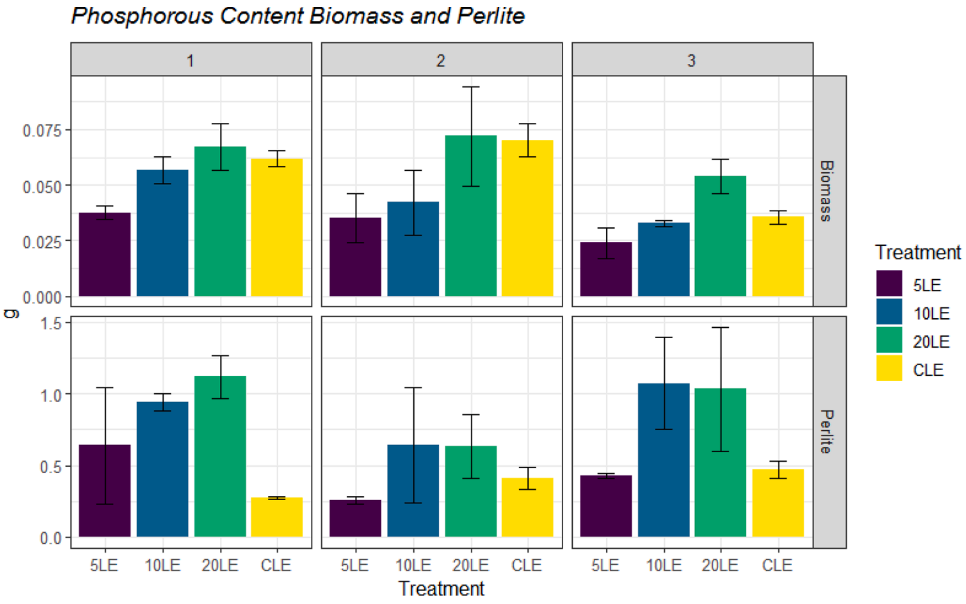


Fig. 2. Amount of P (g) accumulated in the lettuce biomass(above) and perlite (below) for treatments 5LE, 10LE, 20LE and CLE in all three harvests (1, 2, 3) given in total g per plant (in the case of perlite g per pot).

be the dissolved P from the struvite and divided by the liters irrigated to the crop. The dissolution rate was then plotted against the initial struvite amount given to the plant. For the second and third lettuce cycles, the initial struvite was assumed to be the remaining struvite in the perlite after the previous crop cycle.

2.4. Environmental assessment

The Life Cycle Assessment (LCA) used to determine the environmental impact of the irrigation system followed the ISO norms 14,040 and 14,044 (ISO 2006).

2.4.1. Goal and scope and inventory

An environmental assessment of the fertilization method was made, comparing the environmental load to produce 1 kg of fresh lettuce and pepper pods considering the incoming fertilizers and outgoing emissions to water and air, as shown in Fig. 1(C). The life cycle assessment (LCA) tool was used to determine the environmental impact of fertilization for all treatments, which was calculated with Simapro 9.1 software, using the Ecoinvent 3.7 database to account for the background

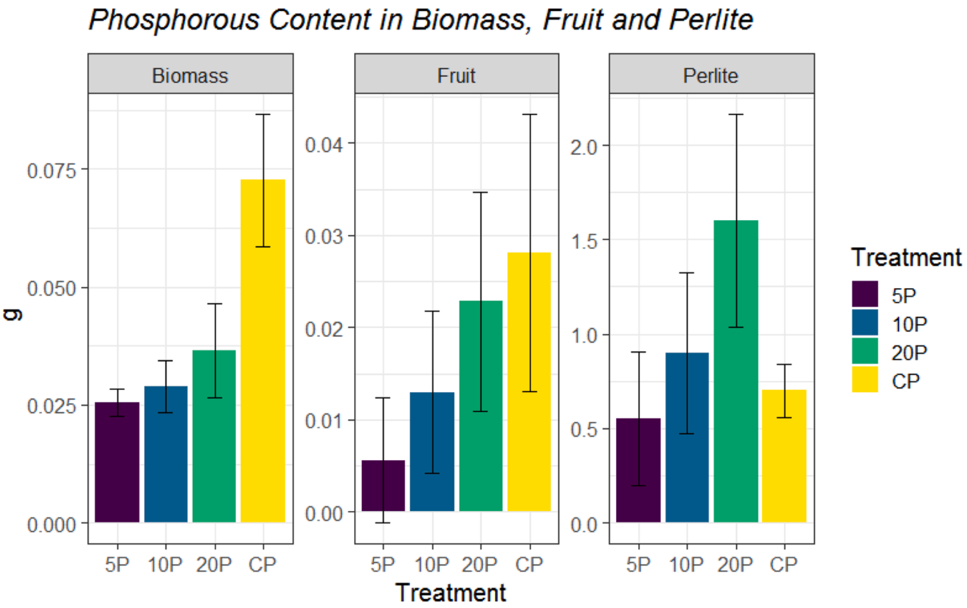


Fig. 3. P amount (g) in pepper biomass, fruit, and perlite for treatments 5P, 10P, 20P and CP at 81 DAT given in total g per plant.

environmental information and the ReCiPe midpoint impact assessment method (Huijbregts et al., 2017). The scope of this attributional LCA was defined as cradle to grave since the production, transport, use and disposal stages for the different fertilizers were considered. On the other hand, the greenhouse infrastructure and production system were not included in the system boundaries, focusing only on the impact of the use of struvite as fertilizer.

2.4.2. *Life cycle incentory*

The inventory for the LCA was comprised by the obtained data from the experiment. The fertilizer applied was obtained from the nutrient solution and irrigation amount controlled daily through the water meters installed in the irrigation system. The incoming irrigation as well as the leachate water was analyzed to obtain the N and P emitted to water (in the form of NO_2^-, NO_3^- and PO_4^-). From the incoming irrigation the calculation of the N emissions to air in the form of ammonia (NH_3), nitrous oxide (N_2O) and nitrogen oxides (NOx) following the emission factors established by Montero et al. (2011).

