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EXECUTIVE SUMMARY

Constant supply of plant-available nutrients such as nitrogen and phosphorus, either as manufactured fertilisers or animal manure, to agricultural soils is needed for global food security. Increased recycling of nutrients back to agriculture from organic waste streams is necessary for increased rural-urban sustainability. Anaerobic digestion of sewage sludge and agricultural wastes is widely applied to stabilize the substrate and also capture its energetic value via biogas production. The liquid phase of anaerobic digestate is a concentrated source of nutrients to which nutrient recovery technologies can be applied. Two such promising technologies that could increase nutrient recycling from e.g. wastewater and thereby contribute to environmental amelioration are struvite precipitation and ammonia stripping. By combining anaerobic digestion and nutrient recovery technologies on the digestate, a treatment process that provides both renewable energy and plant nutrients is achieved. This review examined the effectiveness of ecotechnologies for the recovery and reuse of nitrogen and phosphorus from anaerobic digestate with the aim of reducing the impact of waste on the environment.

We searched for academic and grey literature published after 2013. English language searches were performed in 5 bibliographic databases. Google Scholar searches were done in English and Swedish. Eligibility screening was conducted at two levels: title and abstract and full text. Included eligible studies were subject to a critical appraisal that assessed external and internal study validity. We extracted information on study characteristics, intervention, comparators, effect modifiers, and measured outcomes. Data synthesis included narrative synthesis of each study of sufficient validity. We performed quantitative synthesis on a subset of studies.

The review included 36 studies on the effectiveness of struvite precipitation, 7 studies on ammonia stripping, 22 studies on struvite as fertilizer and 1 study on ammonium sulphate as fertilizer. Both pH and the ratio of Mg to limiting reactant were found to have a clear influence on the effectiveness of struvite precipitation process (and nutrient removal rates). The response to pH was found to be nonlinear, resembling a bell curve with a maximum around pH 9.5. Mg to limiting reactant ratio was found to have a positive effect on removal up to a ratio as high as 4 to 1. However, dosing Mg in excess may be expensive, and it should be noted that relatively high efficiencies were achieved at a ratio as low as 1 to 1 as well. Although the effects of pH and Mg to limiting reactant ratio were clear, the model developed could not accurately predict removal based on these two parameters alone. Studies on ammonia stripping were relatively heterogeneous and different digested substrates were included, including wastewater sludge and different types of manure. Due to a small size of the evidence base, and the heterogeneity between studies, no conclusions are presented regarding the influence of different process parameters on the outcome of ammonia stripping. We provide suggestions as of which data to report in future studies. In conclusion, when performed under the right conditions, both struvite precipitation and ammonia stripping seem to be effective techniques for the recovery of nutrients from the liquid phase of digestate. In a wastewater treatment setting, both methods could be applied to the liquid phase of digested wastewater sludge in order to produce a fertilizer product that contains less contaminants than the sludge itself.

Struvite, most frequently recycled from agricultural waste or sewage sludge, seems to be a suitable phosphorus fertilizer for a diverse set of crops, predominantly including cereals and grasses. Treatment of soils with struvite usually results in comparable yields and P uptake by plants as treatment with conventional, mineral P fertilizers. Even if the direct effect of struvite on yield and P uptake is slightly worse than that of mineral fertilizer, which was the case in some studies, the benefit of using struvite



should offset this difference in the long-term. Future research should focus on field-scale validation of struvite effectiveness, as well as on assessment of its long-term effects.

The evidence base for ammonium sulphate was too small to formulate implications for policy and management on this fertilizer.

1 INTRODUCTION

The degradation of the Baltic Sea is an ongoing problem, despite investments in measures to reduce external inputs of pollutants and nutrients from both diffuse and point sources. Available technological and management measures to curb eutrophication and pollution flows to the sea have not been adapted adequately to the contexts in which they are being applied. Furthermore, measures are often designed based on single objectives, thereby limiting opportunities for multiple benefits.

In addition, there is a general sense that measures to address the deterioration of the Baltic ecosystem are primarily technologically-driven and lacking broader stakeholder acceptance – the "experts" who define these measures have little engagement with industry, investors, civil society and authorities. This problem is magnified by governance and management, taking place in sectoral silos with poor coordination across sectors.

As a result, research shows that regional institutional diversity is presently a barrier to transboundary cooperation in the Baltic Sea Region (BSR) and that actions to achieve national environmental targets can compromise environmental goals in the BSR [0]. The regional dimension of environmental degradation in the BSR has historically received weaker recognition in policy development and implementation locally. However, developments in recent years suggest a new trend with growing investments in environmental protection supporting social, economic, and territorial cohesion.

The BSR is an environmentally, politically and economically significant region and like other regions globally, its rapid growth needs to be reconciled with the challenges of sustainable development in a global setting that demands unprecedented reductions in GHG emissions. This poses a truly wicked problem exacerbated by the fact that many of the challenges in the BSR will also magnify in a changing climate. In order to navigate the uncertainties and controversies associated with a transformation towards a good marine environment, BONUS RETURN will enact an innovative trans disciplinary approach for identifying and piloting systemic eco-technologies.

The focus is on eco-technologies that generate co-benefits within other interlinked sectors, and which can be adapted according to geophysical and institutional contexts. More specifically, emphasis is placed on eco-technologies that reconcile the reduction of present and future eutrophication in marine environments with the regional challenges of policy coherence, food security, energy security, and the provision of ecosystem services.

1.1 Project Objectives

The **overall** aim of BONUS RETURN is to improve the adaptation and adoption of eco-technologies in the Baltic Sea Region for maximum efficiency and increased co-benefits.

The **specific objectives** of the project can be divided into six categories presented below. These categories are interlinked but for the purpose of providing a step-wise description, the following overview of each category proves useful. BONUS RETURN is:

1) Supporting innovation and market uptake of eco-technologies by:

- Contributing to the application and adaptation of eco-technologies in the BSR through an evidence-based review (systematic map) of the developments within this field.

- Contributing to the development of emerging eco-technologies that have the capacity to turn nutrients and carbon into benefits (e.g. bio-energy, fertilizers), by providing an encompassing framework and platform for rigorous testing and analysis.
- Developing decision support systems for sustainable eco-technologies in the BSR.
- Contributing to better assessment of eco-technology efficiency via integrated and participatory modelling in three catchment areas in Finland, Sweden and Poland.
- Contributing to methodological innovation on application and adaptation of eco-technologies.
- 2) Reducing knowledge gaps on policy performance, enabling/constraining factors, and costs and benefits of eco-technologies by:
- Assessing the broader socio-cultural drivers linked to eco-technologies from a historical perspective.
- Identifying the main gaps in the policy environment constraining the implementation of emerging eco-technologies in the catchments around the Baltic Sea.
- Informing policy through science on what works where and under which conditions through an evidence-based review (systematic map and systematic reviews) of eco-technologies and the regional economic and institutional structures in which these technologies evolve.

3) Providing a framework for improved systematic stakeholder involvement by:

- Developing methods for improved stakeholder engagement in water management through participatory approaches in the case study areas in Sweden, Finland and Poland.
- Enacting a co-enquiry process with stakeholders into opportunities for innovations in ecotechnologies capable of transforming nutrients and pollutants into benefits for multiple sectors at different scales.
- Bringing stakeholder values into eco-technology choices to demonstrate needs for adaptation to local contexts and ways for eco-technologies to efficiently contribute to local and regional developments.
- Disseminating results and facilitating the exchange of learning experiences, first within the three catchment areas, and secondly across a larger network of municipalities in the BSR.
- Establishing new cooperative networks at case study sites and empowering existing regional networks by providing information, co-organizing events and engaging in dialogues.

4) Supporting commercialization of eco-technologies by:

- Identifying market and institutional opportunities for eco-technologies that (may) contribute to resource recovery and reuse of nutrients, micro-pollutants and micro-plastics (e.g. renewable energy).
- Identifying potential constraints and opportunities for integration and implementation of ecotechnologies using economical models.
- Facilitating the transfer of eco-technologies contributing to win-win solutions to multiple and interlinked challenges in the BSR.
- Linking producers of eco-technologies (small and medium enterprises SMEs), to users (municipalities) by providing interactive platforms of knowledge exchange where both producers and users have access to BONUS RETURN's envisaged outputs, existing networks, and established methodologies and services.
- 5) Establishing a user-driven knowledge platform and improved technology-user interface by:
- Developing an open-access database that maps out existing research and implementation of eco-technologies in the BSR. This database will be intuitive, mapped out in an interactive geographical information system (GIS) platform, and easily managed so that practitioners, scientists and policy-makers can incorporate it in their practices.



- Developing methodologies that enact the scaling of a systemic mix of eco-technological interventions within the highly diverse contexts that make up the BSR and allows for a deeply interactive medium of knowledge.

1.2 Project Structure

BONUS RETURN is structured around six Work Packages that will be implemented in three river basins: The Vantaanjoki river basin in Finland, the Słupia river basin in Poland, and Fyrisån river basin in Sweden.

Work Package 1: Coordination, management, communication and dissemination.

Work Package 2: Integrated Evidence-based review of eco-technologies.

Work Package 3: Sustainability Analyses.

Work Package 4: Environmental Modelling.

Work Package 5: Implementation Support for Eco-technologies.

Work Package 6: Innovative Methods in Stakeholder Engagement.

1.3 Deliverable context and objective

The current deliverable (Del. No. 2.7) is part of WP 2. The objectives of WP 2 are to systematically collate scientific research of existing and emerging eco-technologies, as well as of the economic models and policy instruments that support the implementation and development of these technologies in the BSR countries.

This deliverable synthesises available evidence on the effectiveness of two ecotechnologies for recovery of nitrogen and phosphorus. Furthermore, it investigates the effectiveness of reuse products of these ecotechnologies, struvite and ammonium sulphate, as fertilisers.

