



Sustainable Phosphorus for Soilless Greenhouse Horticulture

An exploration of potential routes

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Referaat

De transitie van 'lineaire' nutriënten, gebaseerd op eindige grondstoffen, naar 'circulaire' nutriënten, brengt veel vragen met zich mee. Waar moeten ze vandaan komen? Hoe kunnen ze herwonnen worden? Hoe toepasbaar zijn circulaire meststoffen? Hoe kunnen risico's door contaminanten uitgesloten worden? In dit rapport bekijken we dit soort vragen voor de glastuinbouw, specifiek grondloze systemen, die specifieke eigenschappen hebben t.o.v. grondgebonden teelten. Specifiek is naar fosfor (P) gekeken, omdat Nederland een P-overschot heeft en het toch van eindige mijnen komt. We hebben de belangrijkste P-rijke reststromen in Nederland geïnventariseerd en verschillende herwinningstechnologieën vergeleken. Bovendien zijn de herwonnen producten zelf tegen de unieke eisen van grondloze systemen vergeleken, namelijk oplosbaarheid en de aanwezigheid van contaminanten. Niet-oplosbare producten zoals struviet kunnen toegepast worden als ze op locatie in zuur opgelost worden. Het effect hiervan op het nutriëntenrecept is gekwantificeerd. Ook concluderen we dat grondloze systemen vaak veel gevoeliger zijn voor contaminanten dan voorgeschreven door de EU Fertilising Products Regulation (FPR) 2019/1009. Met deze resultaten hopen we de transitie naar circulaire nutriënten voor grondloze teelten te bevorderen met betere informatie en begrip van de belangrijkste afwegingsfactoren.

Abstract

The move from a 'linear' supply of nutrients based on finite resources to a 'circular' one involves many unanswered questions. Where should these nutrients come from? How can they be recovered? How applicable are circular fertilisers? How can the risk of contaminants be mitigated? In this report, we examine such questions for soilless greenhouse horticulture, which has unique properties and requirements compared to soil-based systems. Specifically, phosphorus (P) is examined, since the Netherlands has a P surplus and yet it comes from finite natural reserves. We make an inventory of the most important P side-streams in the Netherlands and compare the different available P-recovery technologies. Moreover, recovered P fertiliser products are evaluated using a new methodology specific to greenhouse horticulture's unique requirements: solubility and purity. Insoluble products such as struvite can be applied if dissolved on-site in a separate tank, and we quantify the effect of this on the nutrient recipe. We also conclude that soilless systems are often far more sensitive to contaminants than is currently reflected in the EU Fertilising Products Regulation (FPR) 2019/1009. With these results, we aim to inform and improve confidence and understanding between parties in the transition to circular nutrients for soilless systems.

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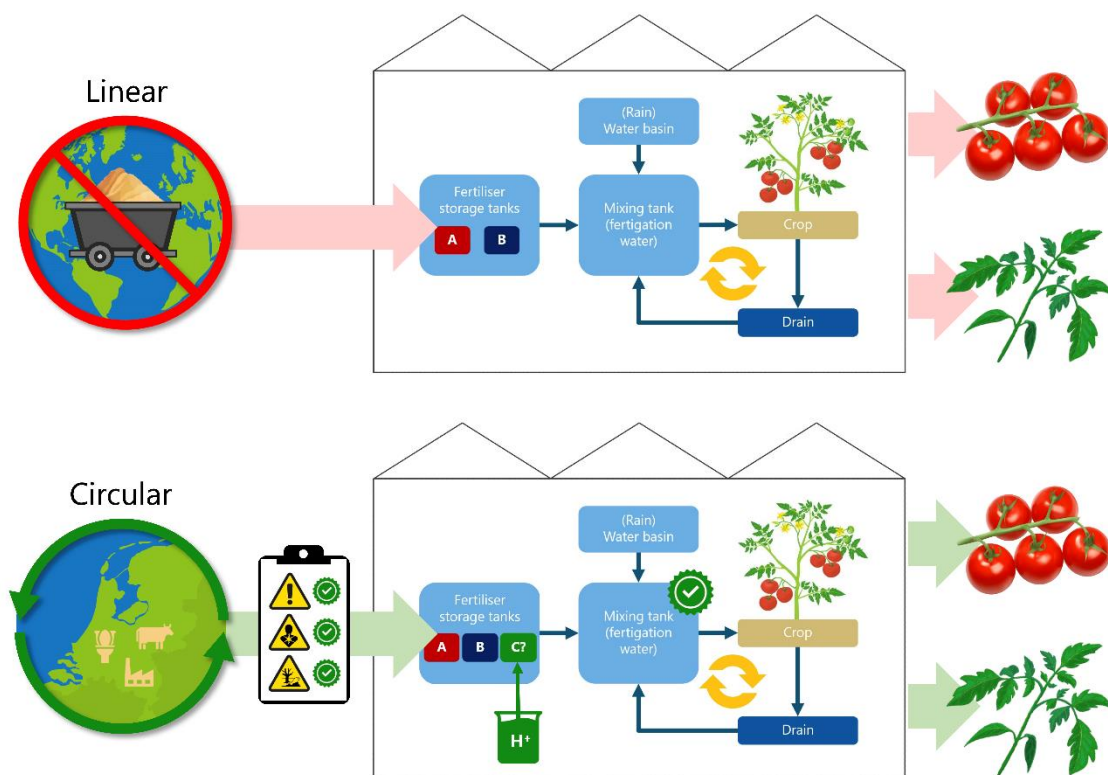
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Summary

Reducing harmful nutrient emissions from the agrifood system is one of the main drivers for establishing circular agriculture as is presented in the Dutch LNV agenda and EU Green Deal. Another driver to close the nutrient loop in the agrifood system is that some nutrients essential for crop growth are sourced from finite natural reserves, putting food production on the long-term at serious risk. Realising a circular agrifood system is one of the main goals of Wageningen University & Research's 'Knowledge Base' (KB, Kennisbasis) programme 'Circular and Climate-Neutral Society' (KB-34), for which this report was written.

In this report we focused solely on phosphorus (P). A significant amount of the P imported and used is lost by emissions (e.g. to wastewater and communal sludge) and yet (unlike with nitrogen) mineral P continues to be imported from finite natural reserves. To identify renewable sources of P potentially suitable for use in greenhouse horticulture, we mapped the possible P sources from side-streams in the Netherlands. We further listed the various P-recovery technologies under development, and evaluated the use-perspective of recycled P in horticulture. Criteria for evaluating the suitability of circular fertilisers based on recycled P were the source volume, state-of-the-art of P-recovery processes, the presence of contaminants and the quality of P, like concentration and solubility.



A graphical abstract of the approach taken in this study, including side-streams, technologies, contaminant risks and adjustments to the fertigation system.

From this study, we conclude that there is more than enough P from side-streams available for soilless greenhouse horticulture in the Netherlands. Within these, there are three categories: manure, sewage sludge, and ashes. There is a trade-off between P concentration and price, but most side-streams leave ample room for processing and recovery costs to produce a fertiliser for soilless systems that could be economically feasible, especially compared to arable farming. The overview of side-streams and technologies may help parties in greenhouse horticulture better understand possible approaches and their trade-offs.

This study also looked at the applicability of P-recovery products not conventionally used in soilless systems. Compounds like calcium phosphates and struvite are not directly soluble, leading to the idea of dissolving them on-site in acid (in a so-called 'C' tank, in addition to the current 'A'- and 'B' tanks). Using simulated chemistry analysis, this was shown to be feasible at a pH of around 3, depending on the compound. The effect of the C tank on the nutrient recipe is also discussed, and is minor for struvite and monocalcium phosphate. Since these products are more affordable than soluble products, this approach may be worth the extra learning curve.

For various recovered P fertilisers, contaminant specifications were used to evaluate potential risks, useful for mitigating risks through 'safety by design'. Assuming 100% accumulation in fertigation water, fruits or residual biomass, we found soilless systems to be orders of magnitude more sensitive to contaminants than the EU Fertilising Products Regulation (FPR) 2019/1009. Many existing recovered P products nearly meet our criteria, however. We hope such contaminant criteria can help manufacturers specify their processes for soilless systems in a way that can gain confidence from growers and consumers.

By looking at various approaches to circular P, taking into account the unique properties and requirements of soilless systems, we hope this methodology and our results can be used to facilitate the transition to circular P, as well being applied to other nutrients

Abbreviations

P	Phosphorus
N	Nitrogen
DM	Dry matter
Ca-P or CaP	Collective abbreviation for three calcium phosphate compounds: tricalcium phosphate ($\text{Ca}_3(\text{PO}_4)_2$), dicalcium phosphate (CaHPO_4) and/or monocalcium phosphate ($\text{Ca}(\text{H}_2\text{PO}_4)_2$)
DCP	Dicalcium phosphate (CaHPO_4)
CFU	Colony forming units
SSP	Single super phosphate ($\text{Ca}(\text{H}_2\text{PO}_4)_2$)
PCP	Precipitated calcium phosphate
MCP	Monocalcium phosphate ($\text{Ca}(\text{H}_2\text{PO}_4)_2$)
P-acid	Phosphoric acid (H_3PO_4)
SS	Sewage sludge
SSA	Sewage sludge ash
MSWI	Municipal solid waste incineration
WWTP	Wastewater treatment plant
FPR	Fertilising Products Regulation
PFC	Product Function Category
CMC	Component Material Category
MRL	Maximum residue limit
DFA	Dutch Fertilising Act
POPs	Persistent organic pollutants
PFASs	Per- and polyfluoroalkyl substances

Term	Definition	Example
By-product	Product other than the main P-containing output from a P-recovery process.	Heavy metal concentrate
Side-stream, residual flow	A side-stream or waste product from agricultural, municipal or other origin (but not P-recovery processes).	Sewage sludge ash
Feedstock	In the context of P-recovery technologies, a resource entering the process. In this report, this always is a residual flow.	Sewage sludge ash

1 Introduction

1.1 Background

Nutrients for crop production are subject to sustainability challenges, especially following large-scale intensification since the mid-20th century (Cordell and White, 2015). Perhaps the most well-known challenge is the environmental impact caused by nutrients leaching into surface water or deposited from the air, which leads to eutrophication (Pluimers, 2001; Wurtsbaugh *et al.*, 2019). This is also the main driver behind the current public debate in the Netherlands on nitrogen emissions (and to a lesser extent, phosphorus emissions). Less commonly-discussed, but at least as important, is the resource depletion associated with the sourcing of these nutrients. Most of the nutrients in synthetic fertilisers come from finite mineral reserves across the globe, with the exception of nitrogen, which is fixated from the air using the Haber-Bosch process, albeit currently with energy mainly from finite fossil fuels (Smith *et al.*, 2020).

To avoid environmental impact and resource depletion generally, various strategies have been proposed (Kirchherr *et al.*, 2017), including increasing resource use efficiency. Efficiency may eliminate environmental impact, but it cannot eliminate resource depletion. It can only slow depletion down. A second strategy is moving from 'linear' to 'circular' supply chains, closing nutrient loops and keeping nutrients in a state of usability within economic production systems. To do this, agricultural systems should source nutrients from side-streams from the agrifood system or consumer waste (de Boer and van Ittersum, 2018; Muscat *et al.*, 2021).

The need for closed resource loops applies to greenhouse horticulture as much as any other agricultural subsector. Soilless closed-loop fertigation systems, such as those used in high-tech greenhouses, are the most nutrient-efficient form of plant production (Goldstein *et al.*, 2016). For example, in Dutch tomato greenhouses, around 95% of the six macronutrients N, P, K, Ca, Mg, and S are taken up by the crop (Van Tuyl *et al.*, 2022). In the Netherlands, greenhouses will be required to have 'virtually zero' N and P emissions by 2027 (Beerling *et al.*, 2014; van der Burgt, 2009), meaning as good as all introduced nutrients will end up in plant biomass.

Table 1 A quantitative overview of the phosphorus balance in current soilless tomato greenhouses in the Netherlands per kg fresh yield, adapted from Van Tuyl *et al.* (2022). Values of zero do not indicate an absence of P, but an amount smaller than the range of certainty of the calculations themselves.

Source/sink	Amount (g P kg ⁻¹)	Amount (g P m ⁻² y ⁻¹)	Proportion of input (%)
Input	0.35	26	100
Tomato fruit	0.21	15	58
Residual biomass	0.14	10	38
Discharge/leakage	<0.00	1	4
Substrate	<0.00	<0	<0

Soilless systems require fertilisers that are rapidly and completely water-soluble, without insoluble residues or contaminants that may accumulate in a closed-loop irrigation system (Sonneveld *et al.*, 2009). In fact, it is precisely the purity and solubility of these nutrients, which are virtually always inorganic, that allows greenhouse horticulture to be so efficient with them (Goldstein *et al.*, 2016). This is different to arable farming, where properties such as low solubility may be tolerable, if not advantageous, as it leads to fewer losses from runoff (Hertzberger *et al.*, 2020). This is just one reason why soilless greenhouse horticulture requires a different approach to circular fertilisers.

Phosphorus (P) is one of the two nutrients mainly responsible for eutrophication, the other one being nitrogen (Khan and Mohammad, 2014). Even if greenhouse horticulture has few emissions left to decrease, other sectors will need to decrease their emissions, capturing and/or repurposing them in a circular economy. At the same time, unlike nitrogen, inorganic phosphorus fertiliser mainly comes from finite reserves of phosphorus rock. Although progress has been made in using nutrients more efficiently in greenhouse horticulture, the source of these nutrients has largely remained the same. At current consumption rates, so-called 'peak phosphorus' has been expected to occur between 2060 and 2100, after which the raw material will be more expensive, of lower quality, or both (Cordell *et al.*, 2009). This leads it to being defined as a 'critical raw material' by the European Union's Raw Materials Initiative (Jama-Rodzeńska *et al.*, 2021).

Previously, over 75% of phosphorus reserves were thought to be in Morocco and the Western Sahara (de Boer *et al.*, 2019). Very recently, a large deposit of high-grade phosphate rock was discovered in Norway, essentially doubling current world reserves and providing phosphorus for an additional 50 years (Harper, 2023; Simon, 2023). These deposits are also expected to be used for batteries, leading to resource competition. Moreover, P-rich by-products such as sewage sludge and manure are still being produced, and greenhouse horticulture may be able to play a role in re-directing them back into the agri-food system.

1.2 Aims

In this report, we examine the possibilities and trade-offs associated with possible routes for circular phosphorus for soilless greenhouse horticulture. We start by giving an overview of the current situation for phosphorus, outlining the requirements of soilless systems (such as purity and solubility) in Section 2. In Section 3, these requirements are used to assess recovered P products that would not normally be used in soilless systems. The report then presents the multiple P-rich flows in the Netherlands in Section 4, before examining and evaluating various processes to turn them into products suitable for greenhouse horticulture in Section 5. Here, the products are evaluated against the requirements of Section 2.3 and other criteria. The report concluded with discussion and conclusion (Section 6).

2 Current Use of Phosphorus in Dutch Greenhouse Horticulture

2.1 Phosphorus Demand

To our knowledge there is little to no data recorded regarding nutrient use in soilless greenhouse horticulture. Only growers who cultivate in soil are obliged to report their phosphorus- and nitrogen use (UO-IMT, 2022). Despite this lack of data, there are some estimates available for phosphorus use. A rough order of magnitude can be established by multiplying P-input of tomato cultivation, $260 \text{ kg P ha}^{-1} \text{ y}^{-1}$ (see Table 1), with the entire Dutch greenhouse area of 10 600 ha (Centraal Bureau voor de Statistiek, 2023a). This approach suggests a total use of 2756 tonnes per year.

However, the total greenhouse area in the Netherlands of 10600 ha is divided over 5900 ha of fruit- and vegetable production (56%) and 4700 ha of ornamental plant production (44%) (Centraal Bureau voor de Statistiek, 2023a). Van der Lugt (2022) assumes a lower P-demand for cut flowers and potted plants, and estimates that greenhouse horticulture uses 2460 tonnes per year. The majority (79%) of this goes to vegetable crops, in part due to a higher productivity and per-hectare nutrient input. In a study regarding sustainable phosphate fertilisers for greenhouse horticulture, similar to this report, Schipper (2022) uses the same estimates as Van der Lugt (2022).

2.2 Fertilisers and Application

Soilless systems make use of fully soluble fertilisers, supplied by manufacturers either as solid salts, or as an already-dissolved aqueous solution, i.e. liquid fertilisers. For the crop, these are functionally equivalent, but solid fertilisers are more labour-intensive as they have to be added to water and stirred before they can be used. An overview of the different phosphorus-containing compounds commonly used in greenhouse horticulture is given in Table 2. Although the most common phosphorus fertilisers in agriculture are calcium salts, this is not the case for soilless systems, as they are not soluble enough. Still, these are used for potted plants in greenhouse horticulture, where they are added to the growing medium and slowly released during the crop cycle.

Table 2 The names and chemical formulae of compounds commonly used in synthetic phosphorus fertilisers in greenhouse horticulture, adapted from Sonneveld et al. (2009). Products with high solubility are sold both in solid and liquid form, with the exception of phosphoric acid, which is only sold as an aqueous solution.

Name	Chemical formula	% P	Solubility
(Ortho)phosphoric acid	H_3PO_4	32	High
Mono potassium phosphate	KH_2PO_4	22	High
Mono ammonium phosphate	$\text{NH}_4\text{H}_2\text{PO}_4$	26	High
Polyphosphate	$\text{HO}(\text{HPO}_3)_n\text{H}$	Trademarks; exact chemical composition not published.	High
Super phosphate	$\text{Ca}(\text{H}_2\text{PO}_4)_2$	20	Low
Dicalcium phosphate	CaHPO_4	20	Low

In soilless systems, fertigation water is mixed with the optimal concentration of different nutrients before being sent to the crop. There are two ways to do this: (1) individual dosage or (2) delivery from so-called 'A'- and 'B' tanks. Individual dosage involves the direct injection of individual fertiliser solutions into the mix, allowing each product to be dosed separately. This is only done with liquid fertilisers. In a system with A- and B tanks, different products are mixed and stored in the tanks before being added to fertigation water. These products can be either solid- or liquid fertilisers. Two tanks are used to avoid nutrients reacting together and forming insoluble precipitates. This happens when phosphorus reacts with calcium and iron. As a result, calcium and iron are only added to (and delivered from) the A tank, and phosphorus is only present in the B tank (Sonneveld *et al.*, 2009).

Recommended concentrations for phosphate in fertigation water range between 0.9 and 1.5 mmol l⁻¹ (De Kreij *et al.*, 1999). Once in the root environment, phosphorus is taken up in the form of H₂PO₄⁻. The pH of fertigation water needs to be controlled to avoid precipitates. At the optimal pH, which lies between 3.5 and 6.5, phosphate is present in the form of H₂PO₄⁻, which does not precipitate together with calcium (Lucas and Davis, 1961). At a pH that is too high, the predominant form of phosphate becomes HPO₄²⁻, which reacts with calcium to form CaHPO₄. Although CaHPO₄ is insoluble, at lower pH levels it becomes soluble again. At extremely high pH levels, phosphate is predominantly present as PO₄³⁻, which reacts with calcium to form tricalcium phosphate (Ca₃(PO₄)₂). Ca₃(PO₄)₂ is also insoluble, but unlike CaHPO₄ this is less easily reversed. Only strong acids can dissolve it again (Voogt, 2023).

2.3 Requirements

2.3.1 Solubility

Fertilisers used in soilless systems must be fully soluble under the typical pH values between 5.5 and 6.5, and between concentrations of 0.9 and 1.5 mmol l⁻¹. Under these conditions, they must also not form precipitates with other nutrients in the solution, to avoid clogging. Certain products, such as calcium salts or struvite, may not be directly soluble in the B tank. However, they could be made soluble in a strongly acidic solution (in a third 'C' tank) before being diluted and added to the fertigation mix. This possibility was investigated using OLI Studio 11.5 (Revision 11.5.1.7) process simulation software (OLI Systems, 2023), and its results are reported in Section 3. Simulations were done to determine whether (1) it was possible to solubilize phosphorus this way, (2) whether the required acid (e.g. HNO₃) and its anion (e.g. NO₃⁻) would still allow the right fertigation mix to be made and (3) which other changes may be necessary.