The generation of struvite was accounted for in the environmental assessment based on the production of the commercial house Ostara®. For the production of 1 kg of struvite, the additional chemical input in the precipitation stage was 0.4239 kg MgCl, 0.766 kg NaOH for pH stabilization and 0.523 kWh energy applied for mixing and aeration (Amann et al., 2018). Impacts related to wastewater treatment, such as an improvement of the effluent or the additional technology required for P removal to the sludge line, were excluded from the system boundaries of this LCA. The transport for all fertilizers accounted for 50 km from the greenhouse. The struvite transport, on the other hand, was estimated to be 30 km, which corresponds to the approximate distance to the two nearest WWTPs of the city (EDAR Besós and EDAR Llobregat), although they currently are not producing or selling struvite.

The environmental assessment was made for a single plant pot, taking into account its fertilization and production. These results can then be further extrapolated to greater production. The detailed inventory and processes can be seen in the supplementary information (Table 5)

2.4.3. *Environmental impact assesment*

The impact categories selected were global warming (GW), terrestrial acidification (TA), freshwater eutrophication (FE), marine eutrophication (ME) and mineral resource scarcity (MRS). These selections were based on the author's expertise and previous literature focusing on the impacts of fertilizers in soilless systems and the use of struvite (Brentrup et al., 2004; Sanjuan-Delmás et al., 2018; Rufí-Salís and Brunnhofer, 2020).

Global warming, expressed in kg CO_2 eq. was chosen due to the documented relevance of greenhouse gas emissions from the production of fertilizers, as well as their transport, and due to the direct emissions occurring at the plant level (Hasler et al., 2015; Chatzisymeon et al., 2017). This case is especially true for nitrous oxide (N_2O). Thus, we considered the additional nitrogen given in the form of ammonia through struvite. The proportional fraction of ammonia released in each treatment, determined through direct measurement in the leachates, was considered when calculating the emission factor as well as the nitrogen given in the irrigation. For the same reason, TA (kg SO_2 eq.) was also chosen to reflect the direct emissions due to the application of ammonia as well as other acidifying agents generated during transportation and manufacturing of fertilizers (Hasler et al., 2015). FE (kg N eq.) and ME (kg P eq.) have been regarded as the most relevant impact categories when analyzing fertilization methodologies, especially considering nitrogen and phosphorous (Hasler et al., 2015; Chatzisymeon et al., 2017; Vatsanidou et al., 2020). These impact categories are especially relevant in this study since slow struvite dissolution can provide insight into the possible reduction of P leaching into fresh and marine waterbodies, and again, the addition of N through struvite can also be reflected in the leachate quantities. FRS (kg oil eq.) was added as a relevant impact category to reflect fossil energy-related emissions that could arise due to struvite precipitation and transport compared to mineral P. Finally, MRS (kg Cu eq.) was chosen to reflect the extraction of finite mineral resources, especially focused on phosphate rock extraction versus the recycling and reuse of phosphorous in the form of struvite.

2.5. *Statistical analysis*

Shapiro–Wilk test $p > 0.05$ was used to test the normality of the data, while homogeneity of variance was tested with Levene's test $p > 0.05$. Duncan's multiple range test was used to assess the statistical significance of treatments when parametric criteria were validated. For nonparametric data, the Kruskal–Wallis test was used. The significance between the treatments is marked with different letters (a, b and c).

3. Results

3.1. *Crop growth and yield production*

The resulting productions for the three lettuce cycles can be observed in Table 1. Here, we can appreciate the average yields obtained for all three harvests and all treatments for their fresh and dry weights. Further information on the specific production within the marked pots (A to O) can be seen in Table 2 in the supplementary information.

We identified a general decrease in yield during the third harvest, most likely due to a remarkable decrease in the overall temperature during 54 DAT and 81 DAT in contrast to the previous crop cycles (1 DAT to 54 DAT). This variation in the climatic conditions can be observed in Fig. 1 in the supplementary information with the recordings of humidity, radiation and temperature during all three cycles. While temperatures still ranged between 20 °C and 25 °C, the sudden change in comparison to the previous two crop cycles may have caused a delay in lettuce growth.

While no great differences can be seen in the overall yield of our lettuce cycles, the close monitoring of our pots can give us the variability of the obtained yield for the lettuces grown with the same initial struvite. This finding means that from the same pot, we can monitor the obtained yield in all three cycles. Table 2 in the supplementary information provides us with such information showing a general decline in production, with the most acute decrease in yield in the 5LE treatment with a −11% difference between the first harvest and the second. On the other hand, the decline for treatments 10LE and 20LE was less pronounced, with −2% for both.

In the case of pepper plant growth and production, Tables 2 and 3 provide insight into the differences spotted between treatments. Table 2 provides the main measurements made of the pepper plants at the end of the experiment (81 DAT).

While no significant differences were seen for the stem weight, an increase in the fresh and dry weight was observed with the increasing amount of struvite applied. The same increase was noted for the leaf weight, number, and LAI, showing significant differences in all but the latter. The control treatment generally showed greater values in all measurements apart from one, the plant stem length.

The yield produced by the pepper plants (Table 3) showed a greater total weight for the 20P treatment. While the total number of fruits was also higher for the 20P treatment, the weight per fruit did not differ greatly from that of the other treatments.

3.2. *Nutrient content in plant biomass and substrate*

The results shown in Fig. 2 depict the P content in the lettuce crop after 27 days of growing in the greenhouse for all treatments. The amount of P found in the lettuce biomass is directly related to the amount of struvite given, being lowest for the 5LE treatment followed by the 10 L and finally 20LE treatments. The amounts of P found in the 5LE and 10LE treatments decrease noticeably over time in the second and third cycles, while the 20LE treatment does not experience a great reduction during the second cycle but rather on the third cycle. It is important to point out that the results found for the second crop cycle show a much greater variability than the first and third ones.

The remaining struvite content in the perlite and therefore P remaining in the substrate were analyzed and plotted in Fig. 2. Here, we can appreciate a great difference between the struvite fertilization treatments and the control, since the nutrient content in the perlite slowly increases over time for the latter, while the P content in the struvite treatments fluctuates and slowly decreases due to its dissolution. Here, again, a much greater variability in the results was observed for the second cycle.

Table 3

Total yield obtained in four pepper fruit harvests (55 DAT, 62 DAT, 72 DAT and 81 DAT) for each treatment. The yield given in g while an average fruit weight given with g/ fruit.