1.4 Outline of the report

This report is structured into the following sub-sections:

- 2.1 Background
- 2.2 Objective of the review
- 2.3 Methods
- 2.4 Review findings
- 2.5 Review conclusions



2 EFFECTIVENESS OF ECOTECHNOLOGIES FOR RECOVERY OF NITROGEN AND PHOSPHORUS FROM ANAEROBIC DIGESTATE AND THEIR REUSE AS FERTILISERS: A SYSTEMATIC REVIEW

2.1 Background

Soil fertility and global food security depend on constant supply of plant-available nutrients, such as nitrogen (N) and phosphorus (P), either in the form of manufactured fertilisers or animal manure, to agricultural soils [0]. But there is a thin line between the optimum amount and timing of N fertiliser and its over-supply. N over-supply can quickly lead to serious environmental problems, since excess N is typically lost from the soil system, contaminating bodies of water [2, 3]. In contrast to N, which is effectively unlimited in its atmospheric form, high-quality rock reserves of P are limited and expected to deplete within a few hundred years [0]. P does not leach through the soil but modern farming practices lead to excessive soil accumulation of P, and as such it is exposed to the risk of erosion into water courses while being sorbed to soil particles [4].

Environmental problems associated with N and P use are particularly pressing in the Baltic Sea Region (BSR), since excessive inputs of nutrients coming from the surrounding land are among the primary causes of the Baltic Sea eutrophication [5]. N and P entering water bodies that originate from the application of synthetic fertilisers or farmyard manure are regarded as *non-point source pollution*. As of 2014, non-point sources in the BSR contributed 46.5% and 35.7% of total N and P riverine loads, respectively [6]. *Point source pollutants* are another significant N and P loads to BSR, and mostly originate from wastewater treatment plants (WWTPs). The contribution of point source pollutants to total riverine load entering the Baltic Sea in 2014 was considerably smaller than that of non-point sources, i.e. 11.7% and 23.5% for total N and P, respectively [6].

In agriculture, nutrient recovery and reuse practices have a potential to address the most pressing problems related to nutrients use along the food chain, such as pollution, depletion of finite resources (such as P), and waste management. Agricultural waste consists of livestock manure, primary agricultural residuals (such as post-harvest crop residuals), and secondary agricultural residuals (from crop processing in agricultural industries). If not properly managed, this waste can be a significant environmental and economic burden [7].

Spreading manure on agricultural land constitutes approximately 53% of the P and 33% of the N applied annually to agricultural soils in the EU27 [8]. However, the spatial segregation of crop-intensive and livestock-intensive areas leads to uneven spatial distribution of manure, creating nutrient-deficient areas on the one hand and nutrient hot-spots on the other [9-11]. Finding cost-effective manure processing technologies that facilitate the transfer of nutrients between these two areas and produce safe and stable fertilisers from organic waste streams is a fundamental quest for sustainable agricultural production.

Domestic wastewater also represents an organic waste stream, from which nutrients can be recovered for agricultural use. The focus within the wastewater sector has, however, traditionally been on *removal* of organic matter and P (including N to a certain degree) from the effluent by applying various treatment methods to protect receiving waters against eutrophication, rather than on nutrient *recovery* per se. Increasingly, however, there is a shift in mindsets towards a circular economy, defined as an economy where the value of products, materials and resources is maintained in the economy for as long as possible, and the generation of waste minimized [12]. This paradigm applied to the

wastewater sector means a shift from the sole focus on waste treatment and nutrient *removal* to the *recovery* of energy and nutrients from waste and further *reuse* of these products [13, 14].

Some nutrient reuse from domestic wastewater (P especially) is already being done via application of sludge to agricultural fields. The P content in sludge depends on whether P removal processes are applied at the WWTP, where P removal from wastewater into the sludge can be achieved by different chemical precipitation or biological removal processes [15]. The suitability of sludge as a fertiliser in agriculture is, however, debated in many countries due to contaminants that can be found in it. In addition, WWTPs are typically not located close to the arable land where sludge from wastewater processing could be applied [11], which increases the difficulty and costs of transporting it. Furthermore, the recovery of N through sludge application is low compared to P recovery rate, since most N is either removed by denitrification or remains in the treated wastewater at conventional WWTPs [13]. For example, Van der Hoek et al. [16] showed that for Amsterdam's WWTP about 38% of the incoming N was captured in the sludge. Out of all N-carrying inflows to the WWTP, urine would be the most interesting source for N recovery if captured separately [16]. Thus, separate collection and treatment of blackwater (wastewater from the toilet only) is able to capture this N-rich stream and, therefore, it is a technical solution on the rise in several countries [17].

Nutrient recovery technologies can be applied to different waste streams. The higher the nutrient concentration in the waste stream the more valuable and economically feasible the recovered product will be. Anaerobic digestion of sludge, agricultural waste and blackwater is widely applied to produce biogas, which can be used as renewable energy. Dewatering of the digestate, often applied to reduce its weight, results in a liquid and a solid digestate phases. The liquid phase of anaerobic digestate is a concentrated source of nutrients, such as N and P, to which nutrient recovery technologies can be applied. By combining anaerobic digestion and nutrient recovery technologies, a treatment process can be obtained that provides both renewable energy and nutrients for plants. Van der Hoek et al. [16] showed that the digestate has a potential of recovering 27% of the incoming N to the WWTP. Nutrient recovery from wastewater and agricultural wastes could decrease the need for mineral P and N fertilisers, reducing the pressure on respective biogeochemical cycles [18, 19]. This makes nutrient recovery an important and integral contribution of the wastewater sector to a circular economy.

2.1.1 Potential solutions

Two promising technologies for N and P recovery identified in systematic maps of ecotechnologies for recovering nutrients and carbon from domestic wastewater [20] and agricultural waste streams [21] are struvite precipitation and ammonia stripping.

Struvite precipitation is an ecotechnology that can be used mainly for P recovery and was one of the most represented ecotechnologies identified in both systematic maps [22, 23]. Struvite is a crystalline mineral composed of equimolar concentrations of magnesium (Mg), ammonium (NH_4) and phosphate (PO4) with the chemical formula MgNH₄PO₄*6H₂O. Struvite is formed under alkaline conditions and the process depends on several parameters, perhaps most notably pH and the molar ratio of NH₄, PO₄ and Mg in the liquid. A simplified precipitation process is depicted in **Figure 1**. Although wastewater typically includes some Mg, the ratio of Mg to PO₄ or NH₄ in the waste stream must often be improved through Mg additions to achieve efficient struvite precipitation from wastewater rich in PO₄ and NH₄. Provided an excess in Mg, the concentration of either PO₄ or NH₄ in the waste stream will limit the reaction (depending on whether PO₄ or NH₄ has the lower concentration). In this review, we refer to this molar ratio as "Mg to limiting reactant". Under optimal conditions, almost complete recovery (100%) of PO₄ or NH₄ can be achieved via precipitated struvite.



Struvite is an effective, slow-release fertiliser with a relatively low content of contaminants, which can replace fertilisers produced from phosphate rock [24]. The value of struvite as fertiliser has only been recently understood and it is now the focus of increasing research attention [25].

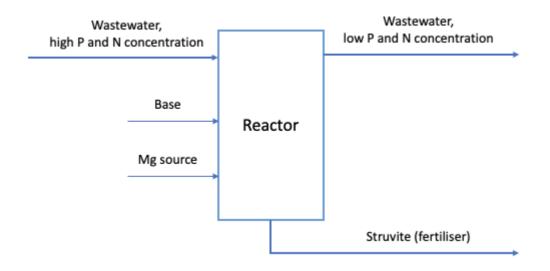


Figure 1. Simplified struvite precipitation process (Source: own elaboration)

Ammonia stripping is applied to liquids containing high concentrations of ammonia [26, 27], and using this method almost complete removal of the ammonia in the liquid can be achieved for a given flow stream [27]. **Figure 2** shows a simplified ammonia stripping process. High temperature and pH increase efficiency of ammonia stripping since this leads to a larger fraction of N as gaseous ammonia. Other parameters that may influence the effectiveness of the process include liquid to gas flow ratio and reactor configuration (i.e. counterflow or cross-current). The stripped ammonia gas is then recovered by absorption to an acid, commonly sulphuric acid. The resulting product is a low pH ammonium sulphate, used as a fertiliser recommended for use on soils with alkaline or neutral reaction [26].

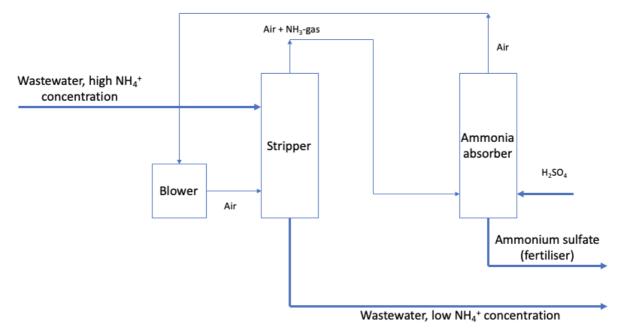


Figure 2. Simplified ammonia stripping process (Source: own elaboration)

Both struvite precipitation and ammonia stripping could potentially be incorporated into existing WWTPs as well as to manure management processes to enhance nutrient recovery, improve WWTP function, and contribute to an increased P recovery. The liquid phase of anaerobic digestate is a concentrated stream of nutrients, commonly produced in the current management process of both manure and wastewater. Therefore, the focus of this review is on the liquid phase of anaerobic digestate as a source of nutrients for recovery. We have chosen struvite recovery and ammonia stripping for this review in order to focus on both P and N recovery, since the N content of struvite is too low to be considered as a fertiliser.