2.3.2 Contaminants

Fertiliser products can contain various contaminants, depending on the production process used and the level of contaminants in the original feedstock entering the process. Little is known about the accumulation of food safety hazards in circular production systems based on recycled P. New circular systems should therefore include a safety by design approach, to develop strategies for mitigation and control. A (food) safety by design approach involves mapping potential hazards in the entire food production cycle and applying a systematic approach of incorporating safety measures and compliance to existing regulation into the design and development of a food production process. One element of it is mapping the sources and transmission of hazards that relate to the use of agri-food side-stream- and communal wastes, which is shown in Figure 1 (adapted from Focker *et al.* (2022)). This figure demonstrates a selection of hazards present in organic residual flows and is far from exhaustive. However, this report focuses on the food safety hazards that are currently in legislation, which are heavy metals, pathogens, and to a lesser extent pharmaceuticals.

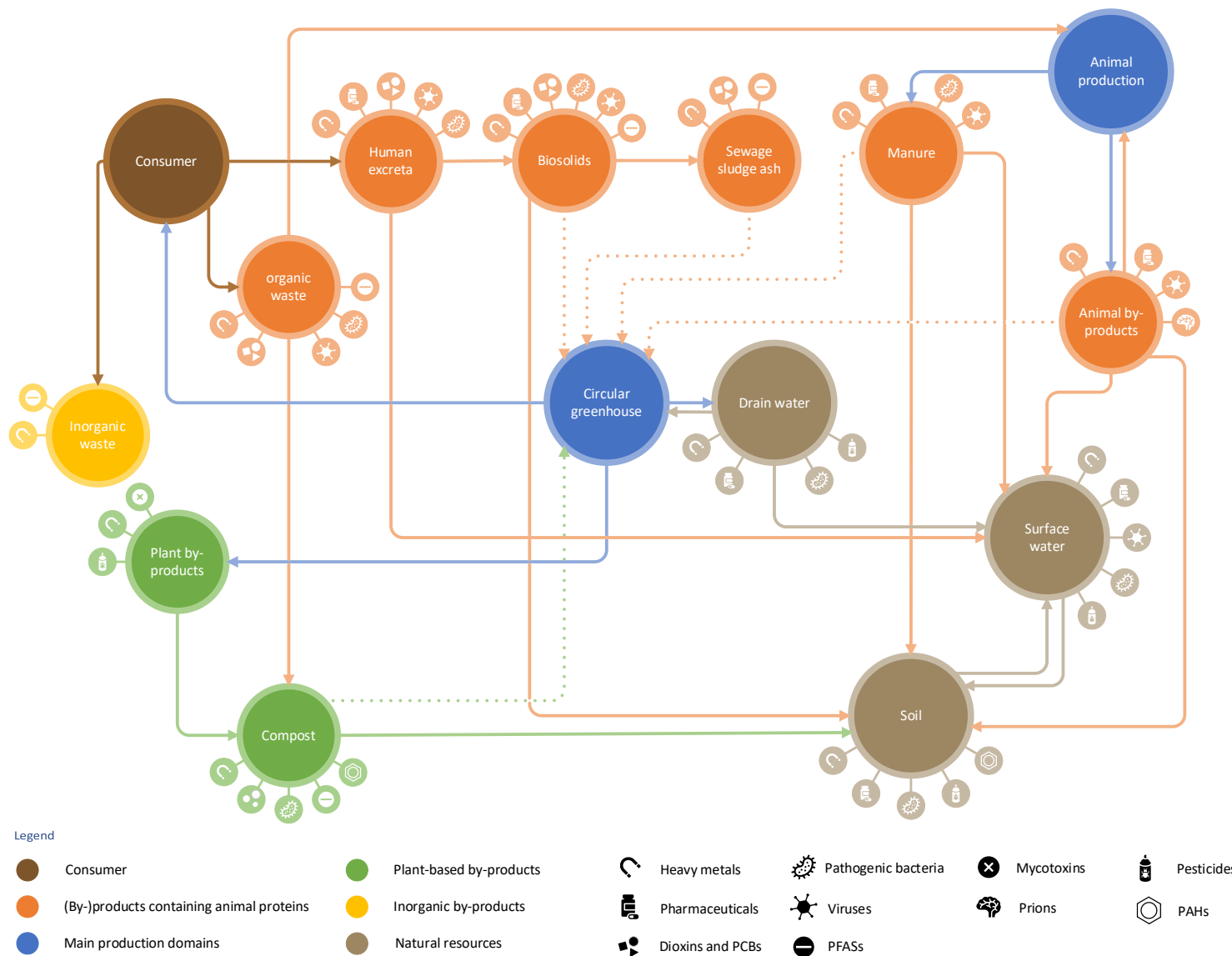


Figure 1 Potential (food) safety hazards in a circular greenhouse resulting from the recycling of side-streams, showing the potential sources and transmission routes into the greenhouse of possible microbial- and chemical contaminants and emission into the environment (surface water, soil). Adapted from Focker et al. (2022).

2.3.2.1 Legislation

The trade and transport of all fertilising products within the European Union is regulated by the new European Fertilising Products Regulation (EU) nr. 2019/1009 (FPR), which came into force in July 2022 and replaces Regulation (EG) nr. 2003/2003 (Faber and Montforts, 2022). The FPR is one of the measures taken by the European Commission to stimulate the circular economy, especially circular agriculture. This is achieved by including a framework in the FPR for promoting the reuse and recycling of nutrients, organic matter, and other valuable resources from organic waste in fertilising products. All fertilising products belong to one of the Product Function Categories (PFC, Table 3) and need to be composed of at least one Component Material Category (CMC). Multiple component materials may be used in a fertilising product, but the component materials must meet the criteria of the listed CMCs. Similarly, the fertilising product (blend) can be composed of different PFCs (Faber and Montforts, 2022). In addition, the FPR includes a framework to protect against risks associated with environmental and health hazards by including criteria for (in)organic contaminants, heavy metals, and pathogens in fertilising products:

- Restrictions for three organic contaminants with a reference point of action in any fertilising product: chloramphenicol (0.15 µg/kg), malachite green (0.5 µg/kg), and nitrofurans and their metabolites (0.5 µg/kg).
- Concentration limits to (heavy) metals. The concentrations are dependent on the (heavy) metal and PFC.
- Concentration limits to polyaromatic hydrocarbons (PAH₁₆)¹: 6 mg/kg in dry compost or digestate.
- Limits for the presence of *Salmonella* spp. and *Escherichia coli* or *Enterococcaceae*.
- Forbids the presence of *Aristolochia* spp. and preparations thereof, chloroform, chlorpromazine, colchicine, dapsone, dimetridazole, metronidazole, and ronidazole.

Table 3 Quality standards to which products regulated by the EU Fertilising Products Regulation (FPR) have to comply.

EU Fertilising product Regulation – product categories	Quality standards
PFC 1. Fertiliser (organic, organo-mineral, or inorganic)	Organic contaminants (when consisting of CMC 3 and/or CMC 5 ¹ or foodstuffs of animal origin ²)
PFC 2. Liming material	
PFC 3. Soil improver (organic or inorganic)	
PFC 4. Growing medium	
PFC 5. Inhibitor (nitrification, denitrification, or urease)	
PFC 6. Plant biostimulant (microbial or non-microbial)	
PFC 7. Fertilising product blend	

1 Organic contaminants listed in EU Fertilising Products Regulation. CMC 3: compost, CMC 5: digestate other than fresh crop digestate.

2 List of pharmacologically active substances prohibited by Annex I (Table 2) to regulation 37/2010 or with a Reference Point of Action as a threshold.

The FPR does not restrict the presence of organic contaminants beyond those mentioned above. However, the FPR does require that the presence of a large number of substances is assessed in component materials for which a Maximum Residue Limit (MRL) is established if that component material would be placed on the market as food or feed (Faber and Montforts, 2022). The fertilising products must be labelled with the maximum concentration followed by a warning if the presence of these substances is above the MRL for food-producing animals described in Commission Regulation EU No 37/2010/37. This labelling is not required when it concerns component materials that do not qualify as food or feed. Compliance with the criteria can be verified with analytical testing or without testing by the manufacturer based on the nature of the production and processing process.

In the Netherlands, the trade and use of fertilisers are currently regulated by the Dutch Fertilising Act (DFA), but the Netherlands is required to incorporate the new FPR into its Fertilising Act. The current DFA contains a long list of limits for organic contaminants (Appendix B, Table B.2) for all products in four out of nine fertilising product categories (Table 4). This list of organic contaminants also applies to waste- and by-products intended as component material of a fertiliser, and for fertiliser or material for co-digestion (Ehlert *et al.*, 2022). However, the FPR has consequences for the analysis of organic contaminants and for the products that are included in the DFA. Thus, in the Netherlands, both the new FPR and DFA are in force, with

¹ Sum of naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene and benzo[ghi]perylene.

some major differences between the two regulations in terms of product categories and contaminants (Ehlert *et al.*, 2022). The Dutch Foundation for Innovation in Greenhouse Horticulture ('Stichting Innovatie Glastuinbouw Nederland', or SIGN) has developed a flowchart for a better understanding of what regulation needs to be followed for using organic waste streams in greenhouse horticulture (Appendix C).

Table 4 *Quality standards for products regulated by the Dutch Fertiliser Act (DFA).*

Dutch Manure and Fertilisers act product categories	Quality standards
Animal manure	-
Growing media	-
EC fertilisers and liming fertilisers	-
Other inorganic fertilisers	Organic contaminants (when fertiliser is from animal or plant origin) ¹
Lime fertilisers, excluding EC-limiting fertilisers	Organic contaminants (when fertiliser is from animal or plant origin) ¹
Sewage sludge	
Compost	
Recovered phosphates	Organic contaminants ¹
Other organic fertilisers	Organic contaminants ¹
Designated waste (when not harmful to the environment	Organic contaminants ¹ , other expected organic contaminants

¹ Organic contaminants listed in the Fertiliser Decree ('Uitvoeringsbesluit Meststoffenwet') and implementation of the Regulation of the Fertiliser Act ('Uitvoeringsregeling Meststoffenwet').

2.3.2.2 Inorganic contaminants

There are various limits for inorganic contaminants, notably heavy metals and other inorganic contaminants (e.g. sodium or aluminium). Firstly, legal limits exist per kg dry matter in a fertiliser, set out by the aforementioned FPR for various fertiliser groups as explained in Section 2.3.2.1. Since soilless horticulture uses inorganic fertilisers, product function category (PFC) 1C applies.

Whilst compliance to EU regulations is necessary, these standards do not provide enough information to know whether these products can be applied in a high-tech recirculating irrigation system. Inorganic contaminants may accumulate in the irrigation water, leading to phytotoxicity risks. They should never reach phytotoxic levels (Ayers and Westcot, 1985). They may also end up in the fruit or residual biomass (stems, leaves and roots), leading to concerns about food safety and applicability for composting respectively, since limits exist for both. Lastly, three of the inorganic contaminants are also micronutrients: copper, manganese and zinc. Ideally, they should remain within concentrations where they can still be taken up by the crop and not accumulate (De Kreij *et al.*, 2003). All these requirements are summarised in Table 5.

Table 5 Criteria for various inorganic contaminants, based on EU legislation, and potential accumulation in three sinks in tomato crop production: fertigation water (phytotoxicity), fruits (food safety) and residual biomass (composting regulations); assuming 100% of the contaminant ends up in the respective sink. The maximum concentration (above which the crop can no longer take up enough to avoid accumulation) is also given. Note the different units per column.

Criterion	EU 2019/1009 (PFC 1C)	Phytotoxicity of fertigation water (accumulation)	Fruits	Residual biomass	Fertigation water (uptake limit)
Unit	g kg DM ⁻¹	μmol l ⁻¹	mg kg ⁻¹	g kg DM ⁻¹	μmol l ⁻¹
Al	-	185.3	-	-	-
As	40	1.3	-	15	-
Cd	60	0.1	0.02**	1	-
Co	-	0.8	-	-	-
Cr	-	1.9	-	50	-
Cu	600	3.1	5*	90	0.75
Hg	1	-	0.01*	0.3	-
Mn	-	3.6	-	-	10.0
Ni	50	3.4	-	20	-
Pb	120	24.1	0.05**	100	-
Zn	1500	30.6	-	290	4.0
Source	FPR (EU 2019/1009)	Ayers and Westcot (1985)	*: EU 2023/915 **: EC 396/2005	Certificeringscommissie Keurcompost (2023)	De Kreij <i>et al.</i> (2003)

To estimate the accumulation of inorganic contaminants in irrigation water, fruits, residual biomass (stems and leaves) and substrate, as shown in Table 5, calculations were done under the assumption that 100% of the contaminants would end up in these sinks. Since these were the most extreme scenarios, values under the limit gave confidence that the related contaminant would not be an issue. This calculation was done assuming a high-wire tomato crop, using numbers from Van Tuyll *et al.* (2022a). The volume of irrigation water assumed to be present in substrate mats during the crop cycle was obtained from personal contact with Wim Voogt. The calculation parameters' values are summarised in Table 6.

Table 6 A summary of the values used to calculate the potential accumulation of inorganic contaminants in various sinks: irrigation water, fruits, residual biomass (stems, leaves), and substrate.

Quantity	Assumed value	Unit	Source
Volume of irrigation water present in system	10	l m ⁻²	Stanghellini <i>et al.</i> (2019)
Fresh yield	73.4	kg m ⁻²	Van Tuyll <i>et al.</i> (2022a)
Residual biomass production	13.9	kg m ⁻²	
Residual biomass dry matter content	0.123	kg DM kg ⁻¹	
Required phosphorus input	25.7	g P m ⁻²	

Using the parameters shown in Table 6, the values of Table 5 were converted to mg per kg P in the fertilising product. The resulting values are presented in Table 7. Since FPR limits are defined in g per kg dry matter and different phosphorus fertilisers have different P contents per kg dry matter, the FPR limits could not be converted to mg per kg P. The limit in fertigation water related to uptake for copper, manganese and zinc was obtained by dividing the concentrations in Table 5 by 1.25 mmol P l⁻¹ (De Kreij *et al.*, 2003).

Table 7 Criteria from Table 5, excluding legislation, converted into mg per kg P based on parameters in Table 6. The strictest criterion is highlighted in orange. In green, the maximum concentrations for the micronutrients copper, manganese and zinc are given, under which the crop can take them up and accumulation will not occur. Since these are higher than the phytotoxicity requirements, these will not accumulate to phytotoxic levels, hence the brackets.

Criterion	Fertigation water (phytotoxicity)	Fruits	Residual biomass	Fertigation water (uptake)
Unit	mg kg P ⁻¹	mg kg P ⁻¹	mg kg P ⁻¹	mg kg P ⁻¹
Al	155 703	-	-	-
As	31.1	-	13 600	-
Cd	3.11	57.1	910	-
Co	15.6	-	-	-
Cr	31.1	-	45 500	-
Cu	(62.3)	14300	81 900	1030
Hg	-	28.6	273	-
Mn	(62.3)	-	-	1 830
Ni	62.3	-	18 200	-
Pb	1 560	143	91 000	-
Zn	(623)	-	264 000	5630

Table 7 shows that the most sensitive sink is irrigation water for all inorganic contaminants except lead and mercury. In other words, the most likely risk is phytotoxicity, should these contaminants fully accumulate in the irrigation water. For lead and mercury, the most sensitive sink are the fruits. The requirements of residual biomass for composting are the least sensitive. If they are violated, the requirements for irrigation water and fruits will have been violated as well, many times over. For the micronutrients copper, manganese and zinc, the maximum concentration to ensure the crop can take up these nutrients is far higher than the phytotoxicity limits calculated assuming 100% accumulation in fertigation water, meaning such accumulation would not occur in reality.

The FPR sets an upper limit of 20% sodium oxide (Na₂O) for inorganic fertilisers, equating to 14.8% sodium. This is a fixed legal requirement, but in recirculating irrigation systems, sodium content must be as low as possible to avoid accumulation. Applying Stanghellini *et al.* (2019)'s equations to tomatoes, using a maximum rootzone concentration of 25 mmol l⁻¹ (Voogt *et al.*, 2022), results in a maximum concentration of 1500 µmol Na l⁻¹ before accumulation becomes a concern. This differs between crops: for instance, for peppers, this should be 300 µmol Na l⁻¹ (Voogt *et al.*, 2021). The concentration for roses should be virtually zero, since roses take up a negligible amount of sodium (Stanghellini *et al.*, 2019). Regardless of the crop, this total concentration is a sum of sodium from (1) irrigation water, (2) other fertilisers and (3) the recovered phosphorus fertiliser studied here. Fertilisers contribute 60 µmol Na l⁻¹ to fertigation water, particularly from chelating agents. Typical rainwater in Westland, the region in the Netherlands with the most greenhouses, contains 230 µmol Na l⁻¹ (Van Staalduinen and Voogt, 2013). With a total of 1500 µmol Na l⁻¹ for tomatoes, this leaves some 1200 µmol Na l⁻¹ for tomatoes that can come from recovered phosphorus fertilisers.

2.3.2.3 Pharmaceutical products and persistent organic pollutants

The current Fertiliser Act and the new FPR legislation do not include limits for pharmaceutical contaminants or persistent organic pollutants (POPs) in fertilisers, with an exception for fertilisers composed from CMCs that could be placed on the market as food or feed. However, the presence of POPs, especially PFAS, in the environment and food products has received more attention in recent years. Thus, it cannot be ruled out that additional legalisation will include limits for pharmaceutical contaminants and POPs in the near-future. These limits are necessary in circular systems because these contaminants are reintroduced into the food production system (Figure 1). This may lead to the accumulation of pharmaceutical compounds and other POPs in edible parts of the plants. Phytotoxicity effects vary depending on the pharmaceutical product, its concentration, the crop species and even the crop variety. Some studies have shown toxicity only in concentrations much higher than typically found in wastewater (Pino *et al.*, 2016), whereas others show a far higher sensitivity (D'Abrosca *et al.*, 2008; Pino *et al.*, 2016).

Scientific organisations across the European Union, including Wageningen University & Research, have written a scientific response document on the new Directive for Soil monitoring and Resilience ((COM(2023) 416). The scientific community welcomes the new directive in this document, and urges to set limits to various organic contaminants including pharmaceutical and POPs to monitor and improve soil health. Thus, the potential food safety hazards need to be controlled before reusing organic waste in food production (Various Scientific Organisations across the EU, 2023).

2.3.2.4 Pathogens

The DFA and FPR describe limits for pathogens in fertilisers. These limits are mostly for *Salmonella* spp., *E. coli*, and *Enterococcaceae*. There are additional limits for microbial plant biostimulants (PFC 6), which are shown in Table 8. However, organic waste such as manure and sewage sludge is allowed for use as fertiliser when pathogens are removed via biological, chemical, or thermal treatments. In addition, long-term storage or any other manner that results in the removal of pathogens is allowed.

Table 8 Overview of limits for human pathogens in biostimulants in CFU g⁻¹.

Micro-organism or its toxins/metabolites	Limit in colony-forming units (CFU)
<i>Salmonella</i> spp.	Absence in 25 g or 25 ml
<i>Escherichia coli</i>	Absence in 1 g or 1 ml
<i>Listeria monocytogenes</i>	Absence in 25 g or 25 ml
<i>Vibrio</i> spp.	Absence in 25 g or 25 ml
<i>Shigella</i> spp.	Absence in 25 g or 25 ml
<i>Staphylococcus aureus</i>	Absence in 25 g or 25 ml
<i>Enterococcaceae</i>	10 CFU g ⁻¹
Anaerobic plate count, unless the microbial plant stimulant is an aerobic bacterium	10 ⁵ CFU g ⁻¹ or ml
Yeast and mould count, unless the microbial plant biostimulant is a fungus	1000 CFU g ⁻¹ or ml

3 Applicability of Insoluble P-Products

As explained in the previous chapter, it is crucial that the fertilisers used in soilless growing systems are fully soluble and do not form precipitates under typical concentrations and pH levels. This is a given for phosphoric acid, polyphosphates and mono-ammonium and -potassium phosphate. Some P-recovery processes can produce phosphoric acid, but others produce struvite, calcium phosphates or vivianite, which are not fully soluble.