Harvest	Treatment	Total Fruit Weight (g)	Fruit number (nr)	Weight/ number (g/ fruit)
1ST HARVEST (55 DAT)	5P	1261.9	22	57.4
	10P	1402.7	21	66.8
	20P	2155.5	25	86.2
	CP	1479.0	22	67.2
2ND HARVEST (62 DAT)	5P	911.0	18	50.6
	10P	1240.0	19	65.3
	20P	1833.4	28	65.5
	CP	1597.0	21	76.0
3RD HARVEST (72 DAT)	5P	528.0	9	58.7
	10P	759.0	10	75.9
	20P	1649.2	18	91.6
	CP	1046.0	13	80.5
4TH HARVEST (81 DAT)	5P	860.0	20	43.0
	10P	940.0	21	44.8
	20P	1860.0	29	64.1
	CP	1225.0	19	64.5
TOTAL	5P	3560.9	69	51.6
	10P	4341.7	71	61.2
	20P	7498.1	100	75.0
	CP	5347	75	71.3

Fig. 3 depicts the P content in pepper biomass, fruit and perlite, showing great variation between struvite fertilization treatments and the control. While our treatments showed a low P content of 1.2 mg/g in leaves and 0.7–0.8 mg/g in the plant stem, giving ranges of 0.02 to 0.03 g of P in the total dry biomass, the control treatment showed values within adequate ranges of 2.1 mg/g (0.08 g of P in the total dry biomass). The amount of P in the harvested fruits reveals the differences between treatments based on the great mobility within the plant. Fruits are an ultimate sink of the phosphorous content in plants, and this result is reflected with a very clear relation to the struvite treatment. The great variability seen in these results derives from the great difference found between harvests within the same treatment, while the first pepper fruit harvest contained greater P concentrations, the third suffered a great reduction for all treatments, even the control (Supplementary information Figure 7). Finally, the amount of P found in perlite responds to the initially given struvite.

3.3. *Phosphorous content in the leachate*

The resulting phosphorous concentrations found in the leachates were calculated for the total outgoing water weekly per plant,

Table 2

Mean values of pepper plant biomass measurements made in the 81 DAT. Stem weight and Leaf weight given in g for their fresh weight (FW) and dry weight (DW). *The LAI calculated in cm2. Significant differences ($p < 0.05$) between treatments marked with different letter (a,b,c).

TREATMENT	Stem weight (g/plant)		Leaf weight (g/plant)		Stem length (cm)	Leaf number (nr)	LAI*
	FW	DW	FW	DW			
5P	169.3^{a}	32.2^{a}	131.9^{a}	21.2^{a}	99.1^{ab}	110.1^{a}	2.5 ^{a}
10P	184.0^{a}	35.2^{a}	166.7^{ab}	26.0^{ab}	107.1^{b}	127.3^{ab}	3.3 ^{a}
20P	204.1^{a}	40.9^{a}	195.2^{bc}	31.4^{b}	96.0^{ab}	138.9^{ab}	3.8 ^{a}
CP	220.1^{a}	43.9^{a}	236.2^{c}	35.5^{b}	90.0^{a}	156.3^{b}	4.1 ^{a}

generating the patterns found in Fig. 4. The accumulation of P in the leachates for the lettuce and pepper crops can be seen in supplementary information Fig. 6.

The results for the lettuce crop show the discharge of phosphorous during all three cycles, recognizing a clear pattern before and after each harvest. This pattern was highly noticeable for the CLE treatment, where the phosphorous content in the leachates decreased with the growth of the plant and rose once the plant was harvested and replaced with a seedling. This same pattern can be observed for all struvite fertilization treatments for lettuce, finding greater amounts for 20LE and less for 10LE and 5LE.

The phosphorous content in water, on the other hand, differs greatly when observing the CP and the 5P, 10P and 20P treatments. The biomass growth, climatic conditions and subsequent irrigation amount define the loss of phosphorous in the CP treatments, showing an overall decrease in the concentration with a peak at approximately Day 37 after transplanting. All treatments with struvite showed very low concentrations in the leachates, especially after 20 DAT.

3.4. Phosphorous balance

The results obtained in the previous sections enable us to generate the nutrient balance for P during these cycles for all treatments. This understanding helps us estimate the P flows into the plant, substrate and water. These nutrient balances were calculated for the P flows in the lettuce and pepper crops (Table 4) and averaged to obtain data for one plant. In addition, the water balances per plant are given in Figures 9 and 10 in the supplementary information. The nutrient balance is subjected to potential inaccuracies given through the sampling of substrate, water and biomass and the generation of mean values for all samples generating approximate values close to 100%.

The balance for lettuce gives an overall picture of the obtained results of the phosphorous flows into the plant biomass as well as leachates. Compared to the control treatment, the phosphorous flow into the outgoing water was approximately 10 to 14 times lower for the 10LE and 5LE treatments, respectively, while the flow into the plant biomass remained similar. The remaining phosphorous in perlite remained high in the 5LE and 10LE treatments, while more than half was reduced in the 20LE treatment. We also appreciate an accumulation of P in the perlite of the CLE treatment.

For pepper, the biomass flows were divided between the fruits produced and the generated biomass on the day the plants were cut and weighed. Here, we can appreciate the great quantity of phosphorous found in the pepper fruits, which equals the total phosphorous found in the plant leaves and stem. The total biomass showed a great difference between the CP treatment and the struvite fertilization treatments, revealing a much greater P content in the control. Due to the greater irrigation needs of pepper plants compared to lettuce plants, the CP treatment received an overall greater amount of P through irrigation compared to the CLE treatments. Therefore, although the P in the perlite and leachates is lower in CP than in the CLE treatments in terms of percentage, the absolute amounts are greater. In the case of the pepper plants fertilized with struvite, the P in the leached water was similar and even smaller than the amount found in the lettuce crop. The outgoing P in the leachates of the pepper plants was 10, 19 and even 35 times lower than that in the control treatment (CP) for the 20P, 10P and 5P treatments, respectively.

The calculated dissolution rates for the applied struvite in lettuce and pepper are shown in Fig. 5. The struvite dissolution was estimated by the P contained in the water leachates as well as P in the plant biomass. This dissolution has a direct impact on the P uptake by the plant that was estimated as the P contained in the P biomass. The results for lettuce show greater dissolution with a greater initial amount of struvite. The dissolution of the struvite was also higher during the first lettuce cycle (marked with number 1 in the figure), showing smaller differences between the second and third cycles (marked with 2 and 3, respectively). The dissolution rate found in the pepper crop was smaller than that in the lettuce crop but followed the same pattern as seen before, with greater dissolution with higher amounts of struvite.