Although there are some relevant reviews on the topic 9, 28, 29], to our knowledge, no systematic reviews of effectiveness of modern ecotechnologies for reuse of N and P from anaerobic digestate have been conducted. Here, we define ecotechnologies as "human interventions in social-ecological systems in the form of practices and/or biological, physical, and chemical processes designed to minimise harm to the environment and provide services of value to society" [30]. This definition was produced by a thematic synthesis of definitions in the literature, encompassing both hard (e.g. mechanical or chemical) and soft (e.g. behaviours and practices) technologies and has been used in two other, preceding systematic maps [20, 21].

2.1.2 Stakeholder Engagement

The topic for this review was initially proposed by the research funder BONUS (<u>https://www.bonusportal.org/</u>). The scope of the project was then refined through expert discussions as part of the process of drafting an application in response to the call by the research funder. The scope and the search strategy were further refined by a stakeholder group consisting of the broader BONUS RETURN project consortium members (see <u>https://www.bonusreturn.com/</u>), local stakeholders from the three BONUS RETURN case study areas in Finland, Poland and Sweden, as well as external experts from these countries, which explains the Baltic Sea basin focus.

2.2 Objective of the review

The primary question for this systematic review is:

Are struvite precipitation and ammonia stripping effective ecotechnologies for recovery and reuse of nitrogen and phosphorus from anaerobic digestate?

The secondary questions are:

SQ1: How effective are struvite precipitation and ammonia stripping for the recovery of nitrogen and phosphorus from anaerobic digestate?

SQ2: How effective are recovered products from these processes (struvite and ammonium sulphate) as fertilisers?

This review focused on *struvite precipitation and ammonia stripping* currently developed and applied globally. We investigated application of recovered products (struvite and ammonium sulphate) in the Baltic Sea and boreo-temperate regions. **Figure 3** describes the study system and the relation between review questions.

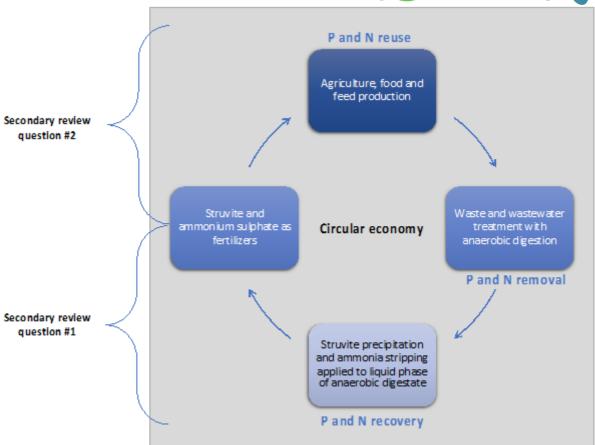


Figure 3 Conceptual diagram with review context and questions. *Note: The diagram is simplified, and it is showing an ideal system (for the purpose of illustration). There are P and N losses along the cycle.*

The first secondary question (SQ1) has the following components:

- *Population(s):* Agricultural residuals and domestic wastewater (including blackwater) globally.
- Intervention(s): Struvite precipitation and ammonia stripping undertaken for the purposes of recovering N and P from the liquid phase anaerobic digestate.
- *Comparator(s):* Before ecotechnology use, a control site without an ecotechnology, a comparison between different ecotechnologies, different intensities of the same ecotechnology, time series after ecotechnology implementation.
- *Outcome(s):* Recovery potential of N compounds (total N, ammonium and/or ammonia) and P compounds (total P, phosphate)) expressed as a recovered percentage in the digestate flow stream and/or total recovery in the wastewater.

The second secondary question (SQ2) has the following components:

- *Population(s):* Ecosystems of boreo-temperate regions, with a focus on the Baltic Sea region (as requested by stakeholders).
- *Intervention(s):* Struvite and ammonium sulphate used as fertilisers.
- *Comparator(s)*: Before the product use, a control site without a product, a comparison between different products, different intensities of the same product, time series after product implementation.
- *Outcome(s):* Crop or biomass yield, N and P uptake by plants, soil N and P content.

2.3 Methods

The review followed the Collaboration for Environmental Evidence Guidelines and Standards for Evidence Synthesis in Environmental Management [31] and conformed to ROSES reporting standards [32] (see **Additional file 1**). It was designed according to the protocol published in Environmental Evidence in early 2019 [33]. Deviations from the originally planned methods were minor during the review process and they were detailed in each sub-section below.

2.3.1 Searching for articles

Bibliographic databases

We searched for evidence in the following databases:

- 1. Scopus
- 2. Web of Science (WoS) Core Collections (consisting of the following indexes: SCI-EXPANDED, SSCI, A&HCI, CPCI-S, CPCI-SSH, and ESCI)
- 3. Electronic Theses Online Service (eThOS)
- 4. Digital Access to Research Theses (DART)
- 5. Directory of Open Access Journals (DOAJ)

Searches were performed using subscriptions of Warsaw University of Life Sciences and Stockholm University. These searches were conducted using English language search terms.

The following search strings were used in bibliographic databases:

Search string for SQ1:

(struvite OR "MgNH4PO4" OR "NH4MgPO4" OR "Magnesium ammonium phosphate*" OR "Crystal green" OR (ammonium AND (sulphate* OR sulphate* OR nitrate*)) OR mascagnite* OR ((stripp* OR scrub*) AND (ammoni* OR NH3 OR nitrogen OR air OR steam))) AND (digest* OR centrate* OR supernatant* OR dewater* OR "solid-liquid" OR "bio refiner*" OR "reject water*" OR effluent* OR "liquid phase") [shown as formatted for WoS]

Search string for SQ2:

(struvite OR "MgNH4PO4" OR "NH4MgPO4" OR "Magnesium ammonium phosphate*" OR "Crystal green" OR mascagnite OR (ammoni* AND (sulphate*" OR sulphate*"))) AND (fertili* OR field* OR farm* OR soil* OR agricult* OR arable OR agron* OR nutrient* OR crop* OR seed* OR food* OR yield* OR produc* OR uptake OR plant* OR vegetat* OR absor*) [shown as formatted for WoS]

Search engines

Searches were also performed in Google Scholar, which is an effective tool for identifying grey literature [34]. Searches were performed in English and Swedish. Google Scholar searches were restricted to articles published after 2013, as above. The first 1000 search results were extracted as citations using Publish or Perish software [35] and introduced into the duplication removal and screening workflow alongside records from bibliographic databases. See **Additional file 2** for results of WoS, Scopus, eThOS, DOAJ, DART and Google Scholar searches.

Testing comprehensiveness of the search

Two lists of articles of known relevance to the review were screened against search results to examine whether the search strategy can locate relevant evidence. Some benchmark articles from the list were not found during the scoping exercise, and search terms were examined to identify the reasons why articles were missed and modified accordingly. The final search strings captured all the benchmark articles.

Assembling library of search results

Results of the searches in bibliographic databases and Google Scholar were combined, and duplicates removed prior to screening. A library of search results was assembled in a review management software (i.e. EPPI reviewer [36]).

2.3.2 Article Screening and Study eligibility criteria

Screening process

Screening was conducted at two levels: at title and abstract level (conducted together for efficiency), and at full text level. The full texts were retrieved, tracking those that cannot be located or accessed. Retrieved records were screened at full text. A list of unobtainable records is in **Additional file 3**.

Prior to commencing screening, consistency checking was performed on a subset of articles (10%) at both title and abstract level and full text level screening. A subset of title and abstract records and full texts was independently screened by up to three reviewers. The results of the consistency checking were compared between reviewers and all disagreements were discussed in detail. Where the level of agreement is low (below c. 80% agreement), further consistency checking was performed on an additional set of articles and then discussed. Following consistency checking (i.e. when agreement is above 80%), records will be screened by one experienced reviewer. EPPI reviewer's machine learning component was not used for screening as it was this component was not publicly launched at the time screening was done.

Eligibility criteria

The following criteria was applied at all levels of screening: **For SQ1:**

- *Eligible population(s):* Liquid phase of anaerobic digestate from agricultural residuals and domestic wastewater (including blackwater). We included studies conducted anywhere across the globe.
- *Eligible intervention(s):* Struvite precipitation and ammonia stripping undertaken for the purposes of recovering N and P.
- *Eligible comparator(s):* Before ecotechnology use, a control site without an ecotechnology, a comparison between different ecotechnologies, different intensities of the same ecotechnology, time series after ecotechnology implementation. In case of SQ1, inlet concentrations served as a control
- *Eligible outcome(s):* Recovery potential of N compounds (total N, ammonium and/or ammonia) and P compounds (total P, phosphate) expressed as recovered percentage in the digestate flow stream and/or total recovery in the wastewater
- *Eligible languages*: English, Finnish, Polish and Swedish

For SQ2:

- *Eligible population(s):* Ecosystems of boreo-temperate regions, with a focus on the Baltic Sea region. Eligible studies were in both hemispheres, with fully humid temperate (Cfa, Cfb, Cfc) and fully humid boreal (Dfa, Dfb, Dfc, Dfd) climates (according to the Köppen-Geiger climate classification [37]).
- *Eligible interventions(s):* Struvite and ammonium sulphate used as fertilisers. In contrast to what was stated in the protocol, we have included struvite and ammonium sulphate regardless of their origin.



- *Eligible comparator(s):* None, before the product use, a control site without a product, a comparison between different products, different intensities of the same product, time series after product implementation.
- *Eligible outcome(s):* Effectiveness of the products expressed as crop or biomass yield, N and P uptake by plants, soil N and P content.
- *Eligible languages*: English, Finnish, Polish and Swedish.

Additional file 4 includes a list of articles excluded at title and abstract level, and at full text level with reasons for exclusion.

2.3.3 Study validity assessment

Eligible studies were subject to a study validity assessment. The assessment evaluated external and internal study validity and categorised relevant studies accordingly. The detailed criteria for the study validity assessment of included studies (i.e. critical appraisal tool) was developed and trialled during the review process in several meetings with subject experts. The critical appraisal tool was tested on a set of 10% of studies for each secondary question and by the entire team. We present study validity assessment criteria and scoring for each secondary question separately. There were no reviewers on our team who have authored articles to be considered within the review.

