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EU-compliant wastewater recycled phosphorus: How much national cereal demand can it meet?

Juan Serrano-Gomez [a,b,*], Geneviève S. Metson [d,f], Tina-Simone Neset [e], Jakob Santner [g], Ludwig Hermann [b], Matthias Zessner [c]

[a] Department of Civil Engineering, TU Wien, A-1040, Vienna, Austria
[b] Proman Consulting GmbH, 2214, Auersthal, Austria
[c] Institute for Water Quality and Resource Management, TU Wien, A-1040, Vienna, Austria
[d] Ecological and Environmental Modelling Division, Department of Physics, Chemistry and Biology, Linköping University, Linköping, 581-83, Sweden
[e] Department of Thematic Studies-Environmental Change, Centre for Climate Science and Policy Research, Linköping University, 58183, Linköping, Sweden
[f] Department of Geography and Environment, Social Sciences Centre Rm. 2403, The University of Western Ontario, London, ON, N6A 5C2, Canada
[g] Institute for Plant Nutrition, Justus Liebig University Giessen, 35392, Giessen, Germany

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ABSTRACT

Finding alternative phosphorus sources is imperative to address negative environmental and societal impacts caused by its current inefficient use. However, the direct use of phosphorus in sewage sludge in agriculture is controversial. This paper uses Denmark, Germany, and Spain as case examples to assess relevant legislation and boundary conditions in agricultural production to identify opportunities and barriers for the utilisation of recycled phosphorus from wastewater in agriculture on a regional level. Only five out of 22 phosphorus recycling technologies considered were in full compliance with legislation across all three countries, and these five were then assessed for their potential to supply phosphorus to major crops within countries. We considered the application of technologies across four scenarios: 1) struvite; 2) vivianite as iron supply; 3) vivianite for calcium phosphate precipitation; and 4) ashes for calcium phosphate precipitation. The most suitable scenario identified for Denmark was vivianite for calcium phosphate precipitation, whereas in Spain vivianite as iron supply was identified as most suitable, and ashes for calcium phosphate in Germany. We found that in 2018, the potential phosphorus supply from recycling technologies was on average 0.38, 0.29 and 0.05 kg of phosphorus per capita for Danish, German, and Spanish regions. These quantities could meet 9.1, 21.7, and 10.0 percent of the phosphorus required to produce major cereals in each country (specifically wheat, barley, and rye). Given current legal constraints, wastewater treatment plant connections and agronomic context, the potential contribution of recycled phosphorus is non-negligible in many sub-national regions. Still, to access the full potential of phosphorus circularity clear product specifications and transport and logistics among regions will be necessary.

1. Introduction

The inefficient use of mineral phosphorus (P), and organic waste products high in P, has led to problematic losses of this nutrient into water bodies (Panagos et al., 2022a), and to landfills as final sinks (van Dijk et al., 2016). P over enrichment of water bodies (i.e., eutrophication) can lead to biodiversity loss, toxic cyanobacterial species occurrence, and even oxygen depleted zones affecting fisheries, recreation, drinking water, and ecosystem integrity (Preisner, 2023). Such anthropological disturbances of the P biogeochemical cycle to supply food, as well as unequitable access to mineral P sources, or phosphate rock (PR), presents an unprecedented sustainability challenge (Chowdhury et al., 2017; Sandström et al., 2023). Hence, there is interest in finding alternative and more sustainable sources than PR. P-containing waste streams such as municipal wastewater are a viable option, but such streams have yet to be fully integrated into fertiliser markets.

Municipal wastewater is defined as the collected water composed of human excreta and waste originated from toilet use, cleaning, washing and cooking, that contain resources such as nitrogen, P, metals and proteins that could potentially be recovered (Ostermeyer et al., 2022). In the European Union (EU), there still is a large untapped potential for reusing P from wastewater. In 2018, the EU imported 1.26 million t of

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* Corresponding author. Department of Civil Engineering, TU Wien, A-1040, Vienna, Austria.
E-mail address: juan.serrano@student.tuwien.ac.at (J. Serrano-Gomez).