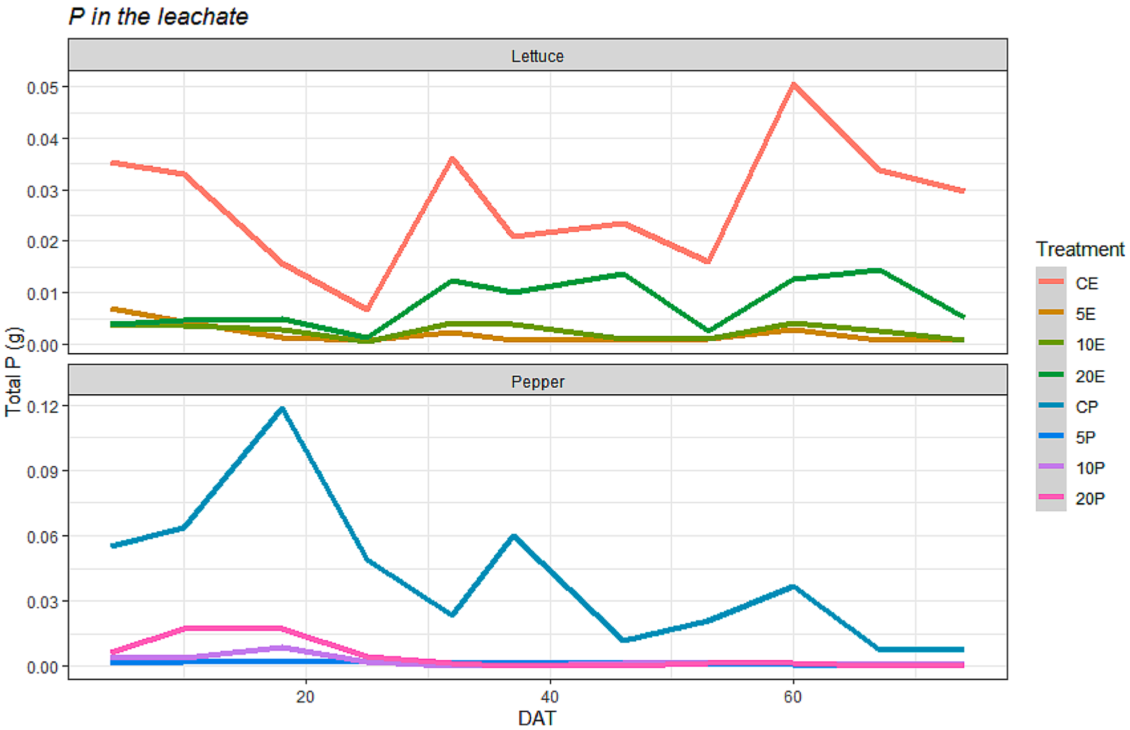


Fig. 4. Total phosphorous (Total g) found in the leachate water from 4 DAT to 74 DAT in Lettuce and Pepper crops for treatments with 5 g, 10 g and 20 g of struvite as well as Control treatments (CP and CLE) irrigated with KPO4H2.

Table 4

Phosphorous balance per plant for the lettuce and pepper crop for treatments 5LE, 10LE, 20LE, CLE, 5P, 10P, 20P and CP. Initial P given through struvite and NS. Biomass 1, 2 and 3 corresponding to the 3 Lettuce cycles at 27 DAT, 54 DAT and 81 DAT. Pepper total biomass corresponds to total P found in Fruit Production and Biomass.* For the biomass the amount of P from the root was also included with the root DW and root phosphorous content obtained from literature (Xu et al., 2004; Pereira-Dias et al., 2018; Erel et al., 2019).

TREATMENT	INITIAL P	BIOMASS 1*		BIOMASS 2*		BIOMASS 3*		TOTAL BIOMASS		PERLITE		LEACHATES		BALANCE
LETTUCE	Struvite /NS g	g	%	g	%	g	%	g	%	g	%	g	%	%
5LE	0.625	0.047	8	0.047	8	0.035	6	0.130	21	0.443	71	0.022	4	95
10LE	1.25	0.064	5	0.050	4	0.042	3	0.157	13	0.885	71	0.028	2	86
20LE	2.5	0.077	3	0.079	3	0.064	3	0.221	9	1.196	48	0.085	3	60
CLE	1.049	0.071	7	0.081	8	0.044	4	0.196	19	0.384	37	0.318	30	86
	INITIAL P	PRODUCTION		BIOMASS*		TOTAL BIOMASS		PERLITE		LEACHATES		BALANCE		
PEPPER	Struvite /NS g	g	%	g	%	g	%	g	%	g	%	%		
5P	0.625	0.029	5	0.069	8	0.099	12	0.561	90	0.014	2	104		
10P	1.25	0.055	4	0.076	4	0.130	9	0.904	72	0.026	2	83		
20P	2.5	0.086	3	0.092	3	0.178	6	1.602	64	0.051	2	72		
CP	2.595	0.106	4	0.189	7	0.295	11	0.709	27	0.501	19	57		