Though vivianite is inappropriate as a fertiliser, it can be dissolved in potassium hydroxide (KOH) to form a potassium phosphate solution (leaving behind iron and heavy metal precipitates) (Wilfert *et al.*, 2018). This potassium phosphate can be applied to greenhouse horticulture. Therefore, vivianite can be seen as an intermediary product rather than a fertiliser in itself. The remaining two products would have to be dissolved under highly acidic conditions on-site, in a separate tank, before being mixed into fertigation water. Whether this is possible and under which conditions is reported in this section, where results from the OLI Studio 11.5 (Revision 11.5.1.7) process simulation software (OLI Systems, 2023) are presented.

3.1 Methodology

The methodology consists of two steps: (1) the determination of the precipitation point in OLI Studio and (2) the allocation of other nutrients to the A-, B- and C tanks to determine feasibility of a nutrient recipe.

3.1.1 Precipitation Point

The C tank was assumed to have a volume of 1 m³, the typical volume of an IBC used as A- and B tanks in greenhouse horticulture. The concentration in the A- and B tanks is usually 100 times the concentration of the nutrient recipe (Van der Lugt *et al.*, 2020). The equivalent amount of phosphate was added to the C tank in OLI Studio for the aforementioned products.

Next, a survey was done in which acid was progressively added until no solids remained, and therefore all product was dissolved. This occurred at the precipitation point. Due to the high concentration of nitrate in most fertigation water recipes (De Kreij *et al.*, 1999), nitric acid was used. The resulting ratio of phosphate to nitrate in the C tank was used in the next step, where nutrient recipes were calculated.

3.1.2 Fertiliser Allocation

In this step, fertiliser products were allocated to the A-, B- and C tanks, to allow for the correct nutrient recipe when water from these tanks were mixed with irrigation water later on. The following recipe for soilless tomatoes was used (Table 9). Since the concentrations of micronutrients are orders of magnitude lower, these were ignored.

Table 9 The nutrient recipe used in the fertiliser allocation calculations, for tomatoes. Concentrations are in mmol l⁻¹; EC is in dS m⁻¹.

	NH ₄ ⁺	K ⁺	Ca ²⁺	Mg ²⁺	NO ₃ ⁻	SO ₄ ²⁻	H ₂ PO ₄ ⁻	EC
Value	1.2	9.5	5.4	2.4	16	4.4	1.5	2.6

Before allocating each fertiliser to a specific tank, the amount added needed to be determined. The amount of phosphorus-containing fertiliser was determined first, to achieve the desired concentration of phosphorus. The amount of nitrate from acid to go with this phosphorus came from the OLI Studio simulation and was

added to the C tank as well, along with any other ions present in the fertiliser (e.g. Ca^{2+} in the Ca-P salts). Once this was done, the rest of the fertilisers were added in the ratio closest to the nutrient recipe, using Excel's *Solver* optimisation tool.

Subsequently, the amounts of fertiliser were allocated to the A-, B- or C tank (Table 10). The C tank of course contained the phosphorus fertiliser and its accompanying dissolving acid. The allocation of the other fertilisers to all three tanks was done such that the total kg added per tank was as equal as possible. This is because viscosity is linked to the mass concentration of fertiliser added, and viscosity should be as equal as possible for the dosing units to supply solution from the three tanks correctly. If an equal viscosity is not possible, the dosing units will need to be re-calibrated. Potassium hydroxide, a lye, is stored in a separate tank, and is used to compensate for extra H^+ ions from the C tank.

Table 10 Possible tanks where fertiliser products can be stored, and lye. Phosphorus fertilisers are not included, since different compounds were evaluated and these were always added to the C tank.

Product	Chemical formula	A	B	C	Lye
Ammonium nitrate	NH_4NO_3	X	X	X	
Calcium nitrate	$\text{Ca}(\text{NO}_3)_2$	X			
Magnesium sulphate	MgSO_4	X	X	X	
Potassium sulphate	K_2SO_4	X	X	X	
Potassium nitrate	KNO_3	X	X	X	
Potassium hydroxide	KOH				X

3.2 Results

In this section, the solubility and applicability of four alternative P compounds is given. The maximum pH and corresponding nitric acid concentration in the C tank is given in Table 11. Based on pH and nitric acid concentration, all four compounds can be fully dissolved in a C tank at a pH of approximately 3. No sulphuric acid was required. However, the resulting $\text{NO}_3^-:\text{P}$ ratio varies considerably between the four options.

Table 11 The results of the OLI simulation for a hypothetical 'C-tank' where P fertilisers are dissolved on-site in acid, with the concentration, the maximum pH required for it to fully dissolve, and the corresponding minimum concentrations of anions from the acid used. Fertilisers are given in descending order of $\text{NO}_3^-:\text{P}$ ratio.

Fertiliser	C-tank concentration (mmol l^{-1})	Maximum pH (precipitation point)	HNO_3^- concentration (mmol l^{-1})	$\text{NO}_3^-:\text{P}$ ratio (-)
$\text{Ca}_3(\text{PO}_4)_2$	75	2.8	310	2.1
Struvite	150	3.3	300	2.0
$\text{Ca}(\text{HPO}_4)$	150	3.0	160	1.1
$\text{Ca}(\text{H}_2\text{PO}_4)_2$	75	3.2	10	0.1

The effects of this ratio on subsequent nutrient mixing is shown in Table 12. This shows that, assuming 100% of phosphorus coverage comes from these fertilisers, it is not possible to obtain the exact recipe, though it is possible to come close.

Table 12 The hypothetical concentrations of different ions in the three tanks (mmol l⁻¹), assuming a C-tank composition as described in Table 11. An asterisk (*) indicates a total sum different to that of the original recipe. Dividing the sum by 100 gets the fertigation water concentration (Table 9). 'Δ' stands for the difference in fertigation water concentration compared to the target recipe.

Concentration (mmol l ⁻¹)		NH ₄ ⁺	K ⁺	Ca ²⁺	Mg ²⁺	NO ₃ ⁻	SO ₄ ²⁻	H ₂ PO ₄ ⁻
Target (Table 9)	A	108	200	540	0	1588	0	0
	B	12	750	0	240	12	440	150
	Sum	120	950	540	240	1600	440	150
Ca ₃ (PO ₄) ₂	A	0	0	438	0	875	0	0
	B	64	406	0	230	256	338	0
	C	54	229	225	17	469	82	150
	Lye	N/A	315	N/A				
	Sum	120	950	663*	247*	1600	420*	150
	Δ	0.0	0.0	+1.2	+0.1	0.0	-0.2	0.0
Struvite	A	0	0	623	0	1245	0	0
	B	0	558	0	131	0	410	0
	C	150	92	0	150	355	18	150
	Lye	N/A	300	N/A				
	Sum	150*	950	623*	281*	1600	428*	150
	Δ	+0.3	0.0	+0.8	+0.4	0.0	-0.1	0.0
Ca(HPO ₄)	A	0	0	501	0	1002	0	0
	B	68	541	0	142	249	298	0
	C	52	244	150	94	369	142	150
	Lye	N/A	165	N/A				
	Sum	120	950	651*	236*	1600	440	150
	Δ	0.0	0.0	+1.1	0.0	0.0	0.0	0.0
Ca(H ₂ PO ₄) ₂	A	0	0	465	0	930	0	0
	B	62	536	0	141	380	250	0
	C	58	406	75	99	290	190	150
	Lye	N/A	8	N/A				
	Sum	120	950	540	240	1600	440	150
	Δ	0.0	0.0	0.0	0.0	0.0	0.0	0.0

For the di- and tri-calcium phosphate, the fertigation water would contain over 1 mmol l⁻¹ too much Ca. Tricalcium phosphate (Ca₃(PO₄)₂) results in slightly more Mg as well. They also result slightly less Mg, but this is within 0.1 mmol l⁻¹ in fertigation water, unlikely to be significant. Tricalcium phosphate is the only calcium phosphate to not allow for enough sulphate (0.2 mmol l⁻¹ shortage), likely due to it requiring the most HNO₃ to dissolve. The more HNO₃, the more H⁺ ions, requiring more KOH to neutralise. This means less K₂SO₄ needs to be added in the end. To solve this, sulphuric acid (H₂SO₄) could be used in the C tank. Encouragingly, monocalcium phosphate (Ca(H₂PO₄)₂) shows no differences compared to the target recipe.

Struvite shows different results. Firstly, since per mol P it contains 1 mol ammonium (NH₄⁺), it is impossible not to exceed the amount of NH₄⁺ in the recipe, though this only happens by 0.3 mmol l⁻¹. This is also likely the cause of excessive Mg. It also shows a 0.1 mmol l⁻¹ shortage in sulphate.

The total mass of fertiliser added to each tank, following the optimised allocation of fertilisers using Excel's *Solver*, is given in Table 13. All new combinations show that the B- and C tanks have similar viscosities, but that the A tank always has a higher viscosity (even if only slightly). This is because calcium nitrate (Ca(NO₃)₂) can only be added to the A tank, and therefore cannot be allocated to other tanks. In the original scenario, identical viscosities are obtained between both tanks, meaning the dosing units do not need to be adjusted.

Table 13 The total mass of fertiliser (kg) added per tank for each phosphorus fertiliser, which is related to viscosity.

kg per tank	Original	Struvite	$\text{Ca}_3(\text{PO}_4)_2$	$\text{Ca}(\text{HPO}_4)$	$\text{Ca}(\text{H}_2\text{PO}_4)_2$
A	121	102	72	82	76
B	121	64	71	68	73
C	N/A	64	71	68	73

4 Phosphorus from Side- and Waste Streams

Organic side- and waste streams, ranging from by-products of agrifood production, to organic wastes from municipal wastewater treatment, to exported or incinerated manures, can be a source of nutrients. These cannot be directly applied in (soilless) greenhouse horticulture, but can be used to recover P and convert it into products that can be. Side-streams were selected based on an analysis of phosphorus flows in the Netherlands, done by Smit *et al.* (2010), supplemented with other publications for a more up-to-date picture as well as data on phosphorus content. An overview of all the data sources used can be found in Appendix A. The sources are by-products of the agricultural sector, households, and industry. For each side-stream, we look at the quantity of phosphorus in the Netherlands, the P concentration, and potential contaminants. Lastly, we examine economic considerations: not only the price per kg of P, but also current uses of the side-stream, with which greenhouse horticulture may need to compete.

4.1 Side- and Waste Streams as a Source of P

4.1.1 Manure

Manure is by far the biggest circular source of phosphorus in the Netherlands. An overview of manures available, their P concentrations and value is given in Table 14.

Table 14 An overview of the total amount of phosphorus ($t P y^{-1}$) for various manure-based residual flows in the Netherlands, along with their P concentrations ($g kg fresh^{-1}$) and costs ($€ t P^{-1}$).

By-product	Amount ($t P y^{-1}$)	P concentration ($g kg^{-1}$)		Cost ($€ t P^{-1}$)
Ruminant manure	17 000	Collected	0.7	-
		Solid fraction	1.9	-
		Liquid fraction	0.5	-
		Digestate	0.7	-11 400
Pig manure	9 000	Collected	1.8	-
		Solid fraction	4.7	-
		Liquid fraction	1.6	-
		Digestate	2.0	-2 800
Poultry manure	7 000		10.8	190

In the Netherlands the largest amount of manure-based phosphorus comes from ruminants, followed by pigs and poultry. The concentration of P in these sources varies, with ruminant manure being the lowest and poultry manure the highest. The way manure is produced, collected and processed affects the P concentration of the different fractions. The P content of the solid fraction is much higher than that of the liquid fraction after separation, and also higher than digestate (after methanogenic fermentation) or manure before mechanical separation. P is present in manure in both organic and inorganic forms. As manure mineralises, organic forms are converted into inorganic forms. About 60% of P in manure is inorganic, though there are variations (Pagliari and Laboski, 2012).

Direct application of liquid manure to arable land is by far the largest use in the Netherlands. To avoid overfertilisation and runoff to the environment, farmers are limited to 40 kg phosphate (13 kg P) per year per hectare of arable land (Rijksdienst voor Ondernemend Nederland, 2023). If more manure is produced than can be legally applied to arable land, the surplus needs to be exported or processed. This is usually done via biodigesters, producing a watery fraction ('dunne fractie' in Dutch) and a digestate ('dikke fractie'),

which contains most of the P. Chicken manure is mostly incinerated, leaving behind an ash rich in P and K, often exported for use in fertilisers (De Graaff, 2017). Ashes are discussed in Section 4.1.3.

On the whole, the Netherlands has a significant manure surplus that cannot be applied to land, leading to an equivalent of 16 000 t P being exported in 2021 (Centraal Bureau voor de Statistiek, 2023b). A supply chain of circular P for greenhouse horticulture could (1) use part of the excess manure or (2) remove part of the P from manure, allowing farmers to apply the remaining P-depleted manure as soil improver to their land.

4.1.1.1 Contaminants

The livestock farming sectors all use various antibiotics: tetracyclines, macrolides and sulphonamides combined with trimethoprim. Aminopenicillins and quinolones are used for poultry and bovines (Bonten and Van Geijlswijk, 2022). In addition, coccidiostats are used extensively in the poultry sector for preventing and treating coccidiosis. A large fraction of these veterinary drugs – generally over half (Kim *et al.*, 2011) – is excreted unchanged via urine and faeces and has been previously detected in faeces from various animals (Berendsen *et al.*, 2015). The levels of antibiotics found range from low µg/kg to mg/kg.

Heavy metals are another concern in animal manure when considering application as fertilisers. Copper and zinc are added to animal feed in concentrations in excess of the animal's nutritional requirements to prevent diarrheal disease as an alternative to in-feed antibiotics and to promote (Zhen *et al.*, 2020) growth (Yazdankhah *et al.*, 2014). Multiple studies have shown that high manure application significantly increases the total concentrations of soil cadmium, chromium, copper, and zinc (Focker *et al.*, 2022; Lu *et al.*, 2014; Nomeda *et al.*, 2008; Zhen *et al.*, 2020).

PFASs are extremely resistant to degradation, and can be found in organic waste products, including livestock manures, urban sewage sludges and composts (Munoz *et al.*, 2021). There are currently no maximum residue limits for PFASs in manure or fertilisers.

Microbiological hazards could be present in manure as well. Pathogenic bacteria of concern in manure are *Campylobacter coli* and *jejuni*, *Bacillus anthracis*, *Brucella abortus*, *Escherichia coli*, *Leptospira* spp., *Listeria monocytogenes*, *Mycobacterium bovis*, *Mycobacterium avium paratuberculosis*, *Salmonella* spp., and *Yersinia enterocolitica* (Focker *et al.*, 2022). In the current legislation, both in the FPR and DFA, there are limits for *Salmonella* spp. and *E. coli*. Other microbiological hazards could be viruses such as avian-swine influenza and Hepatitis E, and parasites might be present in manure, including *Balantidium coli*, *Cryptosporidium parvum*, *Giardia* spp., and *Toxoplasma* spp. (Millner *et al.*, 2009).

4.1.2 Sewage

A significant part of the P used for food production ends up in communal wastewater, making up the second largest amount of P after animal manure. Wastewater is treated in wastewater treatment plants (WWTPs), resulting in a liquid effluent, from which most of the N is removed, and a sludge, which can be incinerated into sewage sludge ash (SSA). As P is removed from incoming sewage water, the effluent contains a low enough concentration of P and N to be released into surface water, whereas the sludge contains solid organic matter and minerals. In the Netherlands about 13 kt P enters the WWTPs annually (Table 15), more than enough to cover greenhouse horticulture's P demand. In the Netherlands, nearly all sludge is incinerated and the SSA used as filling material in asphalt or landfilled (Centraal Bureau voor de Statistiek, 2023c).

Table 15 The total amount of phosphorus from sewage, its P content, and its estimated cost.

By-product	Amount (t P y ⁻¹)	P content (g kg ⁻¹)	Cost (€ t P ⁻¹)
Wastewater	13 100	0.007	N/A
Sewage sludge (total)	11 400	26	-5 000
Sewage sludge (unincinerated)	500		
Effluent	1 700	<0.001	N/A

Most of the P entering WWTPs in the Netherlands ends up in SSA (see Section 4.1.3 on ashes), though 13% still leaves as effluent and is therefore lost to the environment. This lost amount has decreased over the last decade (Centraal Bureau voor de Statistiek, 2022c). WWTPs pay to have their sludge removed and processed, which costs around €100 to €130 per tonne (Brummelaar, 2020; Ruijter, 2023b) but may be higher depending on the exact case (Perree, 2020).

Recovery of phosphate from sewage sludge and its ash is widely investigated, due to multiple factors: (1) legislation in certain countries requires phosphate from sewage sludge (ash) in wastewater treatment plants or sewage sludge incineration plants to be recovered; (2) the drive of these plants to optimise their waste management technologically and/or economically; (3) social and environmental responsibility (Ploteau *et al.*, 2021a; Sichler *et al.*, 2022). For example, in Germany, from 2029 onwards, phosphate will have to be recovered from sewage sludge if the P content is above 20 g kg⁻¹ dry solids. If the P content exceeds this limit, plants will have to either recover 50% of the phosphorus from this sewage sludge, or, alternatively, decrease the phosphate concentration in sludge after recovery to below 20 g kg⁻¹ dry solids. For sewage sludge ash, 80% of the phosphorus will have to be recovered. This upcoming legislation also means that Germany will have less capacity to process Dutch sewage sludge that is exported. In 2017 this was 20% of Dutch sludge (Perree, 2020), whereas now it is around 8% (Centraal Bureau voor de Statistiek, 2022a).

4.1.2.1 Contaminants

Sewage sludge contains a wide range of contaminants related to consumer and industrial products and applications. A recent study by Gustavsson *et al.* (2022) estimated that wastewater in Sweden contains more than 2,000 chemicals. The main contaminants in communal wastewater are PFASs, surfactants, plasticizers, organohalogens (including dioxins, PCBs, PCAs, and brominated flame retardants), pharmaceutically active- and medical compounds (including antibiotics, sedatives, contrast enhancing agents, etc.), polycyclic aromatic carbons, pesticides, organophosphate flame retardants, and heavy metals. Sewage sludge contains a number of microbiological hazards as well, including *Campylobacter jejuni*, *E. coli*, *L. monocytogenes*, *Salmonella* spp. In addition, parasites and viral infectious pathogens are commonly detected in sewage sludge. Especially the viral infectious pathogens including adenovirus, enterovirus, and norovirus are seen as the highest microbiological hazard in sewage sludge (Hamilton *et al.*, 2020).

4.1.3 Ashes

Several ashes resulting from incineration of by-products can be considered a source of P (Table 16), which is present in the form of tricalcium phosphate (Ca₃(PO₄)₂), a compound unsuitable for direct use in soilless fertigation systems. One source, particularly relevant for the Netherlands, is chicken manure from an annual production of 500 million broiler chickens. 30% of chicken manure is incinerated (Gollenbeek, 2022), leaving behind an ash rich in P and K, often exported for use in fertilisers (De Graaff, 2017).

Contrary to many other European countries, virtually all sewage sludge in the Netherlands is incinerated (Centraal Bureau voor de Statistiek *et al.*, 2022; Donatello and Cheeseman, 2013). From a disposal perspective, ash production from sewage sludge is economically beneficial due to mass reduction and the elimination of organic pollutants, microorganisms, and pathogens (Gorazda *et al.*, 2017). The resulting SSA contains approximately 10% phosphorus (Gerritsen *et al.*, 2021). Currently, SSA is used in construction materials or is stored in empty mines (Oerlemans, 2022). Both destinations represent a loss of phosphorus as it takes P out of circulation.