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mineral P and used 1.11 million t of P in agriculture (FAOSTAT, 2022). The largest losses have been attributed to the consumption sector (0.66 million t P in 2005; van Dijk et al., 2016). The consumption sector is composed of plant and animal-based food and non-food products, such as fibres, tobacco, skins/hides, pet food, detergents, wood and paper (van Dijk et al., 2016). The largest share of the total losses from consumption was communal sewage sludge, a by-product of wastewater treatment, with 34.6% (van Dijk et al., 2016). For this reason, the use of P from wastewater sludge as an alternative source for crop production has been widely studied as one of the promising pathways for substituting PR with P recycling (Kanteraki et al., 2022; Sichler et al., 2022).

Currently, direct sewage sludge reuse on cropland is allowed in all EU Member States if it complies with national regulations. EU countries adopted the *Sewage Sludge Directive* EU 86/278/EEC while some countries introduced stricter limits for certain heavy metals (see Table A1) (Gianico et al., 2021). Even though untreated sewage sludge can contain between 0.35% and 1.22% of total P from the total solids, it could also contain pollutants such as heavy metals, organic contaminants, antibiotic-resistant pathogens and microplastics (Egle et al., 2016; Kanteraki et al., 2022). In Europe, approximately 6 kt of microplastics are added every year to soils from sewage sludge (Kanteraki et al., 2022). In contrast, some recycled P products (e.g., from incinerated sewage sludge) have shown lower heavy metal and no organic contaminant accumulation by fertiliser application, due to elimination during the incineration process and post-treatment, as opposed to direct sewage sludge application (Weissengruber et al., 2018), although incineration requires more energy (Egle et al., 2016). Therefore, possible soil pollution from direct sewage sludge reuse in agriculture may be avoided using more processed recycled P products.

Among the EU countries, the conditions for both legislation and infrastructure can greatly differ, not to mention P use in agriculture, which depends on soil properties and type of products. To explore these conditions more fully, we selected three case study countries: Denmark, Germany, and Spain. All three countries are subject to EU regulations but differ in terms of national laws, infrastructure, and agricultural production. Germany has a more stringent regulation when reusing sewage sludge for agriculture and has been implementing mono-incineration as a preferred disposal route for its management, whereas Spain and Denmark still present more sludge reuse in agriculture (Collivignarelli et al., 2019; Mannina et al., 2023). We selected these three countries to identify opportunities to increase the utilisation of products derived from P from wastewater in agriculture at a regional level. Specifically, this study aims to address three questions:

i. What are the current legal and infrastructural preconditions for the implementation of wastewater P recycling technologies for eventual use on agricultural soils?
ii. What P recycling technologies are most compatible with current wastewater treatment plant infrastructure and regional soil properties?
iii. What is the share of P demand in agriculture that can be potentially supplied from P recycling technologies?

2. Material and methods

We first identified relevant legislation for each selected country and the EU regarding P recycling for agricultural use (see Figure A1). Subsequently, we characterised the P recycling technologies from wastewater according to their P-containing stream within the wastewater treatment process and the final recycled P product. Regarding fertiliser legislation, we evaluated if the P product resulting from each technology, listed in relevant European P platforms (e.g., European Sustainable Phosphorus Platform or ESPP, and Nutrient Management and Nutrient Recovery Thematic Network or NUTRIMAN), complied with P content requirements and pollutant limits. The P products that complied with legislation were then selected to represent four implementation scenarios, and we determined the potential P supply of each scenario across each country according to the P inflow of wastewater reported in the Waterbase database from the EU *Urban Waste Water Treatment Directive* (UWWTD). Moreover, we estimated the potential P demand of selected crops based on fertilisation recommendations for soil P maintenance and build-up specified for each crop and region, depending on their soil pH and P content provided by the Land Use and Coverage Area Frame Survey (LUCAS) topsoil database. Lastly, to calculate the coverage of P requirements for the selected crops by recycled P we divided the potential P supply of the most suitable product by the potential P demand.

2.1. Legislation for phosphorus recycling

The relevant legislation that involves the use of recycled P technologies for agriculture is based on how P is managed in wastewater and derived waste streams, and standards that fertilisers must comply with. The legislative map, both national and EU level, is divided into sections of the process (Collection, Treatment, Outlet and Usage), and we summarised their requirements per section (see Table A2). The first two regulate how wastewater is to be collected and treated. The aim of treatment is that the outlet discharges 'clean' water, and this process generates sewage sludge, which contains most of the P removed from the wastewater during the process. Both the outlet water and the sewage sludge produced are subject to management regulation. In addition to collection, treatment, and outlet sections, the use of recycled P products is regulated under fertilisers and soil conditioners ordinances.