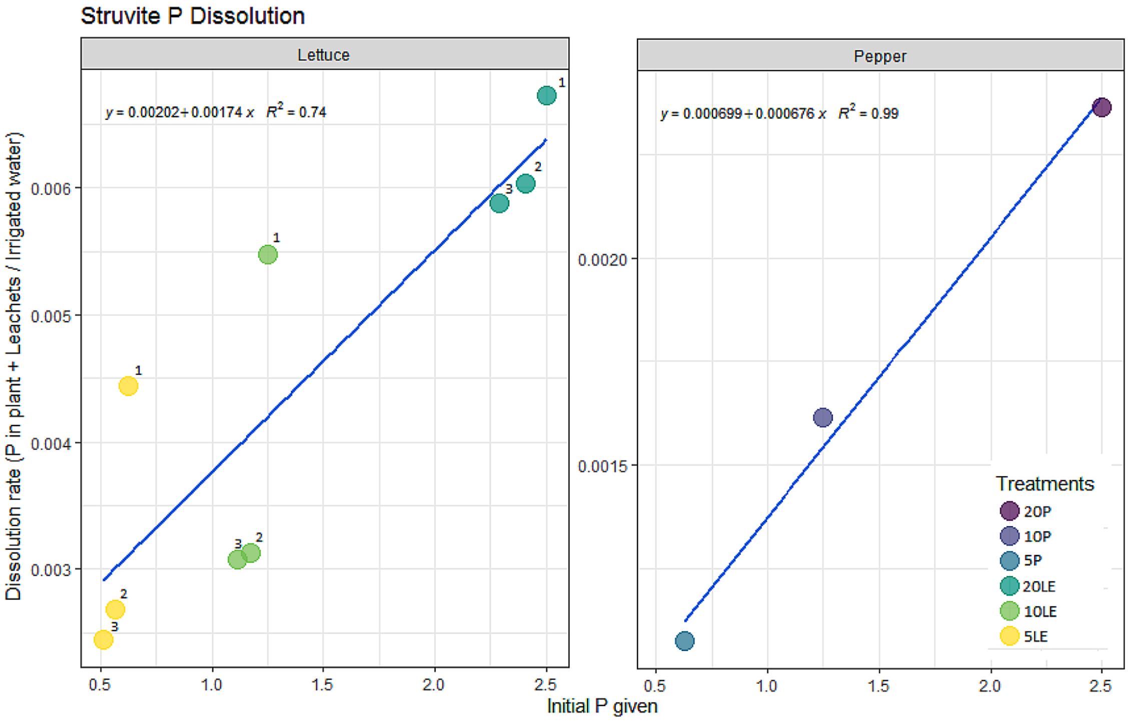


Fig. 5. Correlation of the dissolution rate of the struvite P and initial P given in Lettuce and Pepper for treatments 5LE, 10LE, 20LE, 5P, 10P and 20P For lettuce all three cycles are taken in account, marked as 1, 2 and 3 respectively.

3.5. Life cycle assessment

Fig. 6 shows the results for lettuce, and Fig. 7 shows the results for the environmental assessment of the fertilization treatments. Since only the fertilization of the crops was considered for the analysis (Fig. 1), all differences will be related to the use of struvite instead of monopotassium phosphate (MKP) in the form of KPO_4H_2, leaving out the laboratory infrastructure and auxiliary equipment, as well as the end-of-life processes.

The obtained results for six out of seven impact categories show that fertilization with struvite has lower impacts than the control, and for the cases of ET, MRS, FRS and GW, impacts are also reduced as we increase the amount of struvite applied. In terms of eutrophication, FE, which is directly related to the emissions to water, had the greatest impact on the control irrigated with mineral P, followed by 20LE, which was the treatment with the highest quantity of struvite per plant.

ME, although related to the emissions to water, also does not decrease substantially for the struvite-treated crops due to its relation to nitrogen emissions, which are sustained for all treatments. Furthermore, we can observe that although a reduction of the impacts is occurring for the 20LE treatment, this reduction is most likely not a consequence of a reduced N emission to water but due to greater yields obtained; on the other hand, treatment 5LE is overshadowed by the lower yields generated and a proportionally greater N emission due to the smaller plant growth.

The results obtained for the pepper crop indicate a considerably abrupt decrease in the emissions in all impact categories for treatments 10P and 20P in comparison to the CP treatment. In comparison to the lettuce crop, the ME was severely reduced for these two treatments. The 5P treatment with lower production rates and therefore lower FU experiences much greater values for all impact categories except FE and MRS, which are slightly below the control treatment CP in the latter.

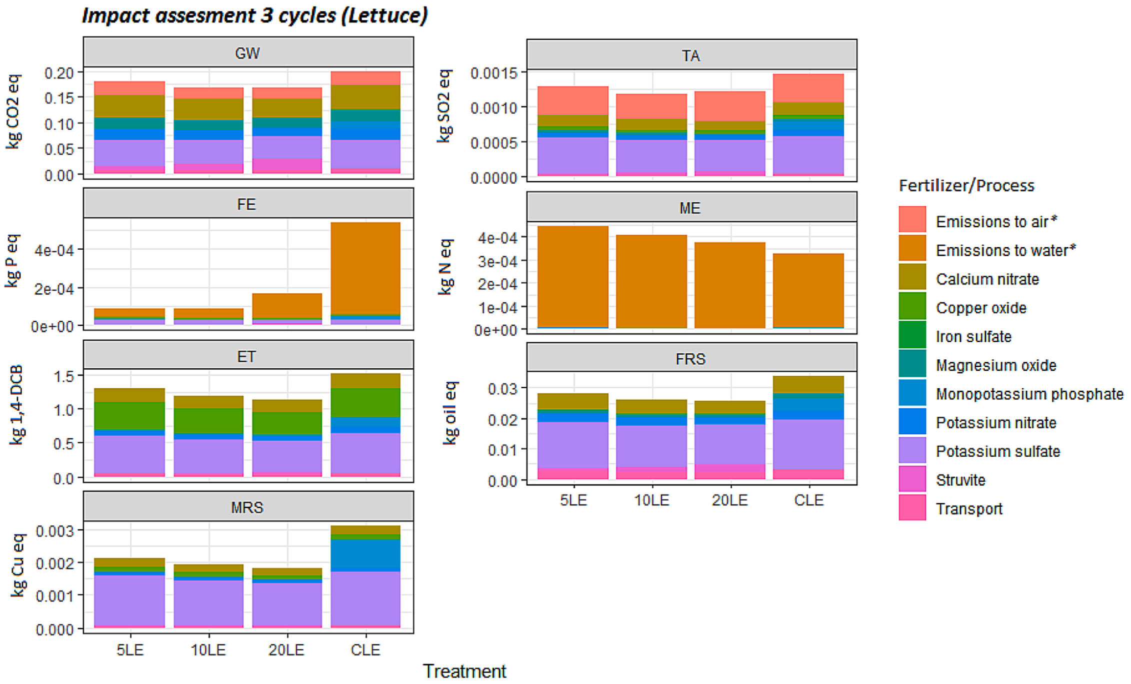
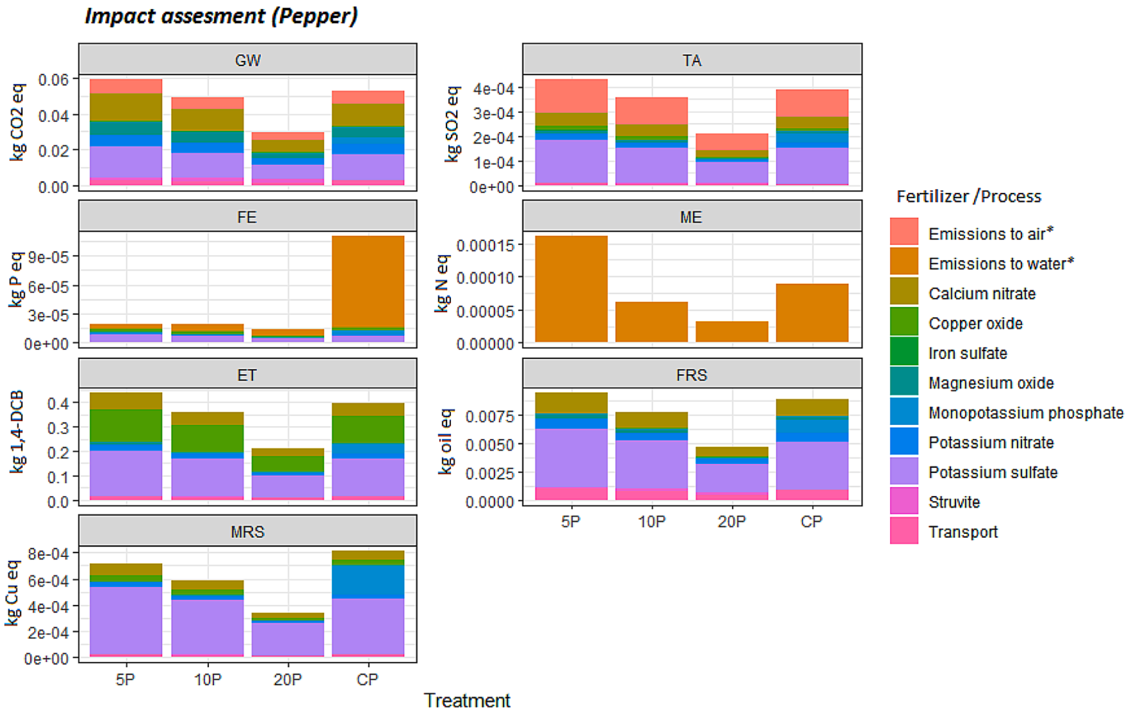