Study validity assessment for studies relevant to SQ1

Study validity assessment for studies relevant to SQ1 included evaluation of 1) study set up and design flaws (due to calculation errors, invalid outcome measurements or failure to control for the effect of additional competing interventions such as irradiation, dialysis or microwave treatment.), and 2) reporting bias (i.e. selective reporting of study findings). A study judged to had flaws in design and set up or reporting bias, was excluded from the narrative and quantitative review. Alternatively, studies without these issues were then assessed for clarity of reporting on reactor input, recovery process and composition of the final product (see details in **Additional file 5**). If a study was deemed to be unclear on two or more of these domains, it was classified as "unclear", and excluded from quantitative synthesis. The included studies that were not judged as unclear, were included in both quantitative and narrative synthesis. Studies on ammonia stripping were not further classified, while studies on struvite precipitation were scored as high or medium validity. The differentiation between high and medium studies was done based on two criteria (as detailed in **Table 1**): 1) input chemicals include contaminants that may influence the process; 2) the rigour of the product analysis.

Domain	Criteria	Points
Input chemicals	Added chemicals are free of contaminants that could potentially disturb the precipitation process (such as potassium, which can lead to struvite-K formation, or calcium, which can lead to formation of calcium phosphates)	1
Product analysis	Article applies at least two methods that complement each other and together produce a complete overview of the product. Examples: SEM/EDX, SEM/EDX/XRD	2
Product analysis	Article applies only one method, or two methods that do not complement each other in a way such that a complete overview is produced. Examples: SEM/XRD, XRD	1
Product analysis	No advanced analytical tools are used at all.	0

Table 1. Scoring scheme for high or low validity struvite precipitation studies.

If a study received at least 2 points in total, it was judged to be of high validity. If it received less than 2 points, it was categorised as having medium validity. The values assigned during the study validity assessment were recorded in a detailed manner. The validity of a study was assessed by one reviewer and checked by a second reviewer. Final decisions regarding doubtful cases were taken by the whole review team. Additional file 5 includes details of critical appraisal criteria and a list of studies excluded based on quality assessment, along with the reasons for exclusion.

Study validity assessment for studies relevant to SQ2

Study validity assessment for studies relevant to SQ2 included evaluation of:

- 1. Level of methodological details reported and overall methodological clarity (which affects the ability to judge the validity);
- 2. Study design
- 3. Number of replicates and scale of replication
- 4. Confounding factors and susceptibility to bias
- 5.

Specifically, the effectiveness of struvite and ammonium sulphate on plant growth had to be tested in a study design that included at least 3 replicates and a control with no fertilizer and/or a mineral fertilizer. In addition, we required that the experimental design (e.g. fertilizers, soils, plants, scale - field or pot) was clearly described and we also reviewed that laboratory analyses and their methods were written in detail. Validity of spatial scale was only assessed for field studies, as it was considered that spatial scale is not relevant for pot studies. It was also checked if the three studied outcomes: dry matter yield, plant phosphorus uptake and soil phosphorus content, were analysed with well-known, standardised methods after at least 4 weeks of plant growth before harvesting. Finally, presence of any confounding factors was assessed as well as evaluation of generalisability of the study findings. Examples of confounding factors could be, for example, unaccounted for weather conditions during field experiments or presence of external factors affecting the solubility of struvite crystals.

Studies with low validity or with no external validity were excluded from the review.

Studies that did not report any measures of variability were judged as 'unclear' and included in narrative synthesis.

2.3.4 Data coding and extraction strategy

Data were extracted from included studies and recorded in spreadsheets that include pre-determined coding adapted for each secondary question. We extracted information on study characteristics, intervention details, comparators, effect modifiers, study validity assessment scores and study findings. Study findings (including the outcome values) were extracted from tables and graphs, using an image analysis software WebPlotDigitizer (<u>https://automeris.io/WebPlotDigitizer/</u>) where needed. Missing parameters (such as unpublished efficiencies or concentrations) were calculated from reported raw data whenever possible. The review team calculated summary statistics if the raw data were provided. All data were obtainable from published materials, and the review team did not ask authors of relevant articles for access to unpublished raw data. All extracted data records are in **Additional file 6** along with excluded data that could not be used in the quantitative synthesis.

A consistency checking exercise was done before coding and data extraction on a subset (10%) of records by all reviewers. All disagreements were discussed, and the coding scheme was clarified where needed.

2.3.5 Potential effect modifiers/reasons for heterogeneity

The following factors, which potentially can cause variation in measured outcomes, were considered and recorded if reported in primary studies. The list was refined during the review process based on consultations with experts on the review team.

For SQ1:

- Specific characteristics of intervention and ecotechnology process parameters including temperature, pH, reactor type, inlet concentrations, molar ratios of reactants, amount of chemicals added and hydraulic retention time
- Type of a substrate used for anaerobic digestion
- Type of waste treatment processes before anaerobic digestion phase and before application of struvite recovery or ammonia stripping
- Study design, including study scale and sampling method

For SQ2:

- Study scale (field or pot)
- Source of recovery (agriculture, sewage sludge or synthetic)
- The difference in fertilizer rate between recovered and mineral fertilizer
- Soil texture (sand, loam or clay)
- Soil pH (low, medium or high)
- Crop groups as in the Indicative Crop Classification of FAO (<u>http://www.fao.org/fileadmin/templates/ess/documents/world_census_of_agriculture/app_endix3_r7.pdf</u>)

2.3.6 Data synthesis and presentation

A narrative synthesis described the validity of the results along with a summary of findings (in a graphical and tabular form) for each included study (see **Additional file 7**) for each secondary question. In addition to the narrative synthesis, quantitative synthesis was performed on a subset of data relevant for SQ1 only and for struvite precipitation specifically. Due to high heterogeneity and lack of data, no quantitative synthesis was performed for ammonia stripping. In the struvite precipitation dataset, the effect of selected process parameters on the removal efficiency of limiting reactant (NH₄ or PO₄) was investigated. Removal, rather than recovery, was chosen as the measure of outcome since this is what authors most commonly reported (removal and recovery rates are relatively similar for struvite precipitation). The investigated parameters were pH, Mg source (such as MgO or MgCl₂) and ratio of Mg to NH₄ or PO₄ (depending what ratio was limiting).

The review also highlighted methodological deficiencies of the relevant studies, and major knowledge gaps in the evidence base for each secondary question. Knowledge gaps were highlighted by identifying un- or underrepresented subtopics using heat maps.

Statistical analysis for SQ1, struvite precipitation dataset

The main objective of the data synthesis was to derive quantitative models of the struvite removal and how it was influenced by pH, ratio of Mg to limiting reactant and Mg source. The statistical analyses involved the following steps:

- 1. Handling of extreme values
- 2. Calculation of mean values
- 3. Fitting and selection of prediction models
- 4. Graphical presentations of models and prediction errors

Scatter charts of the two-dimensional distribution pH and Mg/(limiting reactant) revealed that, with few exceptions, the struvite removal was measured where $7 \le pH \le 11$ and $0 \le ratio$ of Mg to limiting reactant ≤ 4 . Data outside that rectangle was considered to be too sparse for modelling the struvite removal and thus excluded in the subsequent statistical analyses. Data were also excluded for one substrate where the reported values of ratio of Mg to limiting reactant were obviously wrong. Apart from these two exceptions (for details see **Additional file 6**, Outliers) all extracted data were used.

The studies from which data were extracted varied strongly with respect to the experimental design that was used. Among a total of 28 substrates that were examined only 10 were subjected to an experimental design with true replicates, i.e. 2 or more experiments for at least some of the examined combinations of substrate item, pH, ratio of Mg to limiting reactant and Mg source. Furthermore, the studies varied strongly with respect to the distribution of design points. Among a total of 38 combinations of substrate and Mg source that were examined only 10 had a design in which both pH and ratio of Mg to limiting reactant were varied. For 10 of the 38 combinations both pH and ratio of Mg to limiting reactant were held fixed, and for the remaining 18 combinations either pH or ratio of Mg to limiting reactant was fixed. Together, this meant it was not feasible to assign meaningful standard errors to the observed mean struvite removal in individual studies.

To overcome the abovementioned difficulties, we compiled a dataset containing the average struvite removal for each investigated combination of substrate, pH, ratio of Mg to limiting reactant and Mg source. Thereafter, regression models were fitted to the entire dataset and to subgroups representing specific types of substrates. This approach had the advantage that the models were primarily fitted to studies with a good experimental design in which both pH and ratio of Mg to limiting reactant were varied, whereas studies in which these two variables were fixed played a minor role.

An ideal regression model would be able to produce both accurate estimates of the expected removal for a great variety of substrates, Mg sources, levels of pH, ratios of Mg to limiting reactant and also, reliable uncertainty estimates. Considering that the available dataset was too small to enable both a flexible structure of the mean function and an advanced correlation structure, any regression model is a compromise. We decided to prioritize smoothing techniques allowing flexible models of the expected removal and accept that uncertainty estimates and p-values of statistical significance may be underestimated because all observations in such models are regarded as statistically independent. More specifically, we decided to fit generalized additive models (GAM) to data. GAM is a class of smoothing techniques in which a univariate response variable is related to smooth functions of a set of predictors [38]. In its original form, the expected value E(Y) of the response variable (or a function g(E(Y)) of this value) was assumed to be a sum of smooth functions of a set of predictors $x_1, x_2, ..., x_p$, i.e.

$$E(Y) = s_1(x_1) + s_2(x_2) + \dots + s_p(x_p)$$
(1)

where $s_1, s_2, ..., s_p$ are estimated from the given data. If data are divided into classes, equation 1 can be extended with constants representing systematic level shifts between the different classes. Current GAM procedure also makes it possible to fit models of the type

$E(Y) = spline2(x_1, x_2)$

where spline2 is smooth response surface, a so-called thin plate spline, in two variables. The main difference between models 1 and 2 is that model 1 assumes that the response to x_1 is the same for all levels of x_2 , whereas model 2 allows interaction effects of x_1 and x_2

In our case, the removal of struvite expressed in percent was selected as response variable Y, whereas pH or ratio of Mg to limiting reactant, or both of these, were used as predictors. In some analyses of the entire dataset, equation 1 was extended with constants representing different types of substrates. All analyses involving GAM models were performed using a procedure called proc GAM in SAS

(2)



Enterprise Guide, which is the primary graphical user interface of the software package SAS developed by SAS Institute for advanced analytics, multivariate analysis, business intelligence, criminal investigation, data management, and predictive analytics (<u>https://www.sas.com/en_us/company-information.html#history</u>).