The two main sewage sludge incineration plants in the Netherlands, HVC and SNB, have the potential to produce about 2 200 and 2 500 t P annually (Gerritsen *et al.*, 2021). Half of the Dutch regional water authorities ('Waterschappen') send their sewage sludge to these two companies, with the other half getting their sludge picked up and processed on a contract basis (Unie van Waterschappen, 2020). Of these, GMB is an important company, which has been looking at ways to valorise sludge to reduce its volume before incineration (GMB, 2021).

Table 16 Ashes and the potential amount of phosphorus they represent, their P content, and cost. SSA stands for 'sewage sludge ash' and MSWI for 'municipal solid waste incineration'.

P flow	Amount (t P y ⁻¹)	P content (g P kg ⁻¹)	Cost (€ t P ⁻¹)
SSA	9 230	80	0?
MSWI ash	5 000	7	0?
Chicken manure ash	7 000	80	187
Animal carcass ash	3 600	180	535

4.1.3.1 Contaminants

Turning sewage sludge and animal manure into ashes reduces the number of contaminants. However, recent research shows that short-chain PFASs are still present in sewage sludge ashes (Björklund *et al.*, 2023; Liu, S. *et al.*, 2021). The persistent chemicals such as PFASs are still present in sewage sludge ashes indicates that other persistent chemicals such as dioxins could be present in sewage sludge as well. However, there is limited information about the fate of POPs during incineration and the relation between POPs in sewage sludge and sewage sludge ashes. Furthermore, there is no information about the fate of veterinary drugs and coccidiostats during incineration of chicken manure.

Inorganic contaminants, such as heavy metals, are observed to accumulate in ashes of sewage sludge and manure. In particular, arsenic, cadmium, copper, nickel, lead, zinc and mercury remain (Werle and Dudziak, 2014).

4.1.4 Bone Meal

Bone meal produced in the Netherlands represents 10 500 t of phosphorus (Table 17), of which 3 100 t is used as additive in porous ceramics, where it adds plasticity and compressive strength. The remaining 7 400 t is used as a fertiliser (Smit *et al.*, 2010). It has a P-content of around 7-9%, with phosphorus bound to calcium, mostly as apatite (Rey *et al.*, 2009).

Table 17 Bone meals, the total amount of phosphorus they represent, P content and cost (bone meal used as fertiliser in soil).

Bone meal	Amount (t P y ⁻¹)	P content (g kg ⁻¹)	Cost (€ t P ⁻¹)
Total/average	10 500	70	18 874
Bovine	N/A	90	N/A
Pigs	N/A	94	N/A
Poultry	N/A	85	N/A

Bone meal is marketed as a slow release fertiliser, as the nutrients will be released over the span of months. In terms of contaminants, it can contain traces of tetracyclines, an antibiotic group that is reported to accumulate in bone tissue of chickens and pigs (Kühne *et al.*, 2000; Odore *et al.*, 2015), and is among the more harmful pharmaceutical products tested on plants (Pino *et al.*, 2016).

4.1.5 Compost and Champost

Compost (Table 18) has a variety of origins and applications. The most well-known example of this is household compost, called 'GFTe' compost in the Netherlands (meaning vegetable, fruit, garden and food compost, or 'groenten, fruit, tuinafval & etensresten' in Dutch). There is also 'green' compost from municipal green waste, such as hedge trimmings. Lastly, spent mushroom substrate (also known as champost) represents a significant amount of the phosphorus from organic waste streams. Mushroom substrate, sometimes mistakenly called a compost, is a mixture of horse manure, chicken manure, straw, and gypsum or spent lime. After mushroom cultivation has taken place, this waste stream is called 'mushroom manure' or 'champost'.

Table 18 An overview of the various kinds of compost, the total amount of phosphorus they represent, P-content and cost.

P flow	Amount (t P y ⁻¹)	P concentration (g kg ⁻¹)	Cost (€ t P ⁻¹)
Household compost	1 200	2	71 500
Municipal compost	N/A	N/A	N/A
Champost	1 600	2	76 300

Compost contains various contaminants, including dioxins, PFASs, PAHs, and heavy metals (Brändli *et al.*, 2005; Costello and Lee, 2020). Some of these contaminants are found to increase in concentration during the composting process up to a factor of two, including five- and six-ring PAHs, and polychlorinated dibenzo-p-dioxins (Brändli *et al.*, 2005).

4.2 Evaluation of Side- and Waste Streams as source of P

In this section, a number of manures, side- and waste-streams are evaluated as possible source of P. We start by summarising the data presented in the previous section. Then, we evaluate the different side-streams relative to each other and the current properties of P-fertiliser used in soilless growing systems.

Selection of side-streams is based three criteria: (1) the total amount of phosphorus they represent, (2) price per kg P and (3) P concentration. The total quantity of phosphorus should ideally cover the needs of (soilless) greenhouse horticulture. Price before processing is important, also since source costs will add to the cost of the final fertiliser product. Lastly, P concentration is not a hard criterion, but is shown to give an idea about the applicability of a certain P source. The higher the P concentration, the easier P recovery will be and the less material will have to be transported.

Figure 2 visualises these criteria for the different side-streams. Both fertiliser used in soilless greenhouse horticulture and synthetic fertiliser used in arable farming are added for reference.

Looking at each criterion separately, we find the following observations:

1. **Quantity.** All side-streams on their own – except compost and champost – represent enough P. Sewage sludge currently does not represent enough P, but this is because it is nearly all incinerated. Enough sewage sludge is produced in principle, and if it were better to recover P from sludge rather than ash, this would be possible.
2. **Price.** All side-streams except compost and champost are cheaper per t P than soilless fertiliser. Within the side-streams that are affordable enough, ruminant digestate is the most affordable and bone meal is the least affordable.
3. **Concentration.** The differences between side-streams stretch multiple orders of magnitude, with digestates, compost/champost and sludges generally being lower and ashes and bone meal being higher. Only animal carcass ash contains more P than soilless fertiliser.

These observations rule out compost and champost as a meaningful source of phosphorus for greenhouse horticulture. They do not represent enough P and are far too expensive per kg P. Moreover, they already have existing applications, where their added value is based not only on the presence of nutrients, but also organic matter to improve soil properties.

Bone meal is a possible candidate due to its high P concentration. It is cheaper than some soilless fertilisers per kg P, but not cheaper than the median price of phosphorus fertiliser used in soilless systems. It is also successfully applied for gardening products and ceramics production, where the latter market is anticipated to grow. Because of this, using bone meal will not contribute to closing a currently unsustainable nutrient cycle, but rather redirecting an existing closed loop.

The remaining side-streams fall under three categories: manure and digestates, sewage sludge, and ashes. These show a trade-off between P concentration and price. The Pareto front, drawn in light orange, shows

the side-streams for which it is impossible to improve in one criterion without sacrificing the other. For example, poultry manure is not on the Pareto front because other side-streams exist that are both cheaper and have a higher P concentration. The possible side-streams on the Pareto front are ruminant digestate, unincinerated sewage sludge, and sewage sludge ash, poultry manure and animal carcass ash.

Despite this preselection, price and concentration are not the only important factors to look at. The presence of contaminants may make certain side-streams not on the Pareto front easier to work with, and therefore worth the extra cost or lower P concentration. Also the form in which P is present (dissolved or solid, organic or inorganic) will make a difference for recovery. These factors are covered in the next chapter, which looks at technologies, products and contaminant levels.

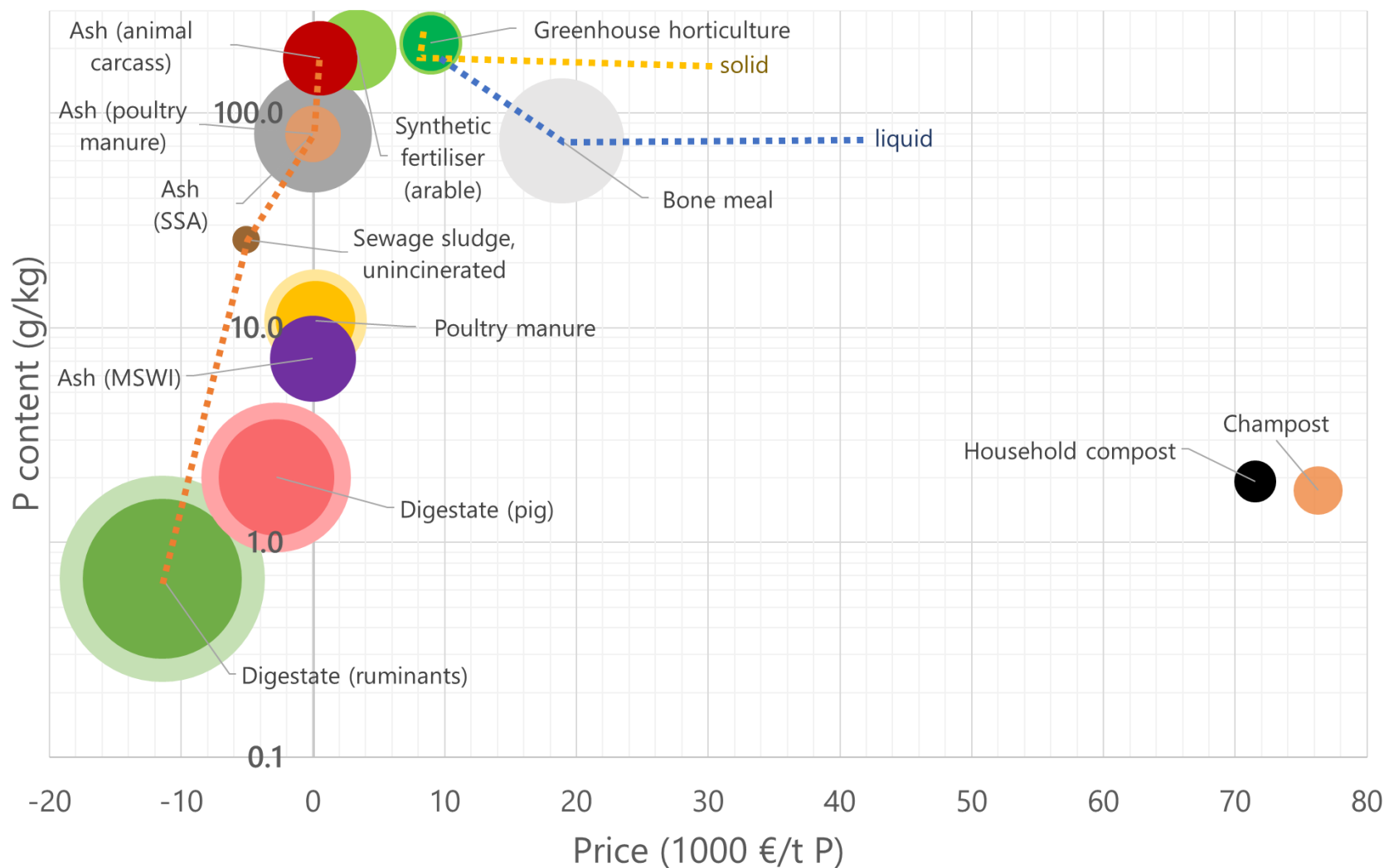


Figure 2 An overview of the major phosphorus-containing residual flows in the Netherlands, excluding composts. The horizontal axis shows the price (1000 € t P⁻¹); the vertical axis (logarithmic) shows the P concentration (g P kg⁻¹). The sizes of the circles correspond to the total amount of P they represent. For manure, the lightly-shaded perimeter represents the organic fraction of phosphorus in the total flow. The dark green part of greenhouse horticulture represents soilless systems. The yellow and blue dotted lines represent the range of possibilities for solid and liquid fertilisers respectively. An orange dotted line is added as a Pareto front.

5 Processes and Products

5.1 Introduction

The recovery of phosphate is increasingly becoming a topic of interest, for reasons explained in Section 1. There are already pilot runs and (almost) full-scale factories applying different processes, as can be concluded from the Phos4You project (Ploteau *et al.*, 2021a; Ploteau *et al.*, 2021b) and information from the recently-held Fosfaat in Perspectief symposium (STOWA, 2023). The aim of this section is to provide an overview of the main phosphate recovery technologies, focused on processes that could be implemented in the short term, and their corresponding pilot/full scale tests, within the context of the requirements of Dutch soilless systems. In particular, where data on contaminants is available, we apply the requirements set out in Section 2.3.2 to various products. Energy consumption is also briefly discussed in Section 5.1.2.

5.1.1 Principles

Phosphate recovery processes use feedstocks that can be classified into two groups: aqueous solutions (dissolved phosphate, e.g. wastewater) and solid materials (e.g. sewage sludge, ashes and manure) (Egle *et al.*, 2016; Witek-Krowiak *et al.*, 2022; Zheng *et al.*, 2023). The appropriate technology depends on the form of phosphate present in the source. Many different brand names exist, each with their own configurations. It would be challenging to include them all in this report, and would obscure the aim of this report as well. Still, most processes make use of the principles described in this section.

In Figure 3, a schematic overview of different recovery technologies from phosphate sources is shown, based on Witek-Krowiak *et al.* (2022) and Zheng *et al.* (2022). When phosphate is dissolved, as in wastewater, it can be recovered or removed by using (a combination of) four principles: precipitation/crystallization, adsorption, biological removal, and membranes. During biological removal, P is accumulated in microorganisms, which eventually ends up in the solid phase (Zheng *et al.*, 2022). Overall, precipitation of P as struvite or vivianite is the most frequently used method to recover dissolved P.

When P is present in solid phase, as is the case with sewage sludge (ash) or manure, wet-chemical processes or thermo-chemical processes are used. Wet-chemical processes, using for example an acid, are the most popular method to extract P from sewage sludge and SSA (Zheng *et al.*, 2023). When P is extracted from solids, again (a combination of) three principles can be used to further purify P: precipitation/crystallization, affinity processes (i.e. adsorption, ion exchange, and extraction), and membranes (Witek-Krowiak *et al.*, 2022).

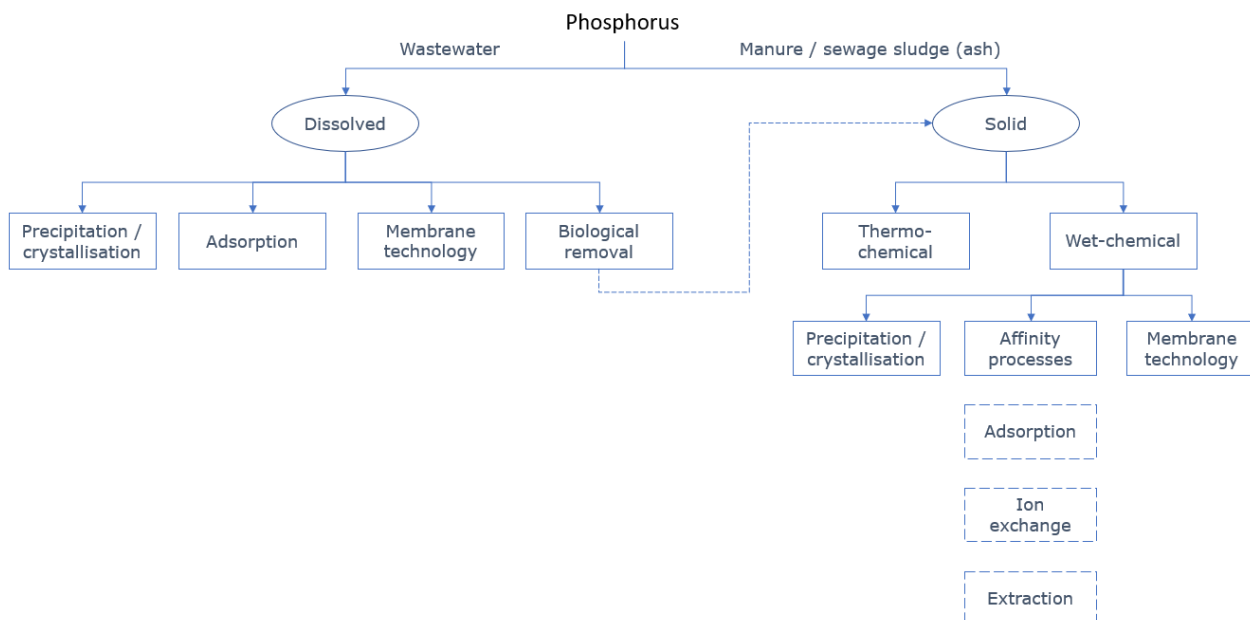


Figure 3 An overview of possible processes to recover phosphorus from different sources, categorised as liquid (where P is dissolved) or solid, based on Witek-Krowiak *et al.* (2022), Zheng *et al.* (2022), and Ploteau *et al.* (2021b).

Following the selection of P-rich flows made in Section 4, the rest of this report focuses on three sources: manure, sewage sludge and ashes, all in which P is present in the solid. The two main methods to recover P from a solid are: (1) wet-chemical treatment to extract phosphate by using, for example, an acid, and (2) thermo-chemical reaction to obtain for example de-mineralized ash or white phosphorus (P_4) (Ploteau *et al.*, 2021a; Van Hooijdonk, 2022; Zheng *et al.*, 2023). White phosphorus is a high-value raw material for flame retardants, lubricant additives, crop protection products, electrolytes for lithium batteries and catalyst ligands (Van Hooijdonk, 2022). Thermo-chemical processes are generally expensive (high operating costs and possible short equipment lifetime due to corrosive conditions); but they show high recovery potential and low consumption of chemicals. Wet-chemical processes require lower energy consumption, are flexible, show high recovery potential, and enable the production of phosphoric acid; but generally consume more chemicals and heavy metals should be removed (Donatello *et al.*, 2010; Gorazda *et al.*, 2017). The low costs and the possibility to make products that could directly be used in the greenhouse horticulture, i.e. P-acid, are two advantages of using wet-chemical processes to recover P from solid sources for application in the greenhouse horticulture.

5.1.2 Operational Energy Consumption

Energy requirements and costs for the processes described in this section are only briefly discussed, as they are not the focus of this project and other papers have investigated this in more detail. Nevertheless, we briefly present operational energy consumption and costs for various processes.

Fahimi *et al.* (2021) and Egle *et al.* (2016) both provide information about the operational energy consumption of different phosphate recovery processes. This information is bundled and summarised in Figure 4. This figure gives an indication on the energy consumption of different principles, though every specific process will be slightly different. The high costs for P recovery from sewage sludge are based on the paper of Egle *et al.* (2016), in which it is mentioned that the costs for extracting P from sewage sludge via wet-chemical leaching is 9-16 € kg P^{-1} . For extracting P from the liquid phase this is 6-10 € kg P^{-1} and from SSA 5-6 € kg P^{-1} .