Fig. 6. Impact assossiated to the system fertilization for 3 consecutive lettuce productions for 81 days. The obtained emissions have been calculated in relation to the resulting yield as FU. GW (global warming), TA (Terrestrial acidification), FE (Freshwater eutrophication), ME (Marinewater eutrophication), ET (Ecotoxicity), FRS (Fossile resource scarcity), MRS (Mineral resource sarcity). *Emissions to air were based on the emission factors of Ammonia, N2O and NOx obtained for the applied nitrogen; the emissions to water were directly obtained from the nitrogen and phosphorous detecte in the water leachate.



Impact assosciated to the system fertilization for pepper production during 81 days. The obtained emissions have been calculated in relation to the resulting yield as functional unit. GW (global warming), TA (Terrestrial acidification), FE (Freshwater eutrophication), ME (Marinewater eutrophication), ET (Ecotoxicity), FRS (Fossile resource scarcity), MRS (Mineral resource sarcity). *Emissions to air were based on the emission factors of Ammonia, N2O and NOx obtained for the applied nitrogen; the emissions to water were directly obtained from the nitrogen and phosphorous detected in the water leachate.

Overall emissions for pepper production were lower than those found for the three lettuce cycles combined. This finding can be explained by the greater weight obtained with pepper production, making a direct comparison between crops difficult with a functional unit only accounting for the obtained yield.

4. Discussion

4.1. Is struvite a good fertilizer for hydroponic production?

The results show that the long cycle of pepper and short cycles of lettuce fertilized with struvite did not differ greatly from each other in the uptake and use of P. We identified that the amounts accumulated in the plant biomass between treatments with the same struvite quantity (5LE and 5P, 10LE and 10P, 20LE and 20P) did not change substantially. This information reveals that little to no effect on struvite uptake can be attributed to the crop cycle duration or needs. This second idea is reinforced by the level of P found in the pepper biomass, corresponding to low concentrations and mirrored in the fruit P content (Hochmuth et al., 2018). Although a clear P deficiency is shown in the plant biomass nutrient content, no such deficiency can be traced in the plant physiology or production capacity (Zelia et al., 2017). Pepper fruit production increases with the given struvite, as well as leaf production and growth, showing significant differences that indicate the relevance of the given struvite amount to the plant. Related to the findings of Talboys et al. 2016 in 90-day experiments with struvite-fertilized crops, the amount of P taken by the plant is lower in the case of struvite but does not affect the final yield, being very similar to the more soluble triple superphosphate (TSP). This finding has been attributed to the struvite residual value in the substrate in comparison to TSP, enabling P uptake by the plant during a sustained timespan.

The leachate P for lettuce and pepper plants was also shown to be a great indicator of the slow solubility of the fertilizer and increased with greater water flow when lettuce was harvested. The higher water demands of the pepper plants could therefore have been a defining factor contributing to low struvite dissolution, as seen in Fig. 5. Although the plants had sufficient irrigation indicated by the daily water drainage, the leaching of phosphorous into the drained water only increased during the early stages of plant growth until 20 DAT. Once temperatures start to rise and drainage is reduced, the emissions of P into the drainage are also reduced. Although greater temperatures have been seen to increase struvite solubility (Rahaman et al., 2006; Ariyanto et al., 2017), its use as a fertilizer unveils that irrigation plays a major role in plant phosphorus uptake (Lunt et al., 1964; Turner, 1985; Silber et al., 2005).

The greater variability obtained in the second lettuce cycle can also be attributed to the increasing temperatures enabling a greater dissolution of struvite in the perlite substrate as well as the slight reduction of the pH from the nutrient solution increasing the struvite solubility (Ariyanto et al., 2017). The uptake and use of the plant could have been affected by the delay in the irrigation adjustment to meet the plant needs.