Wastewater legislation throughout the EU is based on the UWWTD EU 91/271/EEC with its later 1998 amendment EU 98/15/EEC, where the objective is to preserve the environment and protect people from risks associated with pollution. In this directive, wastewater from agglomerations of over 2000 population equivalent (p.e.) must be collected and treated with primary and secondary treatment; agglomerations over 10,000 p.e. must have advanced wastewater treatment for designated areas with a high risk of eutrophication, or sensitive areas (Garrone et al., 2018). Denmark identifies all their surface water bodies as sensitive areas, whereas Germany and Spain only designate some areas as sensitive (Preisner et al., 2020). As a consequence, the discharge P limits are less stringent for certain German and Spanish water surfaces and do not ensure at least 80% P removal (Table A2).

Sewage sludge management plays an important role in P recycling, and the legislation regarding its management and reuse is based on the EU 86/278/EEC *Sewage Sludge Directive* (Hukari et al., 2016). Each country (or even within individual regions) has different thresholds for sludge reuse in agriculture (see Table A1). The use of recycled P products (not sludge) in agriculture is bound to the *Fertilising Products Regulation* EU 2019/1009 legislation. This legislation contains minimal P content requirements for phosphate fertilisers and heavy metal limits depending on the type of P fertiliser (see Table A3 and Table A4). The aforementioned legislative requirements in the selected countries and the EU were the guidelines to evaluate the compliance of recycled P products studied.

The use of recycled P products as fertilisers is regulated by the *Fertilising Products Regulation* amendment of 2019 EU 2019/1009, where under Component Material Categories 12 and 13 (also present in the German *Fertiliser Ordinance* of 2012), which included recycled P products from sewage sludge are covered as "precipitated phosphate salts and derivates" (EU, 2019/1009; DüMV, 2012). Moreover, precipitated phosphates have been approved to be used in organic farming in the latest amendment of products and substances for use in organic production EU 2021/1165 (EU, 2021/1165). Considering the legal requirements, the use of recycled P products in agriculture is a more sustainable alternative to safely supply P.

2.2. Phosphorus recycling technologies and products

In the wastewater treatment process (Fig. 1), there are several

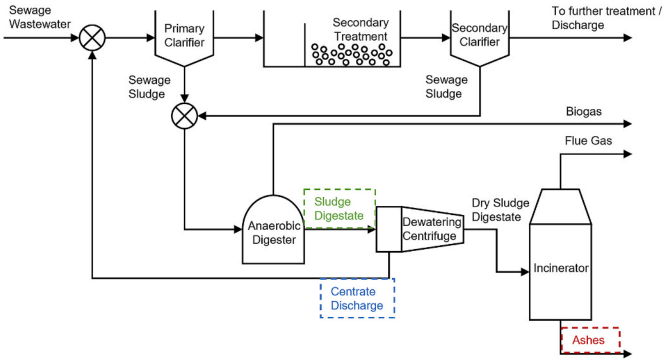


Fig. 1. Process outline of a typical wastewater treatment plant (adapted from Egle et al. (2015)).

streams where P can be recovered (Egle et al., 2015). In this study we focus on three main streams that contain considerable amounts of P: centrate (blue), sludge digestate (green) and sludge ashes (red; colours refer to Fig. 1).

Typically, after sewage sludge is collected and thickened via sedimentation, it is biologically stabilised via anaerobic digestion, while in parallel biogas is produced through the degradation of organic matter by microorganisms in the absence of oxygen. This stabilised sludge, or sludge digestate, is dewatered to reduce the amount of sludge to be disposed and results in a liquid discharge and solid phase (Egle et al., 2015). The liquid discharge (or centrate discharge in blue – see Fig. 1), is recirculated back to the sewage wastewater inlet at the start of the wastewater treatment process. Nevertheless, centrate contains ions that facilitate struvite formation and result in pipeline scaling, which could cause high maintenance cost (Molinos-Senante et al., 2011). On the other hand, the solid phase (or dewatered sewage sludge), due to disposal costs and reduction of methane emissions in landfills, is monoincinerated to ashes when the infrastructure is available.