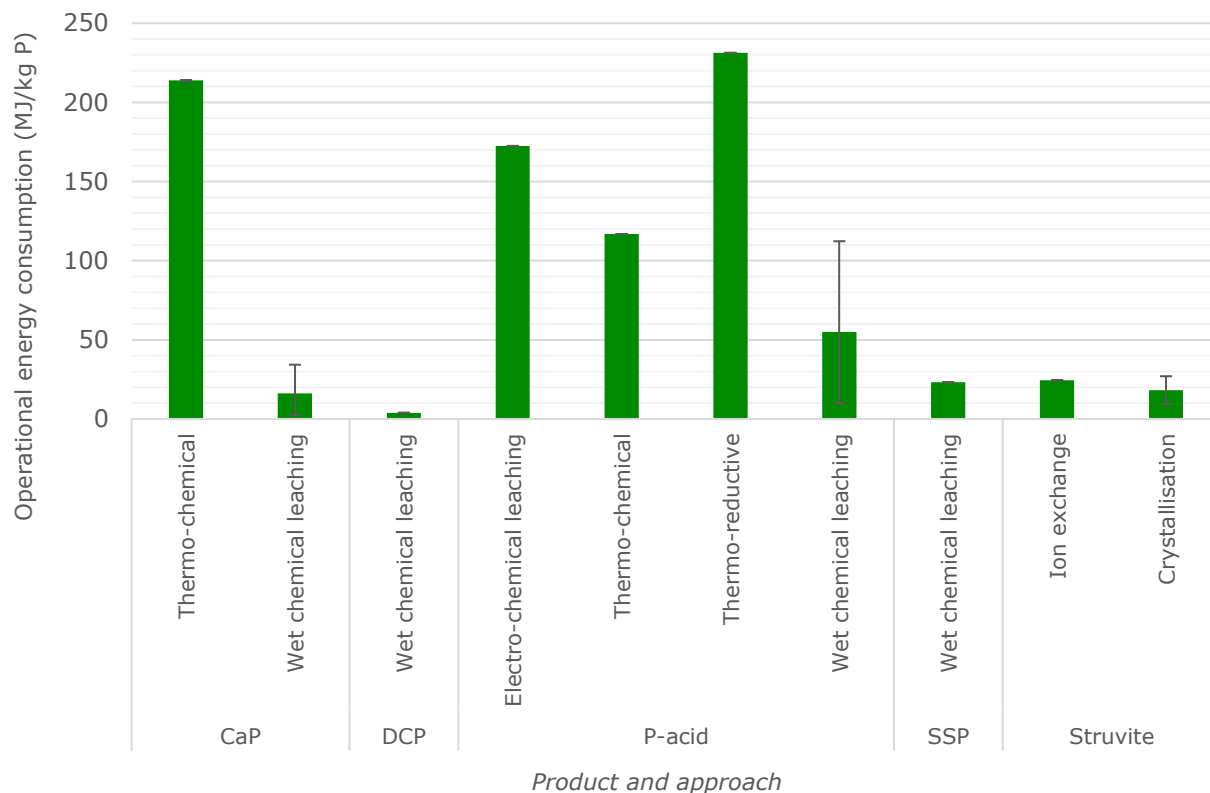


Figure 4 A summary of the operational energy consumption (MJ kg P^{-1}) of various P products and recovery approaches, with data from Egle et al. (2016) and Fahimi et al. (2021). All processes are based on sewage sludge ash, except for those producing struvite, which use sludge as an input. Products: calcium phosphate (CaP), dicalcium phosphate (DCP), phosphoric acid (P-acid), single super phosphate (SSP) and struvite.

5.2 Manure

As discussed in Section 4.1.1, P recovery from manure could make use of manure that currently cannot legally be applied to agricultural land. To our knowledge, there is only one pilot test performed on recovery of phosphate from the solid fraction of manure, which was performed at Groot Zevert Vergisting B.V. (Regelink et al., 2019). Because there is a risk that pathogens and other organic material may be present in the extracted P, additional hygienic measures of the solid fraction are necessary (Regelink et al., 2019).

5.2.1 Precipitates

Groot Zevert Vergisting B.V. tested the RePeat process to separate digested manure in a P-rich solid fraction and liquid fraction, and to obtain a P-fertiliser from the solid fraction. The solid fraction is first hygienised to get rid of pathogens and other propagules. Subsequently, the solid fraction is acidified to extract P, resulting in a P-poor organic soil conditioner and a P-rich solution (Regelink et al., 2019). From this P-rich solution, P is recovered via precipitation in the form of struvite (or calcium-P) by addition of magnesium hydroxide (or calcium hydroxide). From the liquid phase, N-fertiliser can be obtained (Regelink et al., 2019). Overall, from manure, the following products were produced at Groot Zevert Vergisting B.V.: biogas, N-fertiliser, P-fertiliser (struvite), organic soil conditioner (with low P-content), and clean water (Schoumans, 2015). From the RePeat process specifically, the products are P-rich fertiliser and a soil conditioner. A schematic overview of the processes at GrootZevert Vergisting B.V. is presented in Figure 5. This RePeat process was tested at pilot scale at Groot Zevert Vergisting B.V. in 2019, with a capacity of 2 ton solid fraction per hour (Regelink et al., 2019).

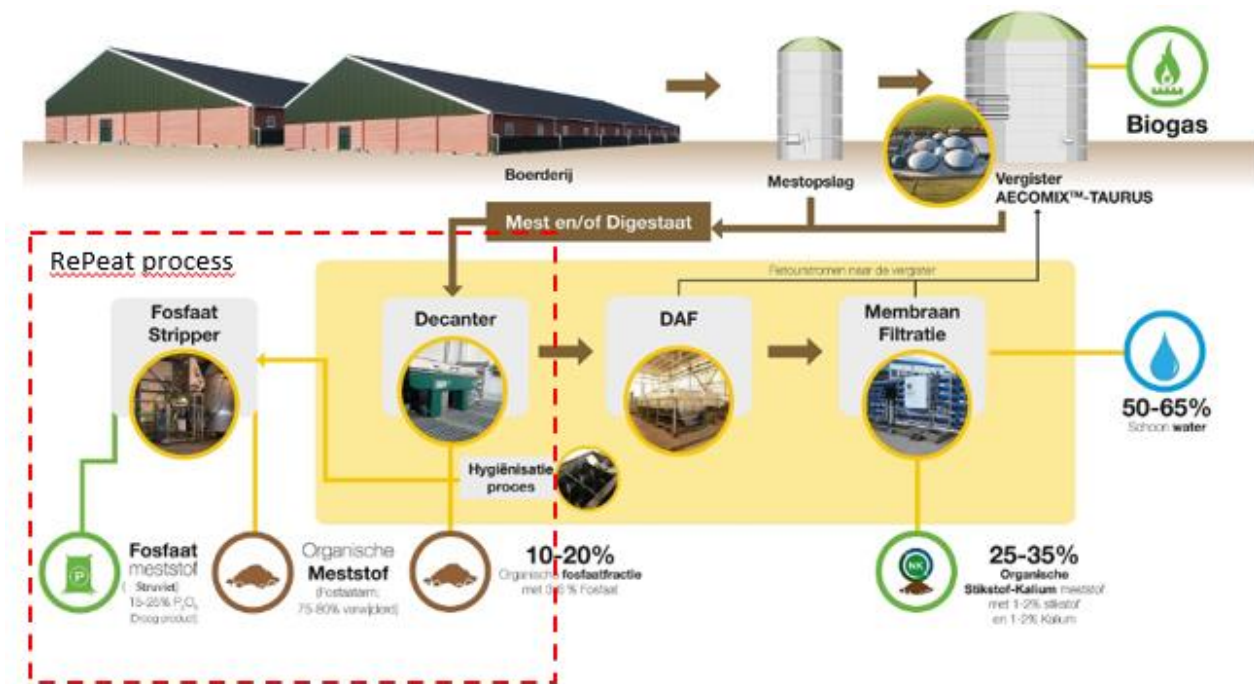


Figure 5 Modified process flow diagram (Schoumans, 2015) of pilot processes at Groot Zevert Vergisting B.V., during which the manure is first digested. Then after digestion, the manure is separated in a liquid and solid fraction. The solid fraction is eventually acidified to extract P, and subsequently struvite is produced. The P-recovery process is outlined with a red box.

An alternative to Groot Zevert's method to recover P from manure was investigated by Schott *et al.* (2023). During this study, raw manure was acidified to liberate P from the manure. Subsequently, calcium was added during anaerobic digestion to recover Ca-P, leading to a P recovery efficiency of 90-95%. Therefore, this could prove to be an interesting method to recover P from manure. For now, it has only been tested at 45 L scale reactors, thus further upscaling is required.

5.3 Sewage Sludge

For sewage sludge, direct recovery of P is a challenge because of a low recovery efficiency (35-70%), high costs, contamination risks with pathogens, heavy metals, organic pollutants and other micro pollutants (Chrispim *et al.*, 2019; Zheng *et al.*, 2022). Witek-Krowiak *et al.* (2022) state the same, and add that organic matter in sewage sludge will dissolve after wet-chemical leaching and subsequently hinder further purification processes. The presence of these contaminants in the final product could also be a risk. This is why in the Phos4You project, only precipitates were investigated (struvite and Ca-P) from sewage sludge after wet-chemical leaching (Ploteau *et al.*, 2021a). Furthermore, the production of de-mineralised ash using thermo-chemical processes was investigated during the Phos4You project. Egle *et al.* (2016) report the same; in this study, different technologies were investigated for the liquid phase (pre-effluent), sewage sludge (SS) and sewage sludge ash (SSA).

Fundamental research is being done to improve P recovery from sludge. For example, as is mentioned in Section 5.3.4, it might be possible to calcinate struvite to remove residual organic content from struvite to facilitate the production of phosphoric acid (Ploteau, 2023). However, current technologies to recover P from sewage sludge focus on crystallisation or thermal metallurgic processes, thus not producing phosphoric acid.

5.3.1 Struvite (and vivianite)

Struvite ($\text{NH}_4\text{MgPO}_4 \cdot 6\text{H}_2\text{O}$) can be obtained from sewage sludge in WWTPs, as is already happening in Amsterdam-West. Sewage sludge is first anaerobically digested. Then, the digested sludge is directed to a

struvite precipitation unit, in which magnesium is added. The sludge is aerated, as a result of which CO₂ is released and the struvite crystallization process is started. Struvite precipitates and is collected from the reactor. Eventually, the crystals are washed (Dir. Duurzame Leefomgeving en Circulaire Economie, 2022). Besides struvite, vivianite can be obtained from sewage sludge, which can be further processed into potassium phosphate as explained in Section 3. In Noord Brabant, for example, Waterschap Brabantse Delta started a project with the aim of producing vivianite from sewage sludge in 2025.

A drawback of struvite precipitation with respect to circularity is its reliance on magnesium salts. Just like phosphorus, magnesium is on the EU's list of critical raw materials. 97% of the EU's magnesium is sourced from China (European Commission, 2023). Moreover, magnesium is given both a higher economic importance and supply chain risk than phosphorus (Blengini *et al.*, 2020). It can therefore be argued that struvite precipitation, whilst closing phosphorus loops, shifts dependence to magnesium instead. That said, alternative (low-cost) magnesium sources have been proposed: magnesite, bittern or seawater (Kumar and Pal, 2015).

An example of a commercial process to recover phosphate as struvite from a liquid stream is Struvia™ (Veolia, 2023). As mentioned in the Phos4You technical report (Ploteau *et al.*, 2021b), the performance of Struvia™ has been investigated in combination with bio-acidification. Bio-acidification is a process during which the slurry is acidified by organic acid that is produced by microorganisms (Regueiro *et al.*, 2022). Without bio-acidification, the P-recovery efficiency was 20% of the total P entering the WWTP. It was found that the best performance of bio-acidification was on undigested sludge combined with a co-substrate (carbon-source). Sludge was bio-acidified, and then separated in a liquid- and solid fraction. The liquid fraction was sent to the Struvia™ unit to produce struvite by the addition of magnesium reactants, the solid fraction was sent to the anaerobic digestion tank (Ploteau *et al.*, 2021b). This process is shown in Figure 6 (Ploteau *et al.*, 2021a). By doing so, at least 50% of the P could be recovered (Ploteau *et al.*, 2021b). Focusing on the recovery of P-products, biogas and organic fertiliser are marked in this study as by-products. It should be noted that these by-products are already produced by current sewage sludge treatment methods.

The Struvia™ process itself is already a commercial process. However, the combination with bio-acidification was new in the Phos4You project, resulting in pilot tests at small waste water treatment plants (Ploteau *et al.*, 2021b).

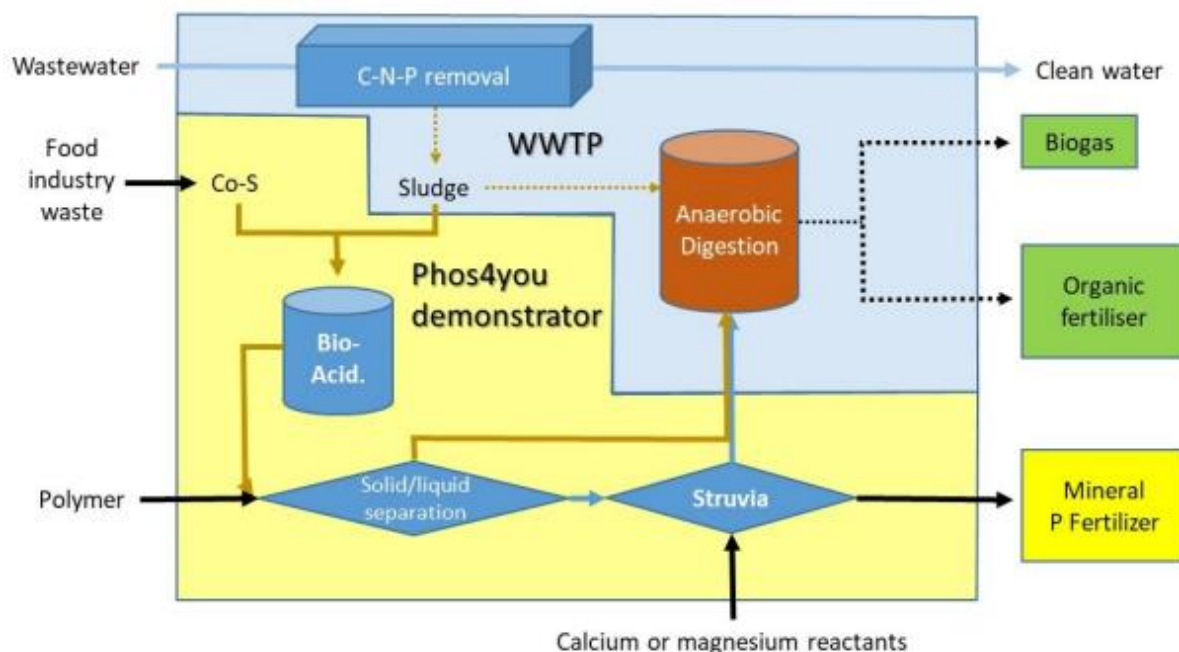


Figure 6 Holistic overview of possible treatment of sewage sludge, by making use of bio-acidification, to enhance P recovery, and struvite formation by Struvia™ (Ploteau *et al.*, 2021a).

5.3.2 Calcium Phosphate

In the PULSE process (Shariff *et al.*, 2023) (Figure 7), de-watered or dried sludge is acidified with hydrochloric acid to extract phosphate from the sludge, with a leaching efficiency of ~70%. Thereafter, metals are removed using a solvent, which should be regenerated by an alkaline solution. Then, the product is further purified via aluminium removal by lime addition. Finally, calcium hydroxide is added to obtain calcium phosphate. It should be noted that also struvite can be produced instead of Ca-P.

The PULSE process was based on the PASCH process and further developed and tested on lab-scale by the University of Liège (Ploteau *et al.*, 2021b). Thereafter, it has been tested at a pilot scale at WWTPs in Belgium, Germany and Scotland, treating about respectively 60, 80, and 70 kg of dried sludge (DM ~95%). Further scale-up studies, and exploration for co-operations, will be performed at the University of Liège (Ploteau *et al.*, 2021b).

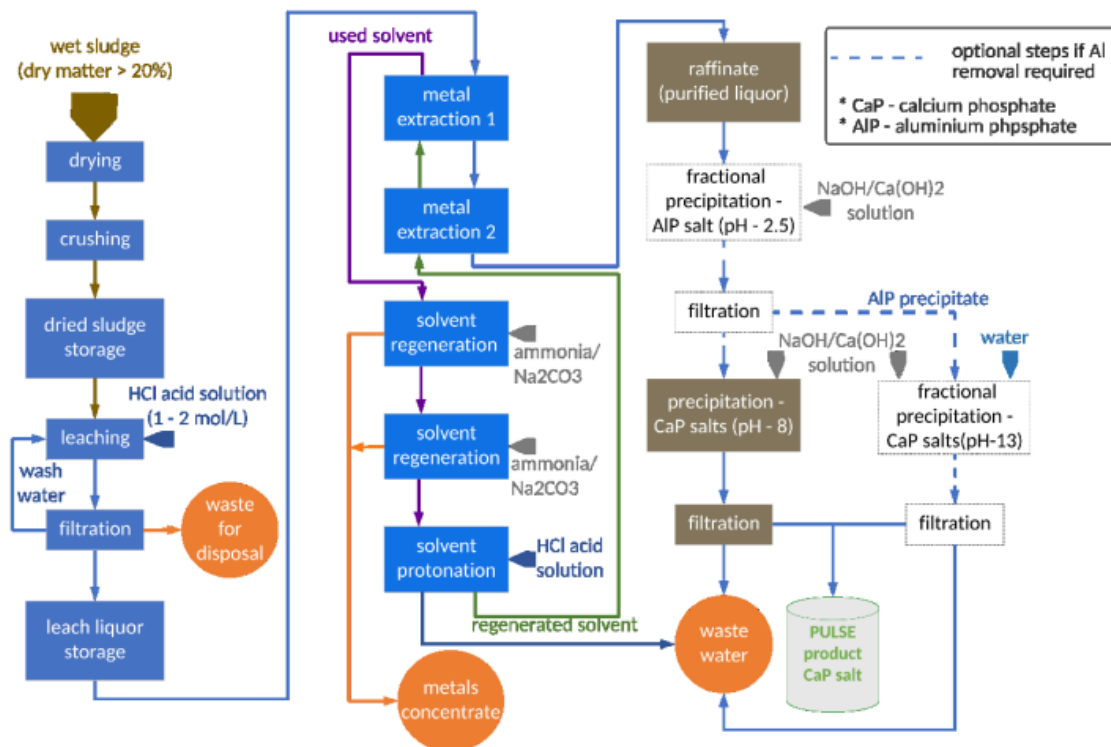


Figure 7 Schematic overview of the PULSE process (Ploteau *et al.*, 2021b). In general terms, P is extracted from sludge using hydrochloric acid. Then, co-dissolved metals are removed using a solvent. Thereafter, aluminium is removed by precipitation with an alkaline solution. Finally, P is precipitated as calcium-P (or struvite) by adding a lime/calcium hydroxide solution.

5.3.2.1 Contaminants

Comparing (Figure 8) the specifications of PULSE to the limits set out in Table 7 (2.3.2.2), there are possible human risks via crop production as several elements may accumulate to toxic levels in fertigation water: aluminium, arsenic, chromium, copper and nickel. Especially aluminium, chromium and copper are multiple times above the limit for fertigation water phytotoxicity/uptake. Lead may also accumulate in fruits.

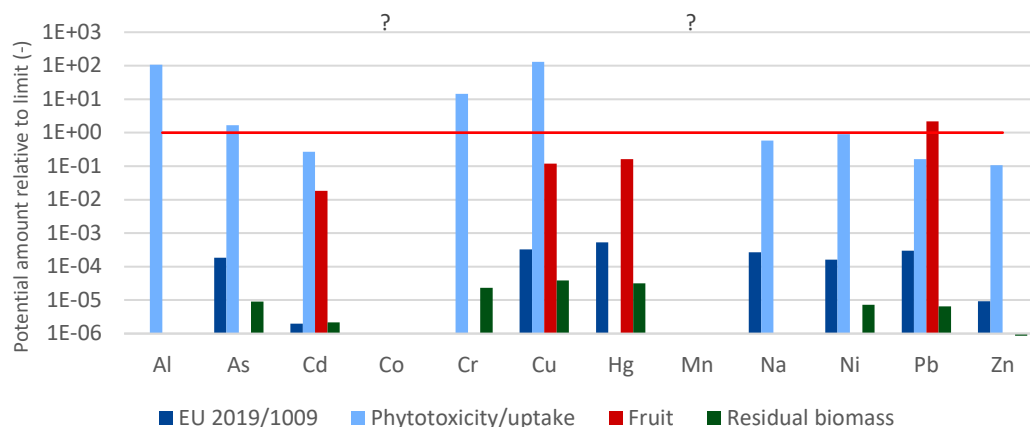


Figure 8 How data (Ploteau et al., 2021b) of the PULSE product compares to the limits set out in Table 7, where the vertical axis (logarithmic scale) represents the actual value divided by the limit, for each of the four limits. Question marks indicate an absence of data where limits do exist.