2.4 Review findings

Although we have used two separate search strings (one for each secondary question) to find relevant literature, we have combined search results and screened them together. This was done as some of the studies included findings relevant for both secondary questions. After screening and coding, analysis and synthesis of findings for each secondary question was done separately. All searches yielded 6473 results (**Figures 4 and 5**). Google scholar searches in Swedish yielded no relevant records (please see **Additional file 2** for details). After removing duplicates (2739), 3743 records were screened on title and abstract. Out of included 881 records, 81% was retrieved and 713 records were screened on full text. At the full text screening stage included studies were marked as relevant to SQ1 or SQ2 and outcomes of critical appraisal and synthesis as well as findings will be presented separately.

2.4.1 Characteristics of studies included in narrative synthesis for SQ1

In the context of SQ1, a study is defined as having a specific treatment trail, i.e. a specific reactor setup and was conducted by the same lead group of authors. Different experiments can use the same reactor setup, but the process parameters (such as pH and temperature) are being varied. In each article, there can be several studies and studies can be reported across different articles. A study can contain several experiments.

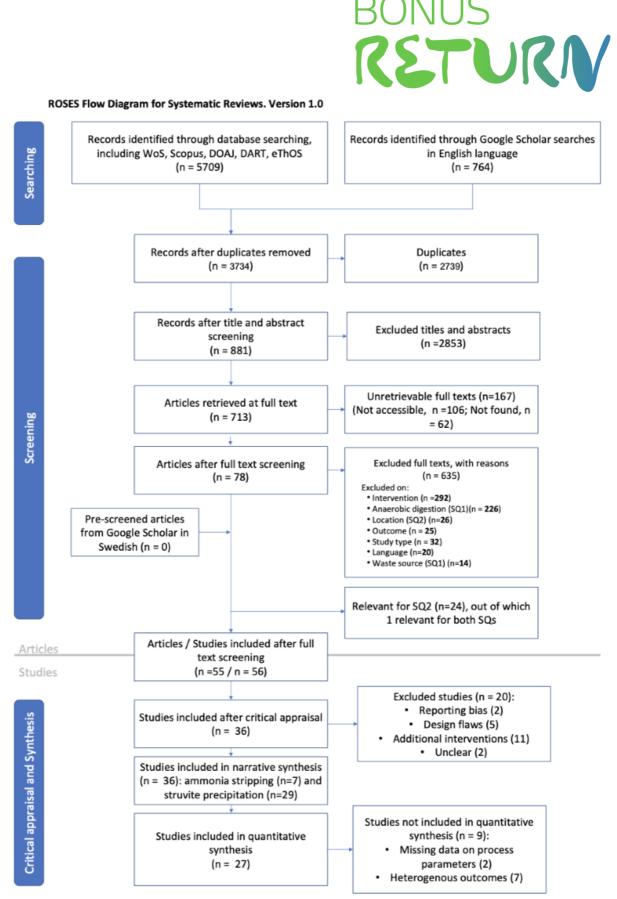


Figure 4 ROSES flow diagram [39] showing all literature sources and inclusion/exclusion process for SQ1 relevant studies.

Only 11% of screened articles at full text (77) were judged to be relevant for SQ1 (**Figure 4**). In the critical appraisal stage, 20 studies were removed. In sum, evidence base of ecotechnologies for struvite



precipitation included 29 studies, and evidence base for ammonia stripping is totalling 7 studies only. Out of these, 27 studies were included in quantitative synthesis (with some excluded experiments as explained earlier and detailed in Additional file 6), whereas all ammonium stripping studies (due to heterogeneity, see details below in Quantitative synthesis for SQ1 section) and 2 struvite precipitation studies could not be included in quantitative synthesis (due to missing data). All the literature sources used in the review and the number of studies included and excluded at different stages of the review process are in **Figure 4**.

All included studies (36) originated from 35 articles (one article included 2 studies), the majority of which were published during 2018 (**Figure 5**) and in English.

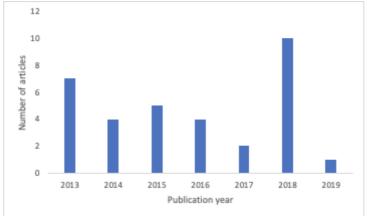


Figure 5 Publication year of included articles relevant for SQ1.

2.4.2 Overview of struvite precipitation evidence base

The evidence base for struvite precipitation included 27 studies and 287 experiments. Most included studies (48%) were in Asia, and specifically in China (**Table 2**).

Continent	Country	Number of studies
	China	13
Asia	Israel	1
	South Korea	2
Australasia	Australia	1
	France	1
Europe	Spain	2
	Sweden	1
North America	Canada	2
North America	USA	1
Couth Amonion	Brazil	1
South America	Colombia	1
No location stated		1

Table 2 Location of included studies

Different digested substrates were included such as swine, poultry and cattle manure as well as wastewater sludge. The most prevalent substrate was supernatant of anaerobically digested sewage sludge, followed by swine manure (**Figure 6**).

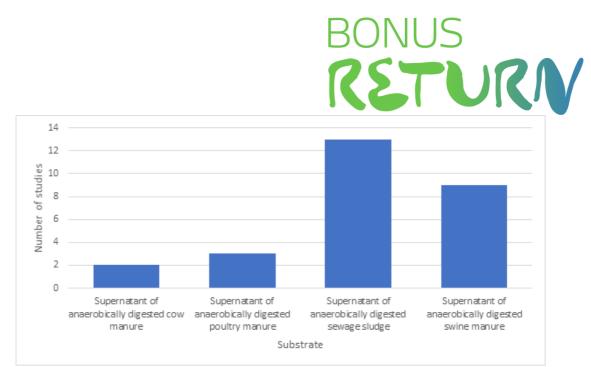


Figure 6 Distribution of struvite precipitation studies across different substrates

The most prevalent studies were conducted in a laboratory context on a small scale (88.8%) (with 274 experiments). These were laboratory or bench scale operations with reactor input flows from a couple of millilitres to up to 10L. Only 11.1% of the studies (with 13 experiments) were classified as medium scale (defined as pilot scale operations with reactor input flows from 100 to up to 5000 L) (**Figure 7**). There were no large-scale experiments (i.e. full-scale wastewater treatment plant operations).

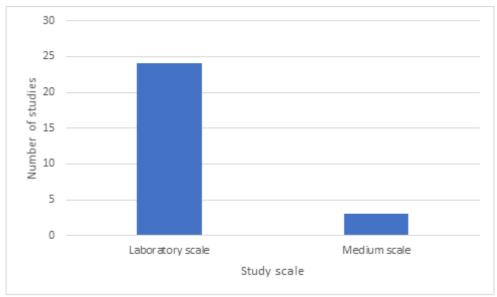


Figure 7 Distribution of struvite precipitation studies across different scales.

The majority of studies were conducted as batch experiments (81.5%), while there were only 18.5% studies set up as a continuous flow (**Figure 8**).

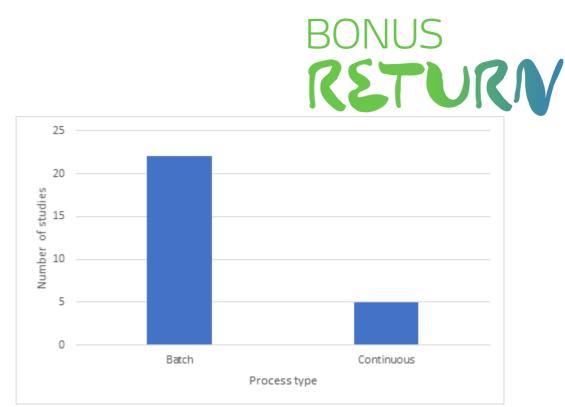


Figure 8 Distribution of struvite precipitation studies across different process types

Sources of Mg included MgO, MgCl₂, MgSO₄, Mg(OH)₂ and bittern (which is a by-product from salt production). Mg was most frequently sourced from MgCl₂ (in 54% of the experiments), while 22.1% of experiments used MgO, and 11.7% used MgSO₄ (**Figure 9**). In 0.3% of experiments existing Mg in the waste stream was used for the reaction (and no additional Mg was used).

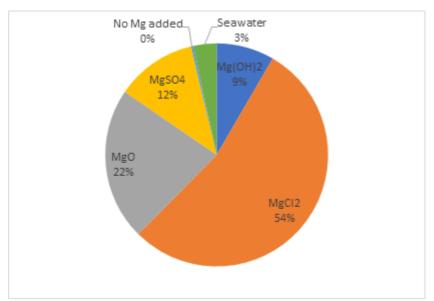


Figure 9 Different sources of added Mg in experiments conducted for struvite precipitation and included in quantitative synthesis.