Technologies to recycle P from different sources within wastewater treatment and their process efficiencies have been widely reviewed (Egle et al., 2015; Santos et al., 2021). P recycling via struvite precipitation from the centrate is one of the most implemented technologies worldwide with around 100 full-scale operations (Jupp et al., 2021). One of the main reasons is that there is a considerable economic benefit of preventing struvite formation in pipes, and also can be directly used as a fertiliser. Instead, P precipitation using iron salts results in vivianite formation, however, it has limited use as fertiliser due to low solubility in neutral soil pH. Consequently, vivianite precipitation could lead to two P-recycled products: vivianite as iron phosphate (Fe–P) for direct application to soils prone to chlorosis or calcium phosphate (Ca–P) through alkaline post-treatment (Prot et al., 2019). Lastly, it is possible to recycle P as Ca–P through acid attack and wet chemical treatment of the sludge ashes (Herzel et al., 2016).

To increase the sustainability of fertilisers used in agriculture, P-recycled fertilisers, such as struvite and Ca–P, have been compared to traditional PR products (e.g., triple superphosphate or single superphosphate) in terms of agronomic P efficiency (Raniro et al., 2022). Recycled P products described in this paper are highly insoluble in water, but present high solubility in neutral and alkaline ammonium citrate, which is commonly used to evaluate the fertilising quality of a non-water soluble P product (Herzel et al., 2016). Several studies on recycled P products have demonstrated slightly lower or similar fertilising efficiency as PR-derived products for different types of crops such as wheat, maize, rye, among others, and soils (Johnston and Richards, 2003; Oliveira et al., 2019). In contrast, the fertilising efficiency of vivianite is limited to specific soils such as calcareous soils ($pH > 7.5$), in which additionally to P it provides iron for its deficiency in plants, or iron chlorosis (Díaz et al., 2010).

Given that the scope of this study was within the EU, the P-recycled products from wastewater found in the catalogue from the ESPP (European Sustainable Phosphorus Platform, 22AD) and NUTRIMAN ("Nutrient Management and Nutrient Recovery Thematic Network," 2022) were evaluated according to their product characteristics and their compliance with the EU and national fertiliser legislation. First, the technologies that provided complete information were compared to national and EU fertiliser legislation (heavy metal limits and minimum P content) and characterised according to the generic P compound (e.g., struvite, vivianite, calcium phosphate). Subsequently, for those recycling P products that complied with legislation in Denmark, Germany, Spain and the EU the potential P supply was calculated according to the regional conditions such as wastewater treatment infrastructure, soil properties and agricultural practices (see Table A5).

2.3. *Conditions for the selected countries*

In this study, the regions of Denmark, Germany, and Spain were studied for 5 specific factors that are directly related to the amount of P to be recycled from wastewater, and the P demand for wheat, barley, and rye production reported in 2018 (Table A5):

- Population
- Cropland area
- Soil properties
- Wastewater treatment infrastructure
- Sewage sludge incineration

Population and sewage sludge incineration data were obtained from Eurostat (except for Denmark – "StatBank"), while cropland area data was taken from national accounts: "StatBank" (Denmark); "Ministry of Food and Agriculture – *Bundesanstalt für Landwirtschaft und Ernährung*" (Germany); and "Ministry of Agriculture, Fisheries and Food – *Ministerio de Agricultura, Pesca y Alimentación*" (Spain). The number of wastewater treatment plants (WWTPs) and total P in influent data was obtained through the Waterbase of UWWTD, and soil properties, such as pH and P content in soil (Olsen method), from LUCAS topsoil database (Bundesanstalt für Landwirtschaft und Ernährung, 2019; Eurostat, 2022a, 2022b; Fernandez-Ugalde et al., 2022; Ministerio de Agricultura Pesca y Alimentación, 2022; StatBank Denmark, 2022a, 2022b, 2022c; Urban Waste Water Treatment Directive, 2018).

Only WWTPs from the Waterbase that reported P removal were considered in this study to assess their potential P supply, given that it ensures the availability of necessary P sources for recycling technologies like centrate, digested sludge, or sludge ash. Those plants were assumed to have the infrastructure of a typical WWTP (see Fig. 1), and at least 80% of the total P in the influent is removed according to the UWWTD 98/15/EEC.