5.3.3 De-mineralised ash

De-mineralised ash is ash where a significant proportion of the heavy metals have been removed. EuPhoRe is a two stage thermo-chemical process that can be used to obtain de-mineralised ash from sewage sludge. First, the sludge is dried. Then, most of the volatiles and some heavy metals are transferred to the gas phase in the reduction step (650-750°C). Finally, in the oxidation step (up to 1000°C), the remainder of pollutants is removed (Ploteau et al., 2021b), although it should be noted that the removal of POPs such as PFAS and dioxins is unknown (Björklund et al., 2023; Liu, S. et al., 2021). Removal of heavy metals is enhanced by the addition of additives, such as magnesium chloride (MgCl₂). As a result, an ash is obtained with a significant lower heavy metal content than regular sewage sludge ash (Ploteau et al., 2021b). This process is presented in Figure 9.

EuPhoRe is operational at two full scale plants, with capacities of 15 000 – 30 000 t sludge per year, and there are six plants planned (30 000 – 135 000) (EuPhoRe® GmbH, 2023).

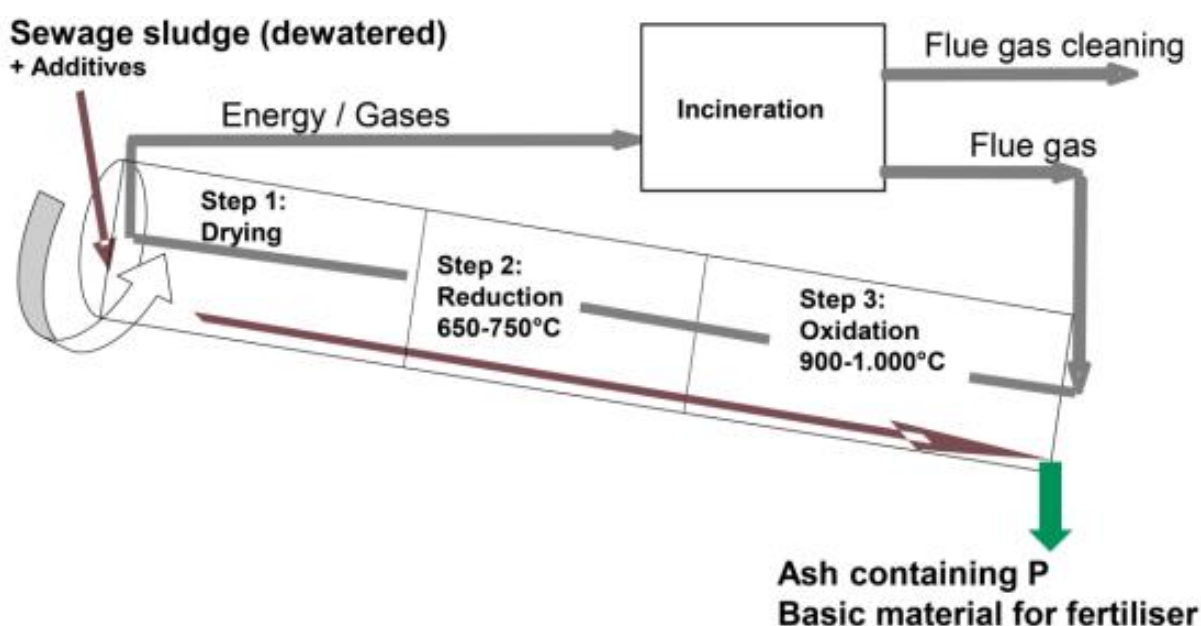


Figure 9 Schematic overview of the EuPhoRe process as presented in the Phos4You technical report (Ploteau et al., 2021b). De-mineralised ash is obtained after thermo-chemical treatment of sewage sludge.

5.3.3.1 Contaminants

The analysed data (Ploteau *et al.*, 2021b) for EuPhoRe demineralised ash shows phytotoxicity risks for nickel (Figure 10). It should be noted that various inorganic contaminants were not measured. Moreover, demineralised ash has a characteristic red colour, which comes from iron oxides (Ploteau *et al.*, 2021b). Data on the iron content was not available, but is crucial for greenhouse horticulture. In soilless greenhouse horticulture, iron is added as a chelate to ensure bioavailability for the crop (Sonneveld *et al.*, 2009), meaning the iron in demineralised ash is likely to be inappropriate at best and harmful at worst.

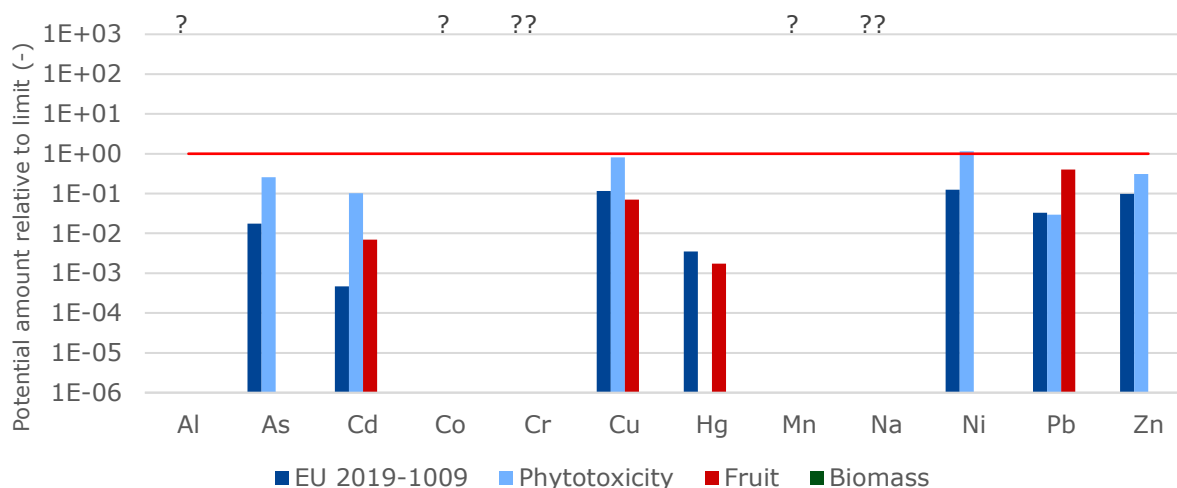


Figure 10 How data (Ploteau *et al.*, 2021b) of EuPhoRe demineralised ash product compares to the limits set out in Table 7, where the vertical axis (logarithmic scale) represents the actual value divided by the limit, for each of the four limits. Question marks indicate an absence of data where limits do exist.

5.3.4 Phosphoric acid

There is currently no information present on the production of phosphoric acid from sewage sludge. The presence of organic matter hinders the purification to phosphoric acid via membranes (Witek-Krowiak *et al.*, 2022). However, there are ideas to calcinate struvite to remove organic matter. Thereafter, P could be extracted again from struvite using an acid, and subsequently further purified to phosphoric acid, as confirmed from email contact with Ploteau (2023). This could facilitate the production of phosphoric acid from struvite, from both sewage sludge and manure.

5.4 Ashes

An advantage of using ashes is the possibility to produce P-acid, as ashes contain no organic matter. For instance, P-recovery efficiency from sewage sludge ash is higher than from sewage sludge, respectively 70-98% and 35-70% (Chripim *et al.*, 2019). This P-acid can directly be used in soilless systems. Several processes and product specifications exist for P-acid, which are discussed in Section 5.4.1. On top of this, ammonium phosphate and calcium phosphates are other possible products, discussed in Sections 5.4.1.5 and 5.4.2 respectively. All processes have been applied to sewage sludge ash, though other ashes could be used, since this could be favourable in terms of contaminants.

5.4.1 Phosphoric acid

Phosphoric acid is produced from ashes using wet chemical leaching, usually with an acid as a leaching agent. An advantage of using sulphuric acid or oxalic acid as leaching agent is the simultaneous formation of insoluble gypsum or calcium oxalate to avoid calcium phosphate formation (Liu, H. *et al.*, 2021). Four specific technologies are discussed in this section: TetraPhos®, RubiPhos, PARFORCE, Phos4Life and SusPhos.

5.4.1.1 TetraPhos® – REMONDIS

An overview of the TetraPhos® process is provided in Figure 11. Phosphate is extracted from sewage sludge ash using phosphoric acid. Thereafter, sulphuric acid is added to precipitate, and then separate, gypsum. Then, ions are separated using ion exchange resins. These resins are regenerated from time to time, resulting in a metal salt solution. Eventually, the remaining phosphoric acid is concentrated using evaporation. Via this process, 85-90% of the phosphate in the sewage sludge ash can be recovered (Ploteau *et al.*, 2021b). In the Phos4You process, TetraPhos® was tested on a pilot/pre-industrial scale in Germany, handling 50-100 kg SSA/h. In 2021, the first TetraPhos® plant started its operation, producing 7 kt P-acid from 20 kt SSA (REMONDIS®, 2023; Sijstermans, 2023b).



Figure 11 Phosphoric acid production scheme of TetraPhos® (Ploteau *et al.*, 2021b). P is extracted from SSA using P-acid. Thereafter, purification steps as filtration, crystallization by addition of sulphuric acid, ion exchange, and evaporation are used to produce P-acid.

5.4.1.2 RubiPhos – TTBS

RubiPhos starts by using sulphuric acid as a leaching agent. During this process, gypsum is immediately separated together with ash-residues. Thereafter, phosphoric acid is further purified using diffusion dialysis and nanofiltration. During diffusion dialysis, only anions can permeate, retaining the cations. Nanofiltration is used as a final purification step to separate the last metal leftovers. Different products can be produced using this process, such as P-acid, struvite and calcium-P. An advantage of this process is that no solvents are used, and less area and chemicals are needed (Ruijter, 2023a).

At the time of writing, RubiPhos was performing pilot trials at HVC's sewage sludge incineration plant in Dordrecht, handling about 10 kg ash per hour (Ruijter, 2023a). Demo trials are planned for 2024-2025, focussing on handling 300 kg ash per hour and on the recovery of metal ions (Ruijter, 2023b).

5.4.1.3 PARFORCE – PARFORCE Engineering & Consulting

The PARFORCE process is similar to the TetraPhos® process. Phosphate is extracted from SSA using an acid (HCl). Then, the acidic solution is separated from the ash residues. This is followed by ion exchange separation and electrodialysis to remove metal ions. To regenerate the ion exchanger, hydrochloric acid or sodium thiosulphate ($\text{Na}_2\text{S}_2\text{O}_3$) are used. A second ion exchange step is used to remove residual aluminium, which can be regenerated using hydrochloric acid or sulphuric acid. Eventually, phosphoric acid is concentrated by evaporation (Figure 12). Phosphorus recovery rates of over 83% were obtained on a pilot scale (REF). Both lab tests and pilot demonstration were performed in the Phos4You project. Further investigation and upscaling to industrial scale was planned in 2023 in Germany, with a capacity of 1 000 t SSA y^{-1} (Ploteau *et al.*, 2021b).

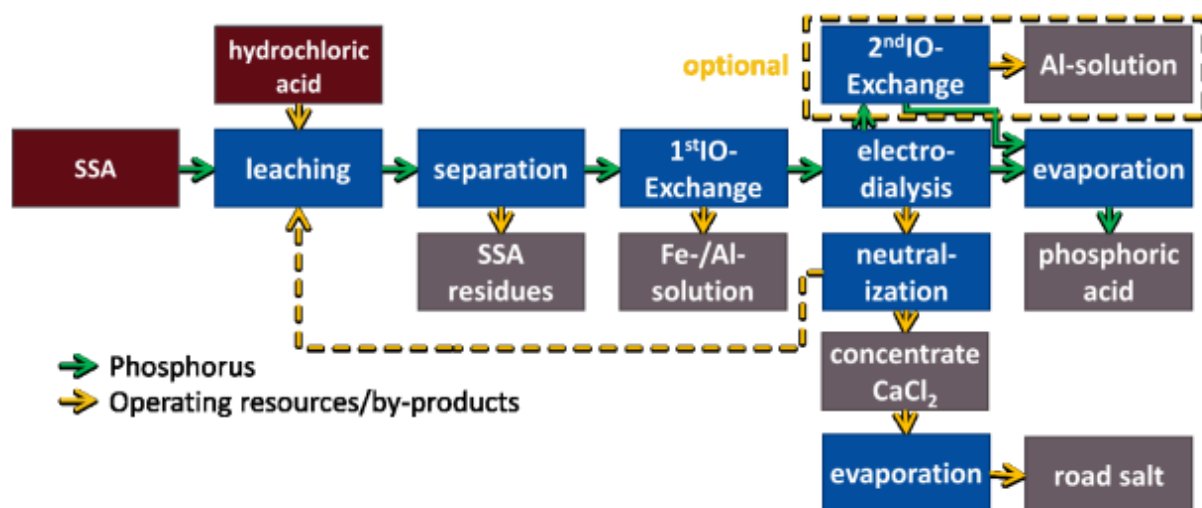


Figure 12 Overview of the PARFORCE process. SSA is acidified with hydrochloric acid to extract P. Thereafter, ash residues are removed via a filter, and the solution is further purified using ion-exchange, electro-dialysis, an option extra ion-exchange step to remove aluminium. Finally, the solution is concentrated by evaporation, and a phosphoric acid solution is obtained (Ploteau et al., 2021b).

5.4.1.4 Phos4Life – Técnicas Reunidas S.A

The Phos4Life process is a relatively new process, with a proof of concept performed in 2020. This process is similar to the TetraPhos® and PARFORCE processes. Phosphate is extracted from SSA using sulphuric acid. Then the acid is separated from ash residues. Further purification of the acid is performed using solvent extraction and re-extraction steps to remove unwanted ions. Finally, phosphoric acid is concentrated using evaporation. This process is shown in Figure 13. Lab-scale tests were performed within the Phos4You project, resulting in leaching efficiencies of 86-96% (Ploteau et al., 2021b). Further investigation is planned, aiming on an industrial scale demonstration in 2027/2028 in Switzerland with a capacity of 30 000 – 40 000 t SSA y⁻¹ (Ploteau et al., 2021b).

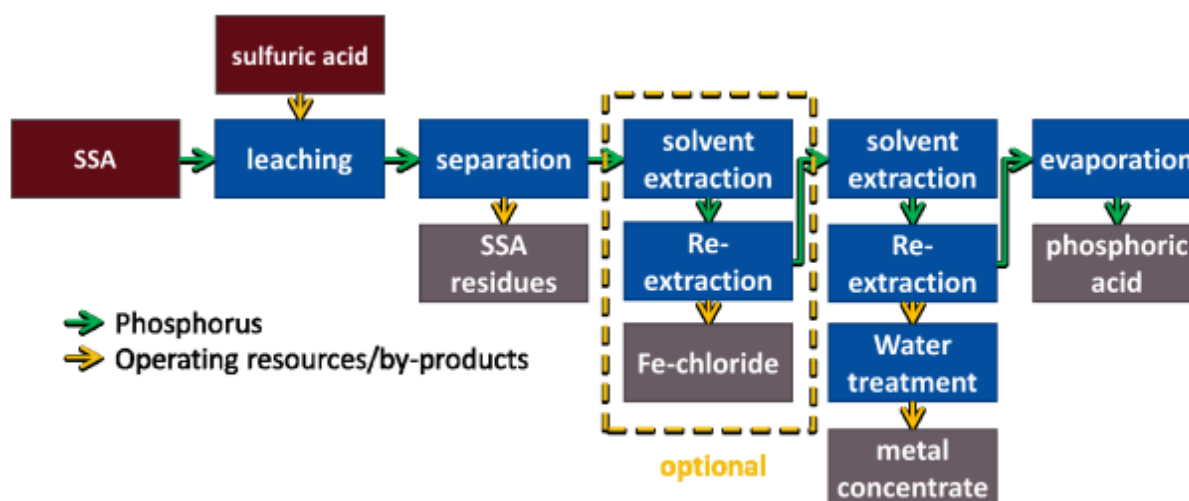


Figure 13 Schematic overview of the Phos4Life process, which starts with the acidification of SSA with sulphuric acid to extract P. Ash residues are removed from the solution, and the solution is further purified using solvent extractions. As final step, evaporation is used to concentrate the phosphoric acid product (Ploteau et al., 2021b).

As an example of the contaminants in a phosphoric acid product from SSA, specifications exist for TetraPhos REPACID®. The specifications are well within the EU 2019/1009 requirements and others specified in this report, with the exception of cadmium and aluminium, which may lead to phytotoxicity risks (Figure 14). That said, although the specification is <1 ppm Cd, Ploteau *et al.* (2021b) showed actual measurements to be lower, from 0.22 ppm Cd down to 0.1 ppm Cd. This shows that phosphoric acid with the right purity can be produced, but that the specification concentrations need to be guaranteed lower for greenhouse horticulture.

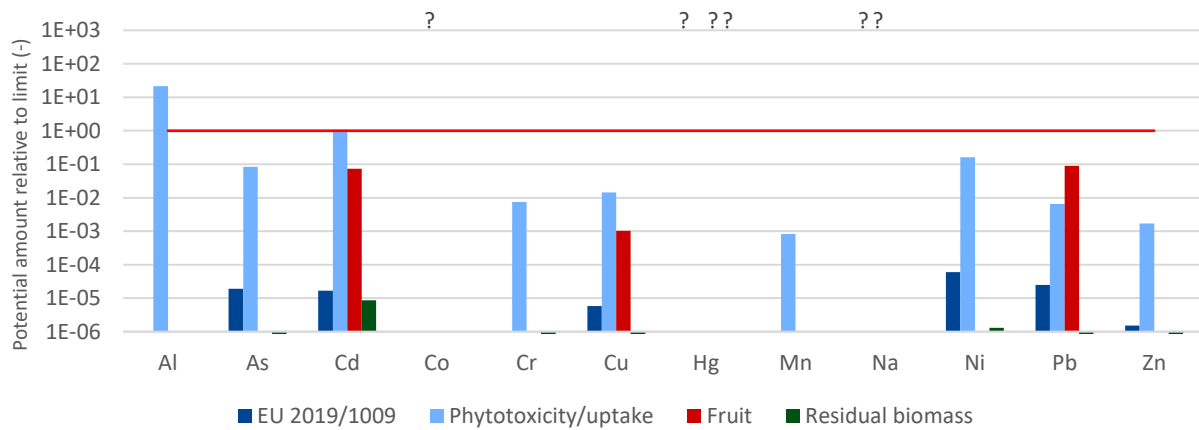


Figure 14 How specifications (Ploteau *et al.*, 2021b) of the TetraPhos REPACID® compares to the limits set out in Table 7, where the vertical axis (logarithmic scale) represents the actual value divided by the limit, for each of the four limits. Question marks indicate an absence of data where limits do exist.