The capacity of struvite dissolution, which has been attributed to different factors in previous literature, like the plant rhizosphere exudation (Kamilova, 2006; Khademi, 2010; Dakora and Phillips, 2002; Talboys et al., 2016), plant growth stage (Degryse et al., 2017) and plant needs. These factors for greater struvite dissolution have not been reflected in these results, indicating a reduced uptake from the pepper plant compared to the lettuce crop. The idea of plant rhizosphere exudation being important for struvite dissolution was also questioned by Rech et al., 2018, who demonstrated the inefficiency of low-concentration root exudates to solubilize granular struvite.

Overall, the quantity of P in the plant biomass (9–21% and 6–12% of the applied P for lettuce and pepper, respectively) as well as the P leachate (2–4% and 2% of the applied P for lettuce and pepper, respectively) in both crops indicate that the amount of dissolved P is very small. This information is reinforced by the analysis of the perlite substrate, indicating that a large amount of struvite remains undissolved in the substrate. This effect was also seen in previous literature with other crops, such as soybean and wheat (Rech et al., 2018) and common bean (Arcas-pilz, 2021a). These low dissolution percentages coincide with dissolutions in media with pH values ranging from 7.5 to 8 (Talboys et al., 2016; Rech et al., 2018), which were mainly found in the present study. While the pepper plants did not reach adequate ranges of P in the biomass with struvite fertilization, the lettuce crops did not differ greatly from the control treatment, especially for 10LE and 20LE. This information reinforces the idea of further reusing the given struvite for consecutive cycles within the same substrate with short cycle crops, such as lettuce.

On the other hand, the dissolution rate seems to be greater during the first plant cycle in all lettuce treatments. The struvite crystal composition and available P could be more prone to dissolve earlier, progressively reducing the dissolution rate with consecutive plant cycles. This same dissolution trend was seen by Rech et al. (2018) when observing the P concentration in the soil solution of wheat and soybean crops with the fertilization of three different struvite types. Concentrations of P were recorded for 40 days, showing a decrease and stability by the end of the experiment. The close dissolution rate of the second and third cycles could indicate this point of stability.

4.2. Does the use of struvite reduce the environmental burden of hydroponic production?

The environmental analysis showed that the 5LE and 5P treatments had the highest impacts since they had the lowest yields. On the other hand, the greater use of struvite can also generate a greater discharge of P into the water system compared to treatments with less applied struvite. This finding is reflected in the case of the lettuce crops for the ME and FE impact categories. While greater yields were achieved for the 20LE treatment, greater P and N leachates were generated, increasing the environmental footprint in comparison to the other struvite treatments. Smaller crop growth in the case of 5LE and 5P can also increase the amount of leachates and discharge of N to the environment. This finding has been observed both for lettuce and pepper, where smaller crop growth leads to greater water and nutrient discharge. However, the P discharge in the struvite treatments was always lower than that in the control and thus impacted freshwater eutrophication.

The impact of the struvite production compared to the monopotassium phosphate seem significantly smaller, being most noticeable in GW and FRS. The production of monopotassium phosphate on the other hand has a large impact on the MRS as predicted, due to the extraction of the finite phosphate rock. The impact of monopotassium phosphate is also noticeable in the ET, TA and FRS categories, responding to the emissions of chemical agents into the environment for the extraction and transport to site. The overall impacts seem to be more dominated by the production emissions associated to potassium sulfate, being present in almost all IC due to its major role in the nutrient solution.

Takin in account the influence of the struvite slow solubility to reduce the emissions of P to water as well as the reduction of the impacts associated to the production of monopotassium phosphate, a great reduction of the impacts of fertilization can be seen.

While the pepper crop shows a clear reduction in emissions related to fertilization with the use of 10 g and 20 g of struvite, sustained production is unclear due to the low content of P in plants. While the production of pepper continues and demands on P can increase, its dissolution and uptake might not be sufficient in time. On the other hand, the lettuce needs were covered for all three cycles for all treatments, showing a P content similar to that of the control treatment. The idea of sustained production for longer periods of time corresponds to the findings of Bonvin et al., 2015 and Rech et al., 2018 urging for the definition of the residual value of the remaining struvite after the initial crop production.

To understand the environmental impact of one year of lettuce cycles, several assumptions were made. To generate the nine-cycle scenario (Supplementary Information Table 4) that would correspond to yearly production, the three initial cycles for our three treatments were taken as references to generate correlations for the P uptake in biomass from the initial P given, as well as the potential yield produced with the P content in the plant biomass (Supplementary Information Fig 8).

Further on, the error detected in this last correlation was subjected to a sensitivity analysis adding a standard deviation of a total 46% to the yield production for a 9 cycle production of lettuce in all treatments. The control was also given a standard deviation of 10%, taking in account that the P fertilization was consistent over time.

The P loss through the leachates was estimated from the average obtained in all treatments due to its direct relation with irrigation. With the following prediction, the total biomass content of yearly production as well as the resulting yield and emissions to water were obtained to further extend the environmental outcome (Fig. 8). The control treatment was estimated with the generated yields and emissions from the three initial cycles. All other fertilizers for all treatments were based on the NS used for the three initial cycles extended for nine production cycles. The obtained emissions were then divided by the obtained total yields.

The LCA for the year's production with the same initial struvite shows a slight emission increase for all ICs, especially for the 5LE and 10LE treatments. The changes observed indicate that the productions obtained for the 5LE and 10LE treatments decrease to a point where the functional unit is reduced and consequently emissions are increased. On the other hand, control treatment yields were sustained in time and maintained close to identical emissions of the three lettuce cycles. The prospective production obtained for the 20LE treatment was similar to that of the control, obtaining results that reduced the environmental emissions for all impact categories compared to the control treatment except ME.

The 20LE treatment maintains the capacity for competitive production in time compared to the other struvite treatments which can also be seen in the sensitivity analysis in fig 11 in the supplementary information, staying below the control emissions in lower production scenarios, especially for FE and MRS. This, however, implies a potential greater emission to water, as reflected in the FE and ME impact categories generated by the leaching of the struvite containing N and P. The use of the discharged water for less demanding crops (Rufí-Salís and Calvo, 2020) can further reduce nutrient leaching into the urban water cycle as well as a reduction and adjustment of the nutrient solution N content with the addition of struvite.