Although the P inflow of WWTPs in Castilla-La Mancha (Spain) was reported, the mass of the P inflow of 264 plants was between 10 and 1000 times higher than WWTPs with similar conditions and possibly an anomaly in the database, thus it was assumed to have a standard value of 1.5 g P $p.e.^{-1}$ day^{-1} (Zessner and Lindtner, 2005). It was also the case of several WWTPs with P removal (213 out of 813) of Spain that did not report P inflow and it was assumed the same standard value to assess their potential. Additionally, the Spanish regions of Ceuta and Melilla did not report data at both Waterbase and LUCAS topsoil databases and were consequently excluded from the assessment.

2.4. *Scenarios of phosphorus recycling*

From the recycled P technologies in the ESPP and NUTRIMAN catalogues that complied with European and national fertiliser legislation mentioned previously (see section 2.3), three recycled P products (struvite, vivianite and Ca–P) were eligible for fertiliser implementation. Moreover, according to the soil pH characteristics of the selected countries, we identified that vivianite could be used directly as Fe–P or as Ca–P fertiliser depending on the soil pH. Consequently, the potential P supply of struvite, vivianite and Ca–P derived into 4 possible scenarios: struvite, vivianite as Fe–P, vivianite as Ca–P and ashes for Ca–P (Table 1).

To consider the established scenarios, four assumptions were crucial to estimate the potential supply:

1. Removal of P by chemical precipitation of vivianite requires the introduction of iron salts that precipitate phosphates present in wastewater influent (Prot et al., 2019). P bound to iron is not available in the centrate after dewatering digested sludge, and thus would make struvite precipitation unviable. Therefore, the first assumption is that the implementation of vivianite restricts P available for subsequent struvite precipitation, and those were considered as different scenarios.
2. It was also assumed the lowest recycling efficiency from Table 1 to provide a minimum expected P supply with the mentioned P recycling technologies.
3. Additionally, the regional soil pH was also obtained via the LUCAS topsoil database 2018, and for calcareous soils ($pH > 7.5$) vivianite for Fe–P was considered more suitable than vivianite for Ca–P.
4. The percentage of dewatered sewage sludge that is incinerated as a disposal route was assumed to be fully monoincinerated and used to determine the potential P supply for ash-based recycling technologies (ash for Ca–P). The remaining percentage of other disposal routes was used to calculate the potential from the scenarios of struvite, vivianite as Fe–P and vivianite as Ca–P.

Finally, all scenarios were evaluated for each region within the three countries, and the scenario with the highest potential P supply was selected as the most suitable (see Table A5). To obtain a comparing unit between regions, we normalised the potential P supply and demand with the population of 2018.

2.5. Phosphorus supply in regions

For each recycling scenario the potential P supply is calculated, and described by Eq. (1):

$$m_{P_{supply,i,j}}(kg\ P\ per\ capita) = \frac{n_{i,j}}{n_{total,j}}\,\frac{m_{P,j}}{u_j}\,\eta_i \tag{Eq. 1}$$

Where the potential supply ($m_{P_{supply,i,j}}$) for i scenario and j region from each country expressed in kg P per capita; number of WWTPs per scenario and region ($n_{i,j}$); the total number of WWTPs per region ($n_{total,j}$); annual wastewater P inflow per region expressed in kg P ($m_{P,j}$); the total population of j region (u_j); and recycling efficiency from P influent of WWTPs for i scenario (η_i).

2.6. Phosphorus demand in regions

The assessment of the potential P demand was conducted by analysing the fertiliser recommendation for the crops that are the most relevant in terms of cropland, as well as soil properties such as P content (by Olsen method, or Olsen P) and pH. A considerable part of the grain cropland area in Denmark, Germany and Spain is used to produce wheat, barley and rye (between 70 and 95%). Hence, those crops were considered to analyse a potential P demand that could hypothetically be supplied by P-recycling technologies in the three countries.