2.4.3 Narrative synthesis for struvite precipitation effectiveness studies

The included studies were relatively homogenous, both with respect to physical conditions and the type of information reported. Studies differed in concentrations of inlet nutrients and total solids. Twenty studies were excluded during critical appraisal due to: competing or additional interventions (11), design flaws (5), reporting bias (2) and clarity issues (2). Out of 29 studies that passed critical appraisal stage, 12 studies were judged to have high validity and 17 as low. During quantitative synthesis, 2 studies were excluded since they lack data on process parameters and 5 experiments were

removed as outliers (see Additional file 6, outliers). All the included studies reported pH, which varied between 5 and 13, as well as ratio of Mg to limiting reactant, which varied between 0.5 to 1 and 14.5 to 1. Removal efficiency varied between 0.1 to 100% with respect to limiting reactant. In general, high removal rates were achieved, with the median removal of limiting reactant reaching 91.1%. PO₄ and NH₄ were the limiting reactant in 49.7 per cent and 4 per cent of the experiments, respectively. For the remaining 46.3 per cent of the experiments, the molar ratio of PO₄ to NH₄ was 1 to 1. Certain process parameters appear to have a clear influence on the outcome, specifically pH and the ratio of Mg to limiting reactant (see Quantitative synthesis section). For a complete list of the studies included in the evidence base and narrative synthesis, see **Additional file 7**.

2.4.4 Quantitative synthesis for struvite precipitation effectiveness studies

Both pH and the ratio of Mg to limiting reactant were found to have a clear influence on the removal rates. The response to pH was found to be non-linear, resembling a bell curve with a maximum around pH 9.5 (**Figure 10**).

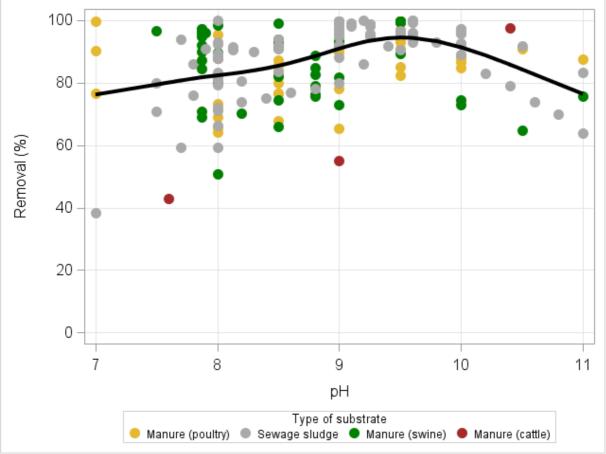


Figure 10 GAM model fitted to struvite removal and pH

The response of removal efficiency to Mg to limiting reactant ratio was found to be almost linear, with an average of around 85% removal at 1:1, increasing to approach almost complete removal at 4:1 (**Figure 11**). No clear conclusions were able to be drawn regarding the effect of Mg source on the struvite precipitation effectiveness.

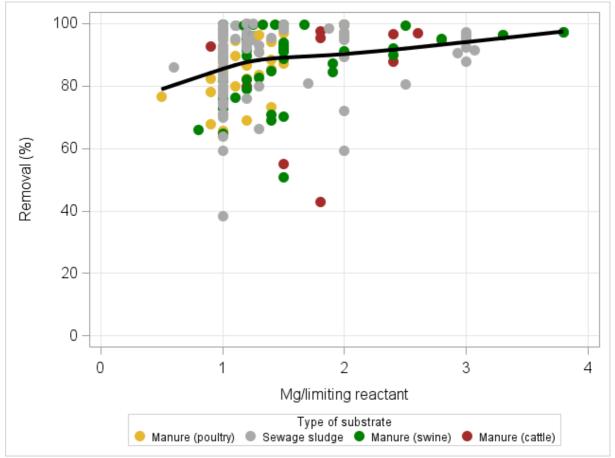


Figure 11 GAM models fitted to struvite removal and ratio of Mg to limiting reactant.

2.4.5 Overview of ammonia stripping evidence base

The evidence base for effectiveness of ammonia stripping was rather small, containing only 7 studies that included 38 experiments in total. Most studies were conducted in Europe (6) and there was only 1 study located in China (**Table 3**).

Continent	Country	Number of studies
Europe	Belgium	1
	Italy	2
	Spain	1
	Switzerland	2
Asia	China	1

Table 3 Location of included studies

Studies included a wide variety of substrates, including poultry, swine, cow, wastewater sludge (Figure 12).



Figure 12 Distribution of ammonia stripping studies across different substrates and locations.

Equal number of studies (3) were conducted in a laboratory context on a small scale and at the medium scale. Large scale studies included 9 experiments and small-scale laboratory studies included 27 experiments. There was 1 medium-scale study (with 2 experiments) (**Figure 13**).

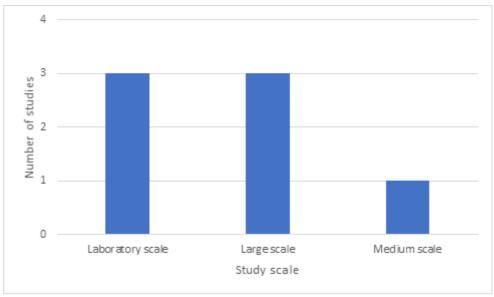


Figure 13 Distribution of ammonia stripping studies across different study scales.

Four studies were conducted as batch experiments (57.1%), and 3 were set up as a continuous flow (Figure 14).

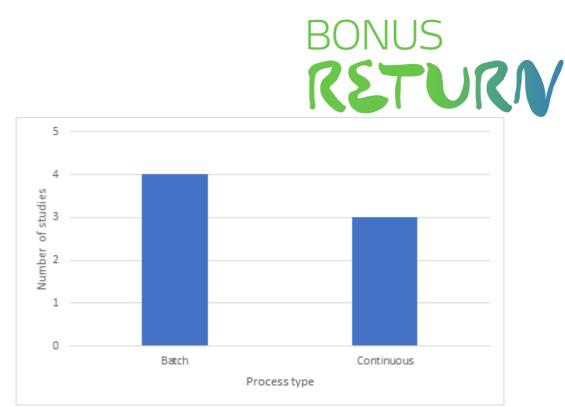


Figure 14 Distribution of ammonia stripping studies across different process types

A majority of studies had a counterflow reactor (5), where 2 studies have not specified the reactor type (Figure 15).

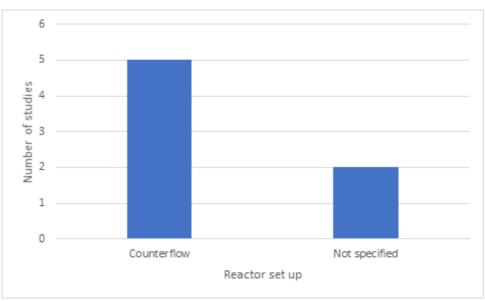


Figure 15 Distribution of ammonia stripping studies across different reactor set-ups.

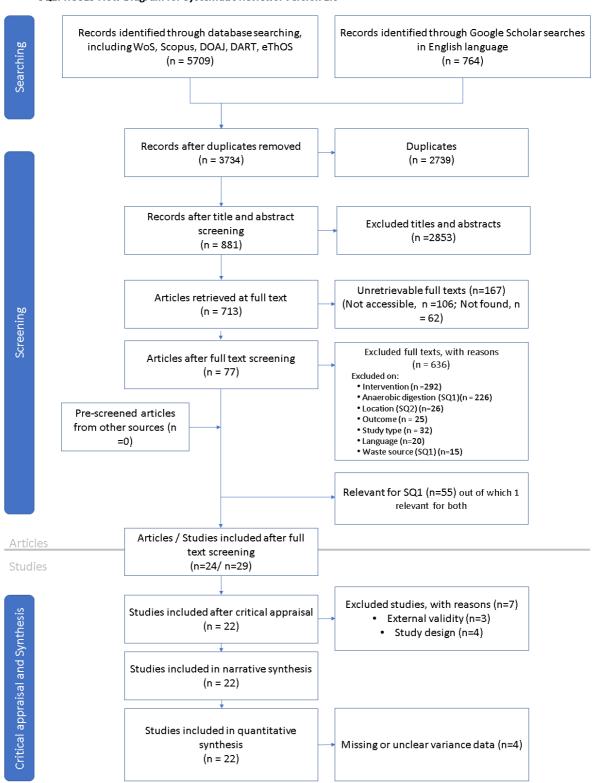
2.4.6 Narrative synthesis for ammonia stripping effectiveness studies

Studies were relatively heterogeneous (see "Limitations of the evidence base" for details). No studies were excluded during critical appraisal, and no distinction was made regarding the validity of the different studies. Different digested substrates were included, including wastewater sludge and different types of manure. All the studies reported pH, which varied between 8 and 12.9. The reported gas to liquid flow ratios varied between 640 to 1 and 6000 to 1. Presented liquid and air temperatures varied between 20 and 60 degrees Celsius, as well as between 20 and 80 degrees Celsius, respectively. Only setups with counterflow reactor configuration were found. Total nitrogen removal varied between 17 and 95%. Due to the size of the evidence base, and the heterogeneity between studies, no conclusions are presented regarding the influence of different process parameters on the outcome



of ammonia stripping. For a complete list of the studies included in the evidence base and narrative synthesis, see **Additional file 7.** Low data availability and high heterogeneity between studies in the evidence base also precluded quantitative synthesis for ammonia stripping dataset (see section "Limitations of the evidence base").

2.4.7 Characteristics of studies included in synthesis for SQ2



SQ2: ROSES Flow Diagram for Systematic Reviews. Version 1.0

Figure 16 ROSES flow diagram [39] showing literature sources and inclusion/exclusion process for SQ2.



All the literature sources used in the review and the number of studies included and excluded at different stages of the review process are in **Figure 16**. Only 3% (24) of articles screened at full text were judged as relevant for SQ2. These articles contained data for 29 studies in total, of which 26 studies were struvite studies and only 3 ammonium sulphate studies. Seven studies (6 struvite and 1 ammonium sulphate) were excluded during critical appraisal. Thus, 22 studies were included in the narrative synthesis, of which 21 concerned struvite and only 1 ammonium sulphate.