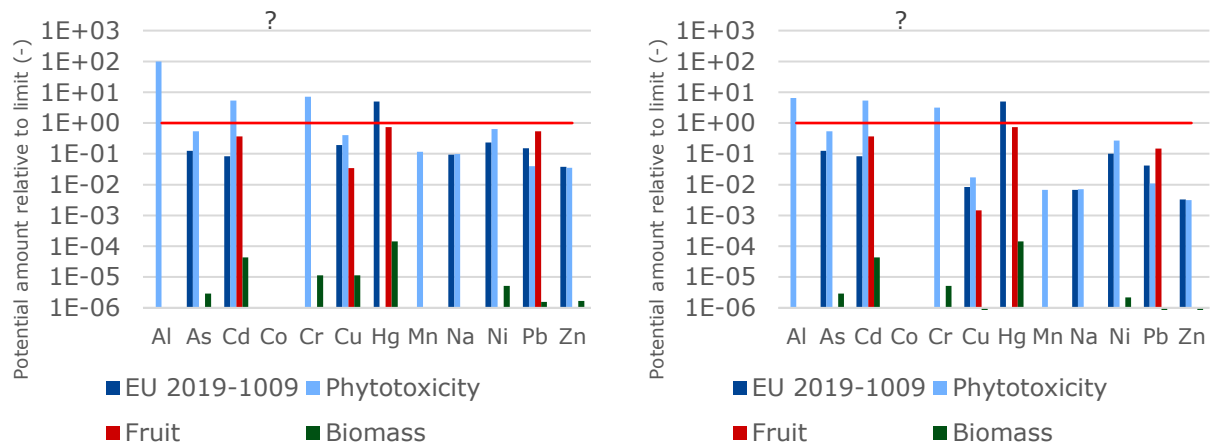


Figure 15 How contaminant data (Ploteau *et al.*, 2021b) of two phosphoric acid batches from the PARFORCE process (left: after 1 IO-exchange; right: after 2 IO-exchanges) compare to the limits set out in Table 7, where the vertical axis (logarithmic scale) represents the actual value divided by the limit, for each of the four limits. Question marks indicate an absence of data where limits do exist.

5.4.1.5 SusPhos

The SusPhos process is developed by the startup SusPhos to produce ammonium phosphate from SSA. SSA is acidified with sulphuric acid to extract P. Then a solvent is used to remove contaminants. The solids are separated from the liquid stream, and subsequently ammonia is added to crystallize ammonium phosphate (Figure 16). The solvent can be reused (De Boer, 2023). At the moment of writing, SusPhos is testing their process at a pilot scale at the sewage sludge incineration plant SNB in Moerdijk. Further testing and scaling up is planned, towards a full scale factory in 2026 in The Netherlands (De Boer, 2023). The company has recently pivoted towards producing phosphoric acid instead of ammonium phosphate.

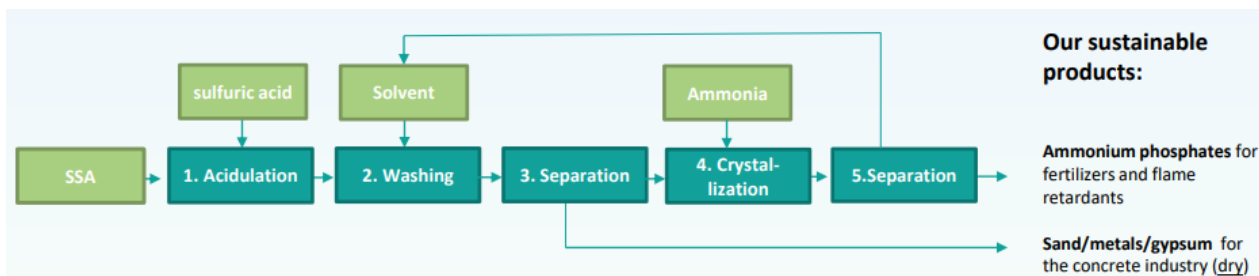


Figure 16 SusPhos produced ammonium phosphates from SSA. First, SSA is acidified using sulphuric acid to solubilise P. Then, a solvent is added for the removal of contaminants. Eventually, ammonia is added to crystallise and collect ammonium phosphates (De Boer, 2023).

5.4.2 Ca-P

Ash2®Phos is a process developed by EasyMining, a Swedish company, and is also based on acidic extraction of phosphate from ashes. Acid (HCl) and lime are needed as input, to produce Ca-P and the by-products iron chloride and sodium aluminate (Sijstermans, 2023a). A schematic overview of this process is provided in Figure 17. A pilot demonstration has been performed, with a first plant planned for 2025-2026 in Germany, based on 30 kt ash/y to produce about 15 kt Ca-P. A second plant is planned for 2026-2027 in Sweden, also for 30 kt ash/y (Sijstermans, 2023a).

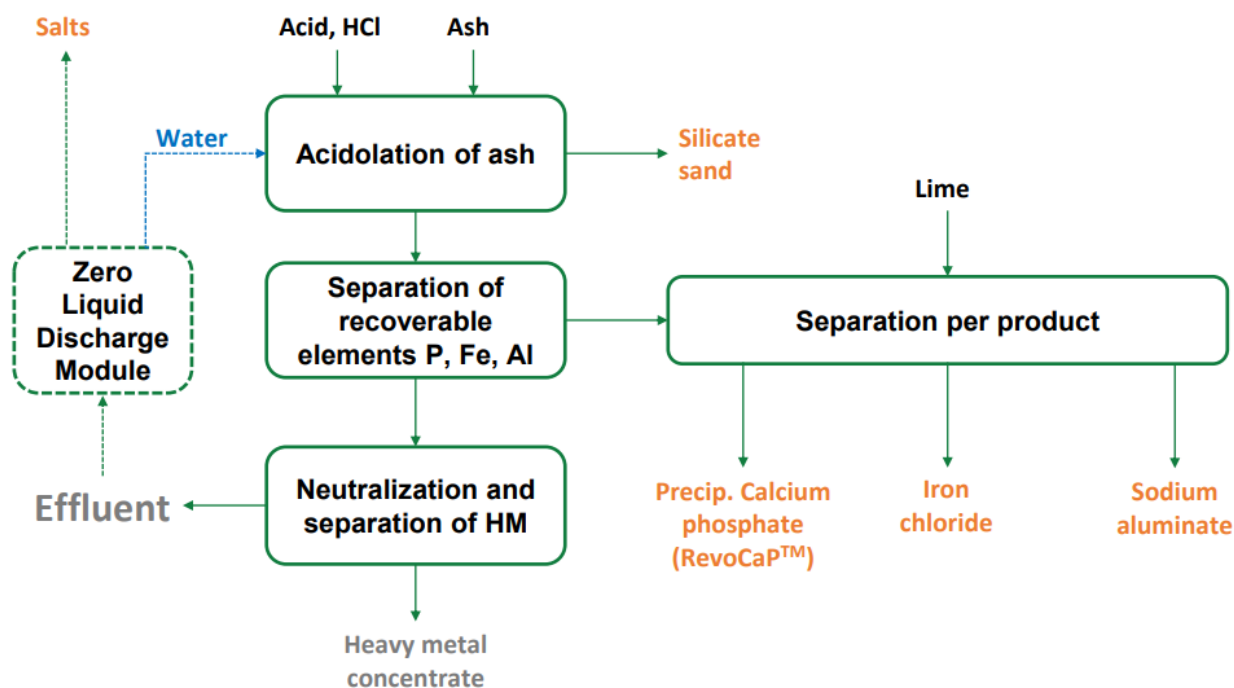


Figure 17 EasyMining's Ash2®Phos process starts with acidification of ash with hydrochloric acid. Then, the recoverable elements are separated. Lime is used to precipitate calcium phosphate and other products as iron chloride and sodium luminate (Sijstermans, 2023a).

EasyMining's Ash2®Phos process produces two products: precipitated calcium phosphate (PCP) and monocalcium phosphate (MCP). Presto Åkerfeldt *et al.* (2023) analysed the inorganic contaminants in both products. Violations of several limits were observed for soilless systems (Figure 18). For PCP, arsenic and cadmium ending up in fertigation water are a possible concern. MCP contains no such violations. Data from Sijstermans (2023a) shows more zinc may be present in tricalcium phosphate from Ash2®Phos than can be taken up by the crop (Figure 19).

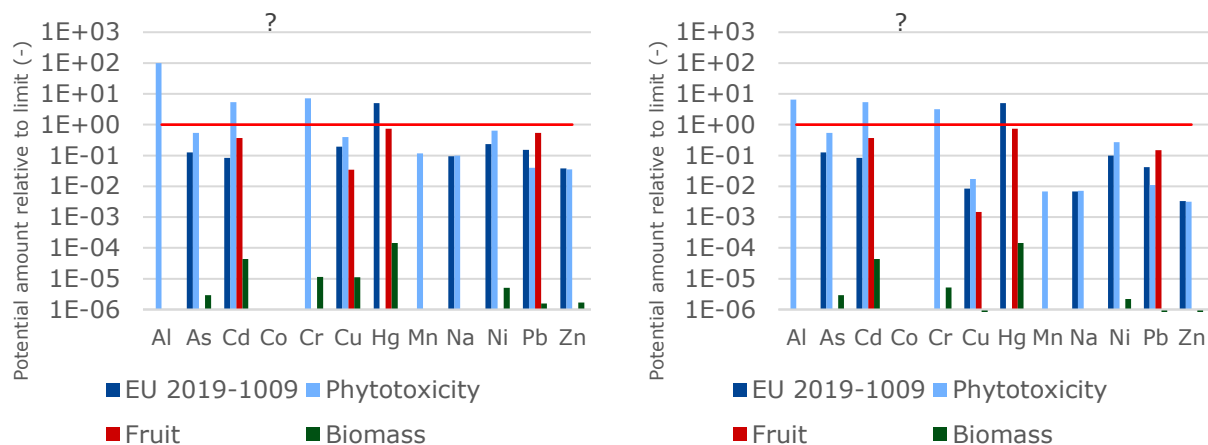


Figure 18 How contaminant data (Presto Åkerfeldt et al., 2023) of two products from the Ash2®Phos process (left: precipitated calcium phosphate; right: monocalcium phosphate) compare to the limits set out in Table 7, where the vertical axis (logarithmic scale) represents the actual value divided by the limit, for each of the four limits. Question marks indicate an absence of data where limits do exist.

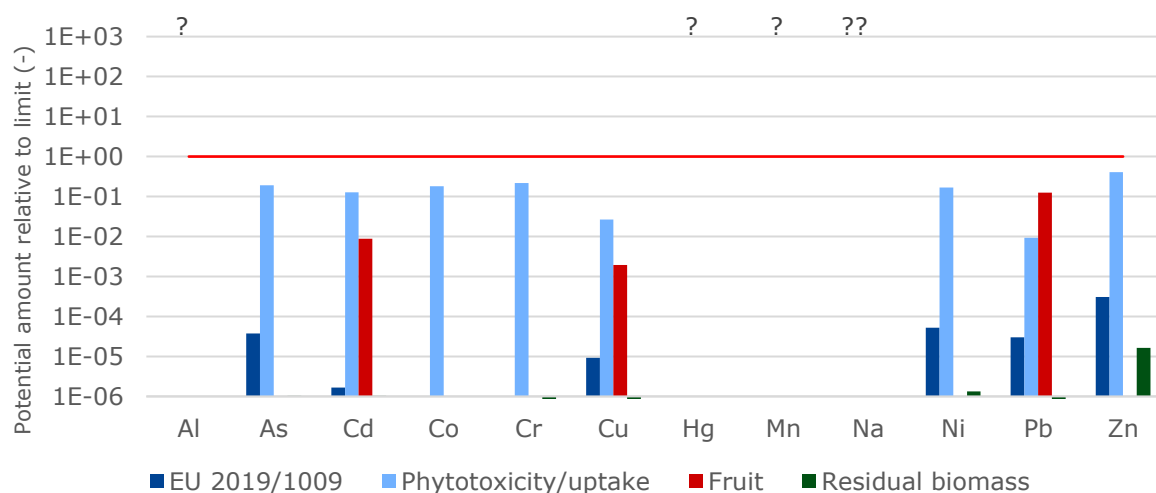


Figure 19 How contaminant data (Sijstermans, 2023a) on Ash2®Phos tricalcium phosphate compares to the limits set out in Table 7, where the vertical axis (logarithmic scale) represents the actual value divided by the limit, for each of the four limits. Question marks indicate an absence of data where limits do exist.

5.5 Summary of processes

In Chapter 5, P-recovery processes for P present in solids were described. The processes were categorised by the feedstocks (manure, sewage sludge, or ashes) needed for each process as a raw material. A summary of these processes, including their main product, other raw materials required, by-products formed is shown in Table 19 below. It should be noted that all the processes use residual streams as feedstock. Therefore, many of the by-products that arise during these processes already existed in the residual streams that were used as a feedstock (though additives and fouled components such as membranes and ion exchangers will be generated too). Nearly all the sewage sludge ash is currently regarded as a waste stream and used as filling material in asphalt or as landfill (Section 4.1.2). The produced by-products of the processes described in this report are, however, split up. For example, the by-products of processes that use sewage sludge ash as feedstock are split up into ash residues, metal solution(s) and possibly gypsum.

The usability of the main products, as described in Sections 3 and 5, depends on the product type and its purity. The processes TetraPhos®, RubiPhos, PARFORCE, Phos4Life and SusPhos all produce products (P-acid or ammonium-P) that could directly be used in the current soilless systems if purity of the product meets the contaminants limits. The other processes that are discussed make products that could not be directly used in the current soilless systems. The products of these processes could be used in a separate acidic 'C-tank', as described in Section 3.

Table 19 An overview of all the processes that are described in Chapter 5. For each process, the required feedstock, the main product that is formed, required raw materials, and formed by-products are shown.

Feedstock	Process	Main P-product	Raw materials	By-products
Manure	RePeat	Struvite (or Ca-P)	(Sulphuric) acid, magnesium hydroxide (or calcium hydroxide)	Soil conditioner
Sewage sludge	Struvia	Struvite	Magnesium chloride, co-substrate (carbon source) for bio-acidification	Biogas, organic fertiliser
	PULSE	Ca-P (or magnesium-P)	(Hydrochloric) acid, metal extraction solvent, alkaline solution (regeneration extractant solvent), lime, calcium hydroxide	Sludge residue, metal solution, waste water
	EuPhoRe	De-mineralised ash	Magnesium chloride	Flue gas
Sewage sludge ash	TetraPhos	P-acid	(P-acid), sulphuric acid, hydrochloric acid	Ash residue, gypsum, metal solution
	RubiPhos	P-acid, Ca-P, or struvite	Sulphuric acid	Ash residue, metal solution
	PARFORCE	P-acid	hydrochloric acid, regeneration agents (1 st ion-exchange: HCl or Na ₂ S ₂ O ₃ ; 2 nd ion-exchange: HCl or H ₂ SO ₄)	Ash residue, metal solutions, road salt
	Phos4Life	P-acid	Sulphuric acid, extraction solvent, regeneration chemicals	Ash residue, metal solution, (Fe-chloride)
	SusPhos	Ammonium-P	Sulphuric acid, solvent, ammonia	Sand, metals, gypsum
	Ash2@Phos	Ca-P	Hydrochloric acid, lime	Ash residue, metal solution, salts, iron chloride, sodium aluminate

As can be seen in Table 19, an acid, mainly sulphuric acid or hydrochloric acid, is necessary to dissolve P in most processes. Then, P can be precipitated or purified to P-acid. The alkaline compounds magnesium hydroxide or calcium hydroxide are used to precipitate struvite or Ca-P. When ion exchange or extraction processes are used, regeneration chemicals are required to recover P. Common by-products during P-acid production processes are sludge (ash) residues and metal solutions. Data about the amount of raw materials required, or the purity of the by-products is not investigated in this project. Besides the purity of the final product, important factors for processes are chemical consumption, energy requirements, wastewater generation, and value-addition to by-products (Ploteau *et al.*, 2021a).

6 Conclusions and Recommendations

In this section, we discuss the methodology used and the implications of our results, leading to conclusions and recommendations. This is done for the three main aspects of circular P covered in this report: side-streams, processes and products, and contaminants.

6.1 Side-Streams

Our analysis of different phosphorus containing side-streams indicated that circular P for soilless greenhouse horticulture is amply available. We compared side-streams in terms of concentration and price, shows a clear Pareto front between these two criteria (Figure 2). The side-streams on this Pareto front are manures, sewage sludge, sewage sludge ash and animal carcass ash.

It is possible that side-streams not on the Pareto front may be worth choosing after all. This is because there are other factors which are not taken into account in this inventory, notably removal of possible chemical contaminants and compliance with legislation. Similarly, the processes discussed were only investigated on sewage sludge, sewage sludge ash and manure. We expect recovery efficiencies and costs to be similar if other side-streams are used. Additionally, using these other sources could potentially come with advantages, for example: (1) lower contaminant levels and (2) other nutrients being present. This is therefore just a first step in identifying possible suitable side-streams for circular, soluble fertilisers in horticulture. Ashes have the highest P concentrations and have the advantage that pathogens and organic pollutants have been removed, though some contaminants may survive or emerge as a result of the combustion process (POPs and PFASs).

P recovered from side-streams is currently more costly than mined P. Still, soilless systems could be a more accessible market for these products, since per kg P, fertilisers used in soilless systems command a higher price than those used in arable farming. Egle *et al.* (2016) found recovered P to cost between 6 and 16 € kg P⁻¹, depending on the side-stream and technology used. This range, applied to the side-streams in Figure 2, shows that it should be possible to produce a cost-effective recovered P fertiliser for soilless systems. Compared to the synthetic fertilisers used in arable farming, the extra price premium of fertilisers for soilless systems may allow some margin to absorb extra costs that come with the recovery process. Until the cost of recovery and processing decreases (also compared to mineral phosphorus), companies providing recovered P from these side-streams may want to look to greenhouse horticulture as a first market to allow for scaling up.

6.2 Processes and Products

The side-streams mapped fall into three categories: ashes, manure (including digestates) and sewage sludge. This report presents an overview of several P-recovery processes for these categories. Many of these processes are not currently operational on an industrial scale. Many of them are also based on the same technological principles. Due to a lack of exact (proprietary) specifications, it is difficult to assess which technology will be best performing in terms of cost, scale, process reliability, etc. Still, the analysis gives a broad impression of which recovery processes will be available in the foreseeable future. We were not able to universally compare different technologies (e.g. wet chemical leaching vs precipitation) for all factors. However, we could use specific implementations/'brands' of a technology as case studies.

Many of the discussed processes produce phosphoric acid, which is directly soluble and applicable in current soilless systems. Our simulations in OLI Studio show the applicability of otherwise insoluble P fertilisers (from precipitation) in soilless systems. These precipitates need to be dissolved in acid and kept separate from

other nutrient solutions, which requires an extra tank (the 'C' tank, in addition to existing 'A'- and 'B' tanks). P products recovered through precipitation may require less operational energy to produce than phosphoric acid, meaning that precipitated P is likely to be the affordable option. Using P precipitates may come with a learning curve associated with the extra tank. Furthermore, applying these products in an acidified C tank likely requires the addition of lye to compensate for the extra acidity, which is comparatively expensive. As the market for recovered phosphorus fertilisers will develop, new approaches may emerge, requiring cost-benefit analyses.

This amount of nitric acid added to the C tank corresponds to an equilibrium state and is a minimum amount required. In practice, more acid may need to be added. This was found experimentally by Carreras-Sempere *et al.* (2021) for struvite. Should this extra amount significantly affect nutrient recipes, sulphuric acid may be appropriate in addition to nitric acid. This should be verified experimentally on a pilot scale.

A sustainability analysis was not the focus of this study, but one element is worth mentioning. The different P-recovery processes showed a large variation in energy consumption. For example, the production of phosphoric acid (most typically through wet chemical leaching) is more energy-intensive than producing precipitates such as struvite and calcium phosphate. Struvite precipitation has the lowest energy consumption, but it relies on magnesium, which, like P, is on the EU Critical Raw Materials list. All other processes do not rely on such critical raw materials, though the inputs for exact proprietary implementations are not fully known.

6.3 Contaminants

In this study, the risk of pathogens and organic contaminants was briefly discussed. Risks from inorganic contaminants – i.e. heavy metals (toxic to humans and plants) and other metals, such as sodium (which can decrease yields) – were quantified based on product measurements and specifications presented in literature. For our safety assessment, we assumed 100% accumulation of certain contaminants over the entire crop cycle. We analysed 100% accumulation in 3 different places: 1) irrigation water (leading to phytotoxicity risks), 2) fruit (leading to food safety risks), and 3) residual biomass (leading to composting risks). Because of such extreme assumptions, there is an asymmetry: if we assess a contaminant to be below the limit, it is almost certain there will be no toxicity risk for plants or non-compliance with food safety regulation. If a contaminant exceeds a limit, this still does not necessarily mean toxicity problems are not possible, but additional monitoring over an extended time period is recommended.