One of the objectives of tackling the P challenge, is to increase the P use efficiency in agriculture, although there has been an accumulation of P in agricultural soils (Panagos et al., 2022a). It has been estimated that in the EU there is on average a surplus of 0.8 kg P ha^{-1} year^{-1} in agricultural soils (Panagos et al., 2022a), and could lead to water pollution. Therefore, there is a need to implement a more efficient estimation of P fertiliser requirements (P_{rate}), and one possible way is based on two components: build-up and maintenance (Eq. (2)).

$$P_{rate,j} = P_{exported,j} + 10BD\,z\left(P_{Olsen,T} - P_{Olsen,S,j}\right) \tag{Eq. 2}$$

$$m_{P_{demand,j}}(kg\ P\ per\ capita) = \sum_{i=1}^{N} \frac{A_{i,j}\,P_{rate,j}}{u_j} \tag{Eq. 3}$$

The maintenance component or P exported of j region ($P_{exported,j}$) describes the amount of P taken away when harvesting (Recena et al., 2022). The build-up component is added to the maintenance component when soil P content of j region ($P_{Olsen,S,j}$) following Olsen P extraction in mg P kg^{-1}, is lower than threshold values ($P_{Olsen,T}$). These are the values above which no response in crop yield can be expected if P fertiliser is applied (Delgado et al., 2016; Recena et al., 2022). In case $P_{Olsen,S,j} > P_{Olsen,T}$, it is recommended for P fertilising rate to only consider the maintenance component ($P_{rate,j} = P_{exported,j}$). There was no significant effect of crop type on the $P_{Olsen,T}$ for most cereals studied which showed values between 8.1 and 17 mg P kg^{-1}, thus the average of 12.6 mg P kg^{-1} was considered (Recena et al., 2022). In addition, the average value for bulk density (BD) was 1.38 t m^{-3} for the LUCAS soil sample, while the soil depth (z) at the LUCAS soil sample was 0.2 m for cropland (Fernandez-Ugalde et al., 2022; Recena et al., 2022).

The potential P demand ($m_{demand\ P,j}$ – see Eq. (3)) for j region was estimated by the sum of cropland area ($A_{i,j}$) of j region and i crop in ha; the $P_{rate,j}$ of j region according to crop production statistics provided by ministries from each country in 2018; and divided by the population (u_j) of each region to normalise the potential P demand.

2.7. Potential phosphorus demand covered by potential phosphorus supply

We assessed the capacity of recycled P products to cover the potential P demand for the selected crops by diving the potential P supply by the potential demand. Nevertheless, regions in Germany such as Berlin, Bremen and Hamburg, as well as Balearic Islands in Spain, did not present information on crop production (Table A5), and it was not possible to establish their potential P demand covered by potential P supply. Additionally, some regions showed over 100% coverage of their potential P demand by P recycling (see Table A6) and could trade with neighbouring regions with lower coverage.

This potential P supply could help increase the potential demand coverage of regions with higher potential P demand per capita.

Table 1
Scenarios for P recycling technologies by source, recycled P product and their efficiency from total P from WWTP influent.

Scenario	P source	P recycling technology	Recycled P product	Recycling efficiency from wastewater influent (η_i)	Reference
Struvite	Centrate	Chemical precipitation of struvite	Struvite	10–35%	(Egle et al., 2015; Santos et al., 2021)
Vivianite as Fe–P	Digested sludge	Direct use of precipitated vivianite	Vivianite	~60%	(Prot et al., 2019)
Vivianite as Ca–P	Digested sludge	Alkaline treatment of precipitated vivianite	Calcium phosphate	~54%	(Prot et al. 2019)
Ash for Ca–P	Sludge ash	Thermochemical treatment of sludge ashes	Calcium phosphate	60–85%	(Egle et al., 2015; Santos et al., 2021)

Consequently, the supply potential and demand of the region with a surplus were added to the neighbouring region with the highest potential P demand. As an example, in Spain, the Valencian Community could supply over 100% of its potential P demand, whereas Castile-La Mancha, its neighbouring region with the highest demand per capita, could cover around 5% (Table A7). In this light, both potential P demand and supply were combined to improve the potential P demand covered by recycled P. As opposed to those cases, the potential P supply of regions that did not report crop production information was directly added to the neighbouring region with the highest potential P demand. We estimated a separate coverage assessment, including the regions that had a surplus or did not present crop production information (Fig. 4b).

2.8. Limitations of the method

In this study we used the mentioned methods to estimate both potential P supply and demand that rely on assumptions, that if not met, might lead to inaccurate quantification of potential for P recycling on a regional level.