Most studies were published in 2018 (**Figure 17**). Note that searches were performed in the first half of 2019, so the study count for this year is likely underestimated. All included studies were journal articles published in English.

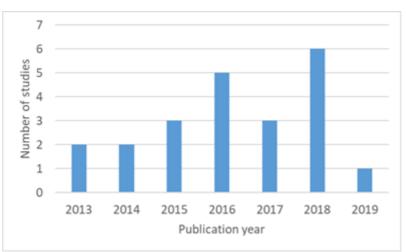


Figure 17 Publication year of included studies relevant for SQ2.

2.4.8 Overview of struvite fertilizer evidence base

All studies included in the evidence base reported their geographic location. The studies in the struvite evidence base were located exclusively in Europe (N=15) and North America (N=6). Germany (N=7) and USA (N=4) were two countries with the highest number of studies (Figure 18).

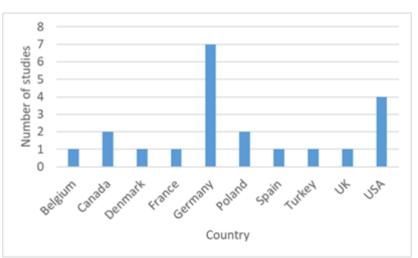


Figure 18 Countries of included studies relevant for SQ2 (struvite).

Table 4 presents the summary of experimental designs for struvite studies included in SQ2 evidence base. In the context of SQ2, the term 'experiment' was used for any unique combination of eight

'factors' within a given study in SQ2: soils, plants, plant parts, fertilizer rates, fertilizer types, placement methods, crop rotations and harvests. The data describing each experiment can be found in **Additional File 6**. Overall, 22 struvite studies contained 166 experiments. Such a definition of 'experiment' results from the nature of studies included in SQ2 and embraces a large diversity of experimental designs. However, most of individual studies contained up to two out of eight factors in their design. The most simple design was that of Cerrillo (2015), which did not include variability in any of the eight factors, whereas the most complex design was that of Katanda (2016), which included variability in five factors: soils, crops, fertilizer rates, placement methods and harvests.

refer to the numbers of		,					enpen			0	C	
Article	Article ID	Study ID	Field/pot	Soils	Plants	Plant parts	Fertilizer rates	Fertilizer types	Placement methods	Crop rotations	Harvests	Number of factors with more than 1 item
Achat (2014)	40526733	1	Pot	1	1	2	1	5	1	1	1	2
Ackerman (2013)	40531652	1	Pot	1	1	1	5	2	1	1	1	2
Bonvin (2015)	40526600	1	Pot	1	1	1	1	1	1	1	3	1
Cerrillo (2015)	40530097	1	Pot	1	1	1	1	1	1	1	1	0
Cala (2010)	40521241	1	Field	1	1	3	3	1	1	1	1	2
Cole (2016)	40531241	2	Field	1	1	2	1	1	1	1	1	1
Ehmann (2017)	40531195	1	Pot	2	2	1	3	1	1	1	1	3
u:h (2010)	40526422	1	Field	2	2	1	3	1	1	1	1	3
Hilt (2016)	40526423	2	Field	1	2	1	2	1	1	3	1	3
Katanda (2016)	40526512	1	Pot	2	2	1	2	1	2	1	3	5
Lemming (2017)	40526230	1	Pot	1	1	1	4	1	1	1	1	1
Rech (2018)	40531710	1	Pot	1	2	2	1	3	1	1	1	3
Szymanska (2019)	40525984	1	Pot	2	2	1	1	2	1	1	1	3
Uysal (2013)	40530620	1	Pot	1	2	1	1	1	1	1	1	1
Vaneeckhaute (2014)	40531914	1	Pot	2	1	1	1	1	1	1	1	1
Vecel (2015)	40531393	1	Pot	1	4	1	1	1	1	1	1	1
Vogel (2015)	40531393	2	Pot	1	2	1	1	1	1	1	1	1
Vogel (2017)	40526341	1	Pot	1	3	1	1	1	1	3	1	2
Wollmann (2018)	40530844	1	Field	1	1	1	2	2	1	2	1	3
Wellmann (2018)	40520044	1	Pot	1	3	1	1	1	1	1	3	2
Wollmann (2018)	40530941	2	Pot	1	1	1	1	2	1	1	1	1
Worwąg (2018)	40526195	1	Pot	1	1	1	3	1	1	1	1	1
Number of studies	with more tha	n 1 ite	m	5	11	4	9	6	1	3	3	

Table 4 Summary of experiment designs for studies included in SQ2 (struvite fertilizer). The values refer to the numbers of soils, plants, etc. that were included in experimental design within each study.

Out of eight considered factors constituting single experiments within each study (**Table 4**), plants and fertilizer rates were the most common ones, with N=11 and N=9, respectively. Six studies included some diversity in the type of applied struvite fertilizer, and five studies included diversity in soil types. Four studies reported outcomes for different plant parts (e.g. shoots and roots), three studies for



different crop rotations (consecutive growing seasons) and also three for different harvests (within one growing season). Only 1 study contained experiments with variable struvite placement methods (seed row and sideband).

Figures 19-24 present distributions of studies across potential effect modifiers: experiment type (**Figure 19**), soil texture class (**Figure 20**), soil pH level (**Figure 21**), FAO crop groups (**Figure 22**), struvite recovery sources (**Figure 23**) and P application rates in struvite (**Figure 24**). If a study had an experimental design with e.g. two soil types, each of them was counted separately, so the total number of studies is not equal throughout **Figures 19-24**.

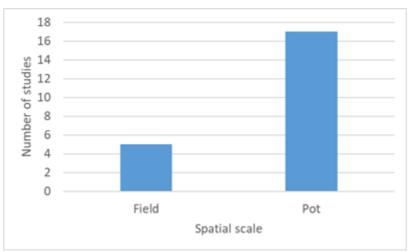


Figure 19 Distribution of studies across different spatial scales.

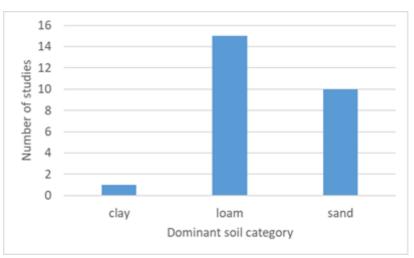


Figure 20 Distribution of studies across different soil texture classes.



Figure 21 Distribution of studies across different soil pH levels.

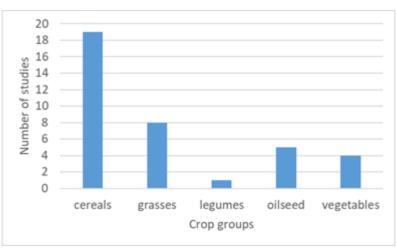


Figure 22 Distribution of studies across different crop groups.

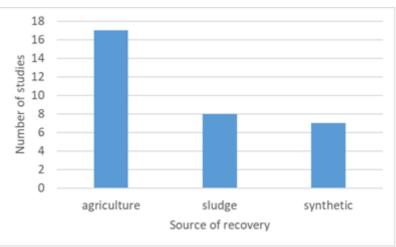


Figure 23 Distribution of studies across different struvite recovery sources.

Overall, studies with pot experiments (N=17) prevailed over those with field experiments (N=5). There was no article containing studies with both field and pot experiments. Loamy soil texture (N=15) was dominant, followed by sandy (N=10) and clayey (N=1) texture. Studies with high soil pH level were the

most frequent (N=13), whereas medium (N=10) and low (N=4) pH levels occurred less frequently. The majority of studies used grain crops (N=19) for testing struvite effectiveness, followed by grasses (N=8) and vegetables (N=7). Some of the FAO crop groups that are relevant for bore-temperate climate zones, such as fruits, root/tuber crops and sugar crops, were not represented at all. As regards to the source of recovery of struvite, agricultural sources (N=17), mainly manure-based, predominated over sewage sludge (N=8) and synthetic (N=7) struvite. In 26 studies equal rates of P in struvite in mineral fertilizer used as control were used, whereas in 12 studies different P rates were applied.

2.4.9 Narrative synthesis for struvite fertilizer effectiveness studies

As already described in the previous sub-section (cf. **Table 4**), the included studies were rather heterogenous. Studies differed in soil characteristics (texture, alkalinity), fertilizer rates and crop groups. Out of 26 struvite studies included in the critical appraisal, five were excluded due to: external validity (2) and study design flaws (3). In addition, four studies were labelled as unclear due to missing or unclear variance, which affects conducting of meta-analysis. For a complete list of the studies included in the evidence base and narrative synthesis, see **Additional file 7**.

As presented in **Table 5**, out of three studied outcomes, crop/biomass yield, as well as P uptake by plants were the most widely studied (18 and 17 studies, respectively), and contained the largest number of experiments (162 and 151, respectively). The least studied outcome was soil P content, for which only 6 studies and 30 experiments were found.

Table 5 Numbers of studies and experiments providing quantitative data for different outcomes. The numbers in parentheses denote numbers of studies/experiments that were reporting values of measures of variability in addition to outcome values.

Outcome	Number of studies	Number of experiments
Biomass/yield	18 (14)	162 (96)
P uptake by plants	17 (13)	151 (87)
Soil P content	6 (5)	30 (27)

A qualitative assessment of the outcomes demonstrates that recycled struvite is a suitable phosphorus fertilizer for diverse plants, including many cereal crops, several grass species and oilseed, vegetable and legume crops. The most reported dry matter yield and P recovery rates were similar between struvite and mineral fertilizer use in most of the studies. In several studies, application of struvite led to higher yields than application of mineral P fertilizer, while some other, but less abundant studies, showed the opposite. Indeed, some plants like tomato and corn, which both have a high fertilizer demand, were able to utilize the struvite fertilizer even better than the mineral fertilizer. Only in very few experiments, e.g. with spring barley and canola, the dry matter yield of plants fertilized with struvite was significantly lower compared with application of mineral fertilizer. This was explained by e.g. soil properties (high pH level), relatively short growth periods (up to six weeks) and the granular form of applied struvite hindering fast dissolution in soil.