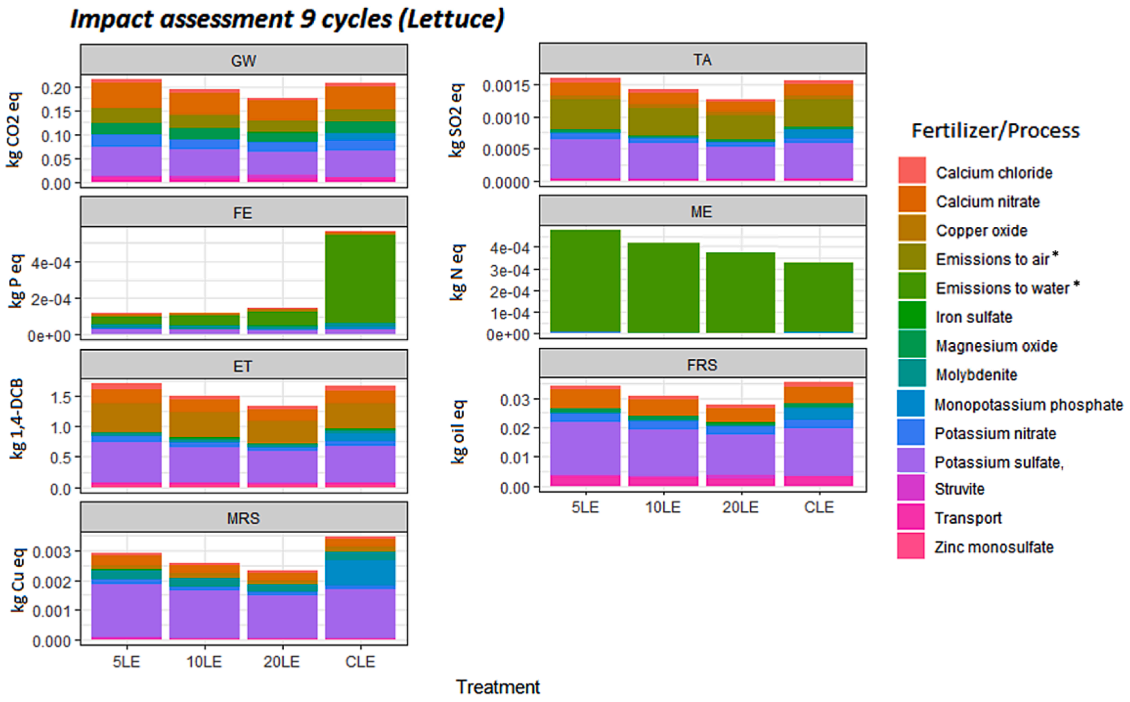
Further loss into the environment can be assessed with a specific analysis of the struvite nitrogen emission factor to the air in the form of ammonia, N_2O and NOx in soilless systems, which is strongly encouraged to determine the GW impact more accurately. This result has been viewed as both interesting and necessary research to understand whether slow dissolution can discourage emission to air or if the composition of N struvite in the form of ammonia will further induce processes of nitrification and denitrification in the substrate.

The findings in this work point out that the successful reuse of struvite in hydroponic production is possible and is been growing in importance, even being used in the fertirrigation for other crops achieving equal results to conventional fertilizers (Carreras-Sempere et al., 2021). Similar work has been made with source separated urine, integrated into hydroponic production as nutrient source, and also using phytoremediation systems for yellow water treatment (Ikeda and Tan, 1998; Yang et al., 2015; Simha and Ganesapillai, 2017; Volpin et al., 2018). These works have found promising results on the reuse of urine although its application can be considered controversial (Simha et al., 2017).

This new way to find circularity in urban ecosystems is deemed as necessary and imposed specially in the waste treatment sector. The capacity to find an added value to the outcome of urban waste can help achieve new environmental goals like the compulsory recovery of P in certain regions of the EU (Kratz et al., 2019). The local P recuperation and local administration can increase the local resilience to P pricing and distribution; therefore the P precipitation and struvite production should be encouraged in WWTP.

5. Conclusions

The main conclusions drawn from the present experiment can be divided into two main aspects, one regarding the production and uptake by the plants and the second regarding the environmental benefits when



Impact assosciated to the system fertilization for 9 consecutive lettuce productions. The obtained emissions have been calculated in relation to the prospective yield as FU. GW (global warming), TA (Terrestrial acidification), FE (Freshwater eutrophication), ME (Marinewater eutrophication), ET (Ecotoxicity), FRS (Fossile resource scarcity), MRS (Mineral resource sarcity). *Emissions to air were based on the emission factors of Ammonia, N2O and NOx obtained for the applied nitrogen; the emissions to water were directly obtained from the nitrogen and phosphorous detected in the water leachate.

compared to the use of mineral fertilizer. We found that the three cycles of lettuce treated with 20 g of struvite had the highest and most sustained overall yield, although such a high struvite concentration resulted in very slow dissolution. In this case, 50% to 70% of the struvite remains undissolved in the substrate after three lettuce cycles, indicating great potential for further consecutive production cycles. Estimations of a year's worth of lettuce production with the same initial struvite indicate sustained production similar to the control, while production for all struvite treatments apart from 20LE would be reduced. While no signs of P deficiency can be seen in the pepper plants, even when obtaining a greater production, the P content was regarded as very low due to the slow struvite solubility. Pepper production was successful in the three-month experiment, although longer production cycles were not tested.

The environmental outcome of the experiment shows a general reduction in the environmental impacts, especially regarding the use and emission of P for freshwater eutrophication and mineral resource scarcity. The production of 20LE is sustained over time, therefore reducing the emissions below the control treatment except for ME. The greater N emissions to water associated with the ME can be reduced by adjusting the nutrient solution, considering the N delivered by the struvite. The findings of this study further encourage the use of struvite in hydroponic production due to the capacity of sustained production of shorter and longer cycle crops as well as the reduction of the environmental impacts compared to mineral fertilizer, such as MKP.

6. Author contributions

All authors were responsible for the conception and design of the study. V. Arcas-Pilz, M. Rufí-Salís, F.

Parada, G. Villalba and X. Gabarrell conceived the original idea for the study. V. Arcas-Pilz, G. Stringari, R.

R. Gonzalez and F. Parada set up, supervised and acquired the data for the experimental tests. V. Arcas-Pilz processed and analysed the data. V. Arcas-Pilz took the lead in writing the manuscript. All authors critically revised the draft for important intellectual content. All authors gave their final approval to the manuscript.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2021.106130.

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