For instance, in the case of potential P supply estimation, we identified that some of the available information regarding P inflows in wastewater treatment was abnormal, while sludge disposal routes are only available at a country level. One assumption we considered in the calculation of potential P supply was that the percentage of sludge incineration and agriculture reuse was identical in all regions, and it might lead to inaccurate potential estimations of the technologies studied. Not only the information provided by Eurostat is unclear on the sludge disposal methods but also does not supply detailed data on a regional level, and it is also mentioned by Anderson et al., 2021. Therefore, a more detailed report of sewage sludge amounts and specified disposal routes on a regional level could lead to higher accuracy in the assessment of potential P that could be supplied for agriculture.

In addition, the estimation of potential P demand was based on soil P content using Olsen P reported in the LUCAS topsoil database. Although Olsen P is one of the most widely used soil P tests for non-acidic soils, it does not predict accurately available P on soils with high chemical property variation (Delgado et al., 2010). Thus, different extraction methods should be used to more accurately predict soil P in other types of soils to provide better fertiliser management (Delgado et al., 2010).

3. Results & discussion

3.1. Phosphorus recycling technologies and regional legislation

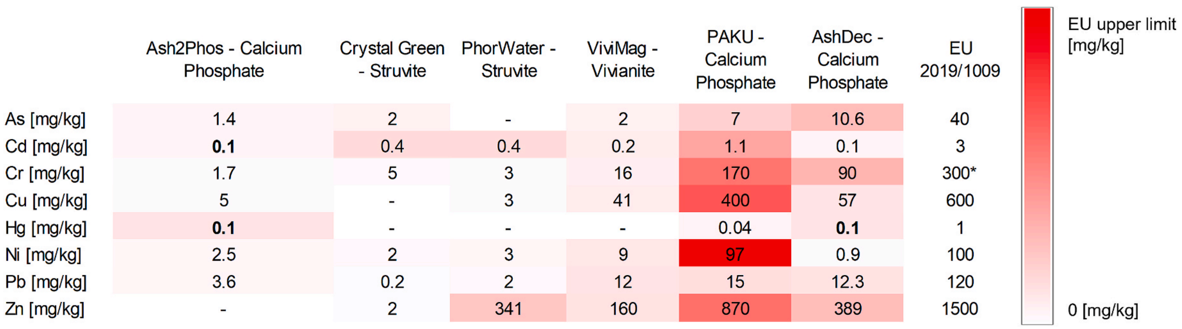
Only six P recycling technologies out of the 22 reviewed provided enough information on the physicochemical properties of end products (Table A4), namely Ash2Phos® (Ca–P), ViviMag® (vivianite), Crystal Green® (struvite), PhorWater® (struvite), PAKU® (Ca–P) and AshDec® (ashes) (Table A8). Most technologies listed in the ESPP and NUTRIMAN databases did not explicitly provide product information, like chemical composition, necessary to analyse their compliance with the current fertiliser legislation. The only heavy metal not specified for any of the reviewed technologies was chromium(VI) which has a limit of 2 mg kg^{-1} in the EU, Denmark and Germany, and undetectable by any official method in Spain.

In general, the composition of the two struvite technologies was similar, and both had lower heavy metal content compared with the ash-based technologies (see Fig. 2). Only PAKU® technology was not in full compliance with the current fertiliser legislation across the three countries and the EU. In the case of this ash-based technology, not only the P content was lower (4.7% total P) than the minimum required by EU fertiliser legislation (7% total P) but also the nickel (Ni) content was higher than permissible limits in Germany, as well as higher zinc (Zn) content than class B labelling limit in Spanish legislation (Real Decreto 506/2013, 2013; DüMV, 2012) (see Table A4). Additionally, AshDec® technology presented a Cr and Zn content higher than the class B labelling limit according to the *Fertiliser Products Royal Decree* in Spain, although, it could still be used. This significant variation of most heavy metals, in this case Cd, Cu, Ni, Cr and Zn, is explained by the fact that there is a high variation of substance content among the different wastewater influents depending on the source (Herzel et al., 2016). The variation could happen for P like in the case of PAKU®, which was under the EU minimum P content. In contrast, Ash2Phos® showed a similar P content to the commonly used PR fertiliser triple super phosphate (~20% of total P) (Cabeza et al., 2011).