2.4.10 Overview of ammonium sulphate fertilizer evidence base

The only ammonium sulphate study included in the evidence base was located in the USA and originated from a scientific journal article published in 2018. This field study had a quite complex experimental setup, with two types of ammonium sulphate, two crops, two crop rotations and two soil types. In this study ammonium sulphate was not used on its own but as an addition to urea.



2.4.11 Narrative synthesis for ammonium sulphate fertilizer effectiveness studies

Out of 3 ammonium sulphate studies included in the critical appraisal, two were excluded due to: external validity (1) and study design flaws (1). One study that was included in the evidence base consisted of 8 experiments and reported crop yield and soil inorganic N, including variance data, as outcome variables. This study showed that ammonium sulphate was an effective addition to urea fertilizer applied to spring wheat and sugar beet crops in a field experiment in Minnesota, USA. A physical mixture of urea with ammonium sulphate was found to increase yield more than a granular fertilizer containing a homogenous blend of urea with ammonium sulphate. No quantitative synthesis was feasible in the case of ammonium sulphate evidence base.

While ammonium sulphate used to be applied in agriculture to a larger extent, other conventional fertilizers have been favoured more recently. This is likely the reason that the fertilizing effects of ammonium sulphate do not seem to have been studied extensively during our investigated time period.

2.4.12 Review limitations for SQ1 and SQ2

The limitations of the map may originate from: 1) the search strategy; and 2) bias in the pool of studies found. We will address both types of limitations consecutively.

Our searches were conducted in a limited set of languages due to the focus of the BONUS RETURN project on the Baltic Sea Region and European contexts and available language skills in the review team. However, searches in other languages (such as Spanish, French, Russian or Chinese) would have probably produced a more extensive evidence base. These additional searches could be easily conducted with more resources. Given the scope of the SQ2 and the focus on boreo-temperate zones, a geographical bias towards developed countries can be noticed in our evidence base (cf. **Figure 18**). Studies from some large countries covering large part of the relevant climate zones for SQ2 (such as Russia) were absent in the evidence base. Moreover, we have limited our search to last 6 years, but future work can capture research published before 2013 for a more extensive evidence base.

Studies examining effectiveness of struvite precipitation included substantial random variation that could not be explained with the available data. The variation might originate from differences in seed crystals, variation in total solids or nutrient concentrations in the inlet. However, the evidence base was neither large enough nor homogeneous enough to investigate the influence of these parameters on the outcome. The variation, however, did not jeopardise the conclusions regarding the impact of pH and Mg to limiting reactant on the outcome, since these relationships were obvious. In addition, and due to lack of data, the predicted removal in some areas of our model was uncertain.

The studies on ammonia stripping displayed high heterogeneity with respect to what parameters and outcomes were reported, even though all the studies applied ammonia stripping to the liquid phase of anaerobic digestate. For example, liquid to gas flow ratio is only presented in three out of seven studies in the final evidence base. When it comes to temperature, some authors present only the air temperature, others present only the liquid temperature, and others still present only the temperature of the reactor. There are also differences in whether the authors present removal or recovery as a measure of outcome, or how they define the two. Moreover, authors apply different techniques for measuring N concentrations, i.e. total N, ammonia, TAN and Kjeldahl N. All these issues contribute to the incomparability of the individual study findings, where synthesis of such evidence base is hard.

In fertilizer effectiveness studies, it is common to carry out pot experiments before conducting largerscale field experiments in real-world conditions. The evidence base for struvite is clearly biased

towards pot-scale studies, whereas the value of field-scale studies is often considered higher. Another potential issue is a low number of studies on heavy soils such as clays as well as on acidic soils. Both clayey soils and soils with low pH are abundant in some parts of the Baltic Sea Region. Although different crop groups were represented in the evidence base, some crops that are cultivated in the region of interest such as root/tuber crops (e.g. potatoes) and fruits were not included. Legumes, vegetables and oilseed crops were also under-represented.

The temporal scale of studies included in the evidence base was rather short. The great majority of studies did not exceed several weeks in length. Even sparse, field-scale studies were usually conducted over only one growth cycle. Only two field studies lasted for more than one season, whereas it is well known that residual fertilization effects should be also considered in the case of P fertilization.

The evidence base for ammonium sulphate consisting of only one study was too small to draw conclusions about bias in the pool of studies. The authors of papers in which pot experiments were reported frequently conclude that field-scale validation of recovered struvite should be one of the important future directions.

2.5 Review conclusions

Here, we describe implications for policy, management and research of the review findings for each secondary review question separately.

2.5.1 Implications for Policy/Management from SQ1

When performed under the right conditions, both struvite precipitation and ammonia stripping seem to be effective techniques for the recovery of nutrients from the liquid phase of digestate. In a wastewater treatment setting, both methods could be applied to the liquid phase of digested wastewater sludge in order to produce a fertilizer product that contains less contaminants than the sludge itself. Note, however, that the potential yield would then be limited to the amount of nutrients in the liquid phase of the digestate. In an agricultural setting, both techniques could be applied to the liquid phase of digested manure in order to produce a fertilizer product that is easier to transport than manure. This could potentially be used to remediate soils with high nutrient contents, thereby decreasing nutrient flows from agricultural land to surface waters.

2.5.2 Implications for Research from SQ1

The evidence base for ammonia stripping was considered too heterogenous to be quantitatively synthesised. Therefore, to ensure comparability among future research, we call on authors to present at least the following parameters when analysing effectiveness of ammonia stripping process:

- pH of the inlet
- Liquid to air flow ratio
- Temperature of both the liquid and the air in the inlet
- Concentrations in terms of ammonia or TAN as well as total nitrogen
- Both removal in the stripping column as well as recovery in the acid scrubber

For struvite precipitation, the quantitative synthesis showed that the maximum efficiency of the process is achieved around pH 9.5. Mg to limiting reactant ratio was found to have a positive effect on removal up to a ratio as high as 4 to 1. However, dosing Mg in excess may be expensive, and it should be noted that relatively high efficiencies were achieved at a ratio as low as 1 to 1 as well. Although the effects of pH and Mg to limiting reactant ratio were clear, the model developed could not accurately predict removal based on these two parameters alone. This could be due to random variation between



experiments, but it may also be due to the influence of other process parameters. Although the evidence base was deemed too limited to draw conclusions regarding other parameters, it is noted that other parameters may exert an influence on the outcome and as such could be interesting to investigate further. These include Mg source, total solid content, initial concentrations of reactants and temperature.

2.5.3 Implications for Policy/Management from SQ2

Struvite, most frequently recycled from agricultural waste or sewage sludge, seems to be a suitable phosphorus fertilizer for a diverse set of crops, predominantly including cereals and grasses. Treatment of soils with struvite usually results in comparable yields and P uptake by plants as treatment with conventional, mineral P fertilizers. Even if the direct effect of struvite on yield and P uptake is slightly worse than that of mineral fertilizer, which was the case in some studies, the benefit of using struvite should offset this difference in the long-term. Specifically, its wider use would reduce the reliance on a finite resource of phosphate rock, help close the nutrient loop and advance a shift towards circular economy in agriculture and wastewater sectors, as well as potentially reduce manure-based environmental pollution. However, this review did not consider the economic aspect of the shift from mineral to recycled fertilizers in agriculture.

The evidence base for ammonium sulphate was too small to formulate implications for policy and management on this fertilizer.

2.5.4 Implications for Research from SQ2

What seems to be currently missing in the context of evaluation of struvite as fertilizer is its more profound validation in terms of both spatial and temporal scale. Due to a strong bias towards pot-scale studies, field-scale validation is particularly important, but also a longer time span of conducted experiments. Future studies could more widely investigate soil P content in struvite treatment experiments as it is a highly under-represented outcome compared to yield and P uptake.

In a more general sense, struvite is not the only bio-based fertilizer that is worth analyzing. A recent systematic map of ecotechnologies in agriculture for the recovery and reuse of carbon and nutrients in the Baltic Sea Region maps [22] showed that compost, biogas residues and biochar were the most frequently used bio-based fertilizers. As such, a systematic review that would compare the effectiveness of these different recycled products would be also of high interest.

The evidence base for ammonium sulphate was too small to formulate implications for research.

2.6 List of additional files

Additional file 1. ROSES form for systematic review (https://drive.google.com/file/d/1OAc6_R53b0cPyFfN_IL5g3hWcuoX9WG5/view?usp=sharing) Additional file 2. Search strategy and results (https://drive.google.com/file/d/1wQgMPnzVZo9cjJBaSwE4PbbBPIV_CmgB/view?usp=sharing) Additional file 3. List of unobtainable articles (https://drive.google.com/file/d/1GYCc4ITxzpKL6_g_JNu8OdJ1XzMTcvZF/view?usp=sharing) Additional file 4. List of excluded articles at full text screening (https://drive.google.com/file/d/1h7iscslu8wor51cvSETCEDyS1DLoH6TS/view?usp=sharing) Additional file 5. Critical appraisal of study validity (https://drive.google.com/file/d/1Vt_gsZcZA6NnD9RWk_K7H62fHRcPCVP7/view?usp=sharing) Additional file 6. Data extraction (https://drive.google.com/file/d/1dXRYEw49e-Jpf_ONQ33p9JmpYva1iFM1/view?usp=sharing)



Additional file 7. Narrative tables

(https://drive.google.com/file/d/1Bpr2tyQFjQDGb_pSvxAG7Tdz2IBqrzcG/view?usp=sharing)

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