In recycled nutrient systems, the risks of inorganic contaminants like heavy metals accumulating in fertigation water to levels harmful for the crop were found the most likely to occur. Accumulation in fruits may exceed legal limits for human consumption only in the case of mercury (Hg) and lead (Pb). If used for composting, the accumulation of contaminants in residual biomass is not of concern. The four recycled P products assessed for this report showed a lack of data for some other metals, in particular sodium. This is relevant since plants grown in soilless systems are more susceptible to sodium than in soil based systems. Therefore, for manufacturers of circular fertilisers, carefully monitoring contaminant and sodium levels is crucial to build trust and acceptance from growers.

The criteria for limits used in this study are far stricter than the EU Fertilising Products Regulation. These can help fertiliser manufacturers determine appropriate specifications for their products and the processes used to make them, as is being done in projects such as KNAP ('closing the cycle of nutrients from waste- and process water'). When switching to using recycled P, monitoring is recommended to confirm that concentrations contaminants or salts in fertigation water do not affect plant health and that concentrations in harvested crop products do not exceed safety limits for human consumption. Moreover, the user of recycled P should be aware that contaminants may still enter the system from other sources. Examples include sodium and boron (Guidi *et al.*, 2011) from irrigation water, but also zinc from galvanised components (Voogt and Sonneveld, 1997). Combined with circular fertilisers, this could still lead to limits being exceeded.

6.4 Overview of Options

In Table 20, we summarise our findings for the various options for circular phosphorus for soilless greenhouse horticulture. For all criteria except one (i.e. operational energy consumption), phosphoric acid, recovered from ashes using wet chemical processes is the best product. It is also soluble in water and directly applicable to greenhouse horticulture. That said, it is 2-3 times as energy-intensive as products recovered by precipitation, such as struvite and Ca-P. This means it will likely be more expensive than phosphorus fertiliser currently used (in fact, phosphoric acid currently is the most expensive phosphorus fertiliser for greenhouse horticulture).

Should the cost of phosphoric acid be prohibitive, struvite or Ca-P can be applied, but this will require changes to the fertigation system by installing a third 'C' tank and adjusting the fertigation strategy accordingly. The applicability in the fertigation system of Ca-P depends on whether the Ca-P salt is tri-, di-, or monocalcium phosphate, with the former requiring the most acid and the latter requiring the least.

Table 20 also shows a number of possible inorganic contaminant violations for recovered P, compared to the criteria derived in this report. For inorganic contaminants where measurements were unavailable, this was counted as a half violation.

Table 20 An overview of how various circular P routes for greenhouse horticulture compare for various criteria, in terms of the three main P-rich by-products and the various fertiliser products currently recovered. For 'directly applicable', 1 means the product can be applied with no changes to the fertigation system, 2 means the 'C' tank is required but with no further adjustments to nutrients, and 3 means significant changes to the nutrient supply strategy are needed, on top of a C tank.

	Manure		SS		SSA		
	Ca-P	Struvite	Struvite	Ca-P	H ₃ PO ₄	NH ₄ -P	Ca-P
Operational energy consumption (MJ/kg P)	2-30	9-30	9-30	2-30	10-110	?	2-30
Critical raw materials (#)	0	1	1	0	0	0	0
Inorganic contaminant violations (#)	?	?	?	7	1	?	2.5
Pathogen risk (Y/N)	Y	Y	Y	Y	N	N	N
Pharmaceuticals (Y/N)	Y	Y	Y	Y	N	N	N
Directly applicable (1-3)	2	2	2	2	1	1	2

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Appendix A

Table A.1 A summary of the sources used for data on manure, presented in Section 4.1.1.

Quantity	Unit	Sources
P content	g P kg ⁻¹	Witek-Krowiak <i>et al.</i> (2022) Termorshuizen and Postma (2021) Veeken <i>et al.</i> (2017) CBGV (2016)
Digestate costs	€ t ⁻¹	Personal contact with Freek Lemmen, Looop (2023)
Chicken manure costs	€ t ⁻¹	Van Loon (2022)
Total production of manure	t y ⁻¹	Gollenbeek (2022)
Proportion of chicken manure incinerated	%	Gollenbeek (2022)
P potential of manure	t P y ⁻¹	Centraal Bureau voor de Statistiek (2022b) Smit <i>et al.</i> (2010)

Table A.2 A summary of the sources used for data on sewage sludge, presented in Section 4.1.2.

Quantity	Unit	Sources
P potential, WWTPs & unincinerated sludge	t P y ⁻¹	Centraal Bureau voor de Statistiek (2023c)
P potential, WWTPs	t P y ⁻¹	Gerritsen <i>et al.</i> (2021)
P content	g P kg ⁻¹	Witek-Krowiak <i>et al.</i> (2022)
Costs	€ t ⁻¹	Personal contact with Josien de Ruiter (HVC) (2023) Brummelaar (2020) Perree (2020)

Table A.3 A summary of the sources used for data on ashes, presented in Section 4.1.3.

Quantity	Unit	Sources
SSA potential	t P y ⁻¹	Centraal Bureau voor de Statistiek (2023c) Gerritsen <i>et al.</i> (2021)
Proportion of municipal waste incinerated	%	Rijkswaterstaat (2023)
P content, SSA	g P kg ⁻¹	Witek-Krowiak <i>et al.</i> (2022)
P content, MSWI ash	g P kg ⁻¹	Kalmykova and Fedje (2013) Loginova <i>et al.</i> (2019)
P content, bone ashes	g P kg ⁻¹	Witek-Krowiak <i>et al.</i> (2022)
P content, chicken manure ash	g P kg ⁻¹	Ehlert (2017)
P content, animal carcass ash	g P kg ⁻¹	Cohen (2009)

Table A.4 A summary of the sources used for data on bone meal, presented in Section 4.1.4.

Quantity	Unit	Sources
P content	g P kg ⁻¹	Hermie (2023) Royal Brinkman (2023) Witek-Krowiak <i>et al.</i> (2022) Termorshuizen and Postma (2021) Jeng <i>et al.</i> (2007)
Costs	€ kg ⁻¹	Hermie (2023) Royal Brinkman (2023)

Table A.5 A summary of the sources used for data on compost, presented in Section 4.1.5.

Quantity	Unit	Sources
Total potential	t P y ⁻¹	Smit <i>et al.</i> (2010)
P content, all except champost	g P kg ⁻¹	Termorshuizen and Postma (2021) Veeken <i>et al.</i> (2017) CBGV (2016)
P content, all	g P kg ⁻¹	Regelink <i>et al.</i> (2019)
P content, champost	g P kg ⁻¹	Baars and Sonnenberg (2015) Groeigoed (2023a) Van Herwijnen (2023)
Costs, champost	€ t ⁻¹	Groeigoed (2023a)
Costs, compost	€ t ⁻¹	Groeigoed (2023a) Groeigoed (2023b)

Table A.6 A summary of the sources used for data on synthetic fertilisers.

Product	Quantity	Unit	Sources
Total amount, arable farming	4350	t P y ⁻¹	Smit <i>et al.</i> (2015) - Van der Lugt (2022)
Costs, synthetic fertiliser for arable farming	3300	€ t P ⁻¹	Van der Meulen (2023)
Concentration, synthetic fertiliser for arable farming	197	g P kg ⁻¹	Triple super phosphate

Appendix B

Table B.1 Kilograms of antibiotics used by livestock sector and for all livestock sectors combined and sold in 2021, by pharmacotherapeutic group (Bonten and Van Geijlswijk, 2022).

Pharmacotherapeutic group	Kilograms used, according to delivery records									
	Broiler farming sector	Turkeys farming sector	Pig farming sector	Dairy cattle farming sector	Veal farming sector	Non-dairy farming sector	Rabbit farming sector		Other poultry farming subsectors	All livestock sectors combined
1 st -choice antibiotics	2,396	1,051	40,991	9,875	36,864	4,676	284	1,799	97,937	114,902
% 1 st -choice of total	41.6%	81.7%	78.8%	81.0%	83.4%	80.9%	75.5%	82.4%	79.1%	79.4%
Amphenicols	0	0	1503	472	1923	362	0	0	4261	4,315
Fixed-dose combinations	0	0	0	0	0	0	0	0	0	662
Macrolides/Lincosamides	342	461	3691	586	12715	1310	57	723	19886	20,744
Other	0	0	0	0	0	0	52	0	52	648
Penicillins	453	58	4148	3387	421	290	0	517	9273	9,527
Pleuromutilins	0	11	172	0	0	0	37	13	232	183
Tetracyclines	541	489	19254	1578	17112	2178	20	297	41469	46,857
Trimethoprim/Sulphonamides	1	33	12224	3852	4693	536	119	249	22765	31,967
2 nd -choice antibiotics	3,350	221	10,115	2,300	7,316	1,093	92	194	24,681	29,607
% 2 nd -choice of total	58.2%	17.2%	19.4%	18.9%	16.5%	18.9%	24.5%	8.9%	19.9%	19.5%
Aminoglycosides	8	0	131	292	260	35	88	0	814	1,089
Aminopenicillins	2,734	209	9,290	1,340	5,961	766	0	91	20391	22,842
1 st and 2 nd gen. cephalosporins	0	0	0	16	0	0	0	0	16	451
Quinolones	579	12	66	3	1073	149	4	102	1986	1,938
Fixed-dose combinations	29	0	535	645	8	140	0	0	1357	1,992
long-acting macrolides	0	0	94	5	14	4	0	0	117	130
3 rd -choice antibiotics	6	14	905	20	28	12	0	190	1175	1,287
% 3 rd -choice of total	0.1%	1.1%	1.7%	0.2%	0.1%	0.2%	0.0%	8.7%	0.9%	0.9%
3 rd and 4 th -gen. cephalosporins	0	0	0	0	0	0	0	0	0	5
Fluoroquinolones	4	14	0	16	5	2	0	12	52	116
Polymyxins	3	0	905	4	23	11	0	179	1,123	1,166
Overall	5,752	1,286	52,011	12,195	44,208	5,782	377	2,184	123,793	144,630

Table B.2 Maximum level of organic contaminants in fertilisers according to the Dutch Uitvoeringsbesluit Meststoffenwet. The values are given in mg contaminant/kg of component which adds nutritional value to the fertiliser. The maximum value that applies depends on the component that reaches a threshold value (amount) first during the application. These are for example: 80 kg of phosphate (P_2O_5), 100 kg of nitrogen, 150 kg of potassium (K_2O), 400 kg of neutralising value or 3000 kg of organic matter.

	Phosphate P_2O_5	Nitrogen N	Potassium K_2O	Neutralising value	Organic matter
Σ PCDD/PCDF	0.019	0.015	0.010	0.0038	0.00051
α-HCH	310	248	165	62	8.3
β-HCH	12	9.6	6.4	2.4	0.32
γ-HCH (lindane)	1.2	0.96	0.64	0.24	0.032
HCB	31	31.2	20.8	7.8	1.0
Aldrin	7	5.6	3.7	1.4	0.2
Dieldrin	7	5.6	3.7	1.4	0.2
Σ aldrin/dieldrin	7	5.6	3.7	1.4	0.2
Endrin	7	5.6	3.7	1.4	0.2
Isodrin	7	5.6	3.7	1.4	0.2
Σ endrin/isodrin	7	5.6	3.7	1.4	0.2
Σ DDT + DDD + DDE	23	18.4	12.3	4.6	0.6
PCB-28	18.5	14.8	9.9	3.7	0.48
PCB-52	18.5	14.8	9.9	3.7	0.48
PCB-101	75	60	40	15	2
PCB-118	75	60	40	15	2
PCB-138	75	60	40	15	2
PCB-153	75	60	40	15	2
PCB-180	75	60	40	15	2
Σ 6-PCB (excl. PCB-118)	375	300	200	75	10
Naphthalene	600	480	320	120	16
Phenanthrene	750	600	400	150	20
Anthracene	600	480	320	120	16
Fluoranthene	185	148	98	37	4.9
Benzo(a)anthracene	230	184	123	46	6.1
Chrysene	230	184	123	46	6.1
Benzo(k)fluoranthene	270	216	144	54	7.2
Benzo(a)pyrene	290	232	155	58	7.7
Benzo(g,h,i)perylene	210	168	112	42	5.6
Indeno(1,2,3-c,d)pyrene	235	188	125	47	6.3
Σ 10-PAH	11500	9200	6133	2300	307
Mineral oil	935000	748000	498668	187000	24933

Appendix C

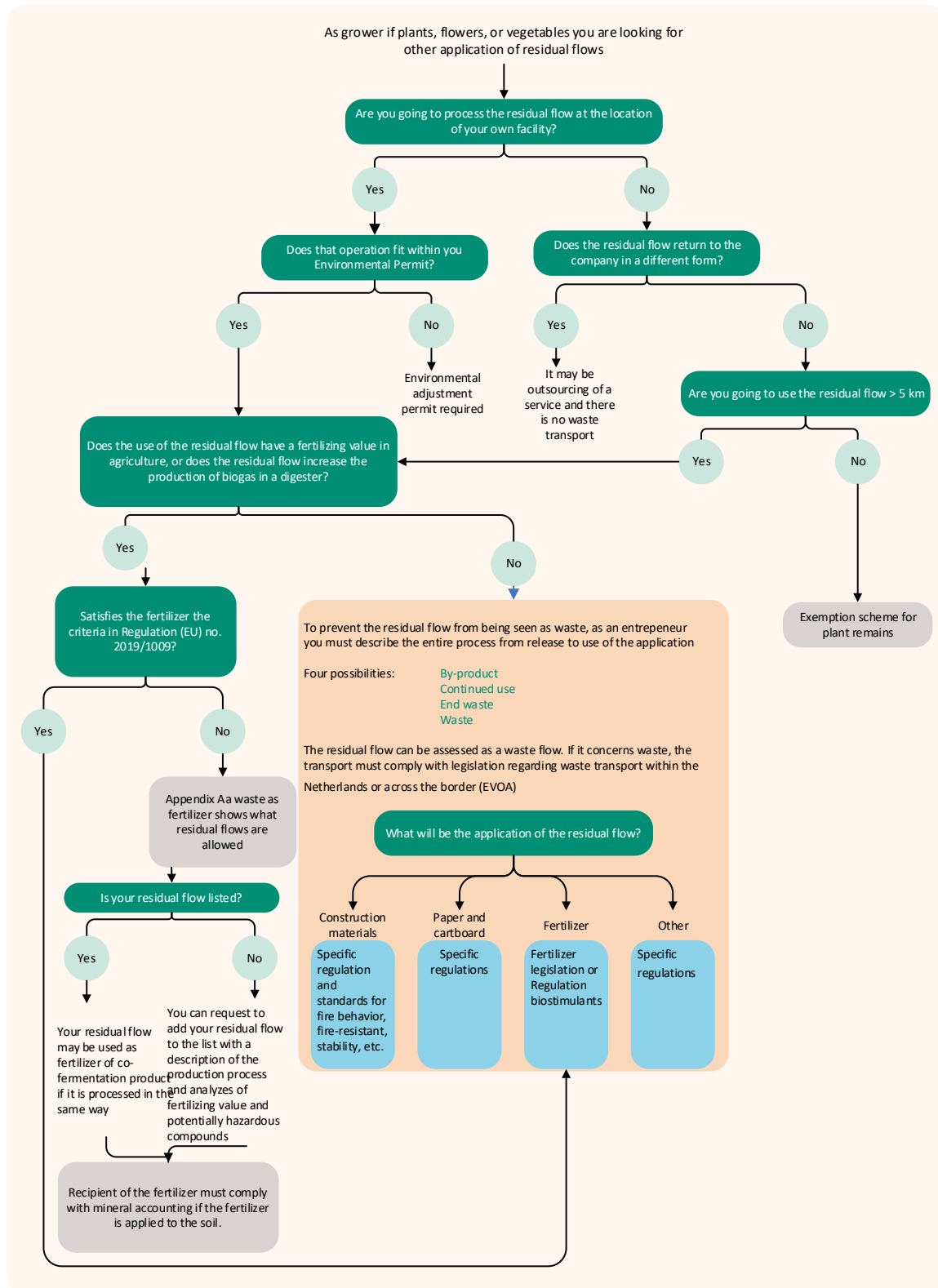


Figure C.1 Flowchart on legislation relevant to circular fertilisers. Translated from *Op weg naar een circulaire tuinbouw: wet- en regelgeving reststromen*, A.D. Hartkamp en P.T. Oei. Stichting Innovatie Glastuinbouw Nederland. 2020.

https://www.innovatieglastuinbouw.nl/media/registered_downloads/s/signaal_folder_reststromen_v2_interactief.pdf.

Appendix D

Equation 1 translates the end requirements of biomass (edible and residual) into maximum levels of contaminant Z in the fertiliser containing nutrient X:

$$[Z]_X = \frac{[Z]_{bio} \cdot m_{bio}}{m_X} \quad (1)$$

In which:

$[Z]_X$	Maximum concentration of contaminant Z in fertiliser for nutrient X	$\text{mg}_Z \text{ g}_X^{-1}$
$[Z]_{bio}$	Maximum concentration of contaminant Z in biomass	$\text{mg}_Z \text{ kg}^{-1}$
m_{bio}	Total mass of biomass produced during the crop cycle	kg m^{-2}
m_X	Total mass of nutrient X given during the crop cycle	$\text{g}_X \text{ m}^{-2}$

A similar approach can be used to translate the maximum concentrations of a contaminant in fertigation water. Assuming 100% accumulation, a contaminant's concentration can be determined by using the volume of water typically present in the fertigation system (Equation 2), not to be confused with the total volume of water given to the crop in a year.

$$[Z]_X = \frac{[Z]_{water} \cdot V_{water}}{m_X} \quad (2)$$

In which:

$[Z]_X$	Maximum concentration of contaminant Z in fertiliser for nutrient X	$\text{mg}_Z \text{ g}_X^{-1}$
$[Z]_{water}$	Maximum concentration of contaminant Z in fertigation water	$\text{mg}_Z \text{ l}^{-1}$
V_{water}	Volume of water typically present in the fertigation system at any given time	l m^{-2}
m_X	Total mass of nutrient X given during the crop cycle	$\text{g}_X \text{ m}^{-2}$

Despite assuming 100% accumulation in fertigation water, some contaminants may not accumulate. For example, copper and zinc are essential plant micronutrients. Micronutrients will only accumulate if present above certain concentrations, beyond which the crop cannot take them up fast enough (De Kreij *et al.*, 2003). Although sodium is not a plant nutrient, its behaviour is similar: crops can take up sodium in low concentrations, but it tends to accumulate in recirculating systems as there is always sodium present from other sources. This is why sodium content must be as low as possible, or not exceeding a concentration that can be taken up by the crop. This input concentration can be calculated for various crops, using the equation in Box 13.7 of Stanghellini *et al.* (2019), which is about maximum sodium concentration:

$$\frac{C_{U\infty}}{C_{in}} = \frac{100}{p} \quad (3)$$

In which:

$C_{U\infty}$	Maximum sodium concentration	mmol l^{-1}
C_{in}	Input concentration of sodium initially entering the fertigation system	mmol l^{-1}
p	Proportion of sodium taken up as a percentage of the rootzone concentration	%

In the case of contaminants that are micronutrients (and sodium), the ratio of contaminant Z to nutrient X should be the same as the ratio of their desired concentrations in fertigation water at the start of the crop cycle (Equation 4):

$$[Z]_X = \frac{[Z]_{water}}{[X]_{water}} \cdot 10^3 \quad (4)$$

In which:

$[Z]_X$	Maximum concentration of contaminant Z in fertiliser for nutrient X	$\text{mg}_Z \text{ g}_X^{-1}$
$[Z]_{water}$	Maximum concentration of contaminant Z in fertigation water	$\text{mg}_Z \text{ l}^{-1}$
$[X]_{water}$	Concentration of nutrient X in applied fertigation water	$\text{mg}_X \text{ l}^{-1}$

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of nature to
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