The five technologies we retained for further investigation (Ash2Phos®, Crystal Green®, PhorWater®, ViviMag® and AshDec®) complied with both EU and national current fertiliser requirements of minimal P content and pollutant limits. Nevertheless, at the moment of evaluating the potential use of recycled P products in agriculture, a complete and standardised characterisation of physicochemical properties is crucial to compare those with commonly used mineral P fertilisers.

More ambitious goals are being set by the EU to achieve the *Sustainable Development Goals* and legislation is expected to change accordingly within the EU (El Wali et al., 2021). At the time of this study, a revision proposal of the UWWTD submitted in October 2022, aims to strengthen the P discharge limits from 1 to 2 mg P L^{-1} to 0.5 mg P L^{-1} (Proposal for a Directive of the European Parliament and of the Council concerning urban wastewater treatment, 2022). In the case of approval by the European Parliament and Council, the P content in sewage sludge could increase, thus providing more P recycling potential.

Some countries in the EU have also adopted measures to make P recovery mandatory and foster P recycling technologies. In 2017, Germany introduced the *Sewage Sludge Ordinance* amendment which will further restrict sewage sludge reuse in agriculture will be more restricted



	Ash2Phos - Calcium Phosphate	Crystal Green - Struvite	PhorWater - Struvite	ViviMag - Vivianite	PAKU - Calcium Phosphate	AshDec - Calcium Phosphate	EU 2019/1009
As [mg/kg]	1.4	2	-	2	7	10.6	40
Cd [mg/kg]	**0.1**	0.4	0.4	0.2	1.1	0.1	3
Cr [mg/kg]	1.7	5	3	16	170	90	300*
Cu [mg/kg]	5	-	3	41	400	57	600
Hg [mg/kg]	**0.1**	-	-	-	0.04	**0.1**	1
Ni [mg/kg]	2.5	2	3	9	97	0.9	100
Pb [mg/kg]	3.6	0.2	2	12	15	12.3	120
Zn [mg/kg]	-	2	341	160	870	389	1500

Fig. 2. Specified heavy metal content of the recycled P products in reference to the limits established by the *Fertiliser Products Regulation* (EU) 2019/1009. Values in bold reported the lower limit measured and values with dash were not specified. For specified zinc of PhorWater – struvite the upper limit of the range was considered (0.4–341 mg of Zn per kg of product). *The limit of total chromium is not specified by the *Fertiliser Products Regulation* (EU) 2019/1009 and the highest limit of Germany and Spain (300 mg of Cr per kg of product) was considered.

from 2029 onwards (AbfKlärV, 2017). Furthermore, in 2023 it is expected that WWTPs in Germany report a P-recycling plan for agricultural use.

This has been carving the path for more monoincineration of sewage sludge, which is expected to increase ash-derived technologies potential in Germany (Roskosch and Heidecke, 2018). However, P recycling lacks support in other countries within the EU from the perspective of agricultural and environmental legislation (Garske et al., 2020); more specifically, stricter pollutant limits in mineral fertilisers and sewage sludge, as well as requirements for removal of P from wastewater in useable forms. The unification of such changes in legislation on an EU level would foster a more sustainable and safer use of P to tackle the challenge.

3.2. *Regional potential phosphorus supply*

The most suitable scenario for Denmark was vivianite for calcium phosphate precipitation, whereas in Spain it was vivianite as iron supply, and ashes for calcium phosphate in Germany (Fig. 3a). More specifically, vivianite as Ca–P was selected for Denmark because the pH was lower than 7.5 in all regions. In contrast, most regions in Spain had pH higher than 7.5 and were assessed with vivianite as Fe–P, except Galicia, Principality of Asturias, Cantabria, Castile-Leon and Extremadura which had pH lower than 7.5 and vivianite as Ca–P was considered for potential P supply estimation. The potential P supply per capita per region ranged from 3×10^{-4} kg P per capita (Canary Islands, ES) to 0.46 kg P per capita (Central Region, DK), with an average of 0.38, 0.29, and 0.05 kg P per capita for Danish, German and Spanish regions, respectively (see Table A6).

In Denmark alone, the potential P supply was similar across regions and ranged between 0.30 and 0.46 kg P per capita (Southern and Central regions, respectively). In Germany, the region with the lowest potential P supply was Saxony (0.18 kg P per capita) and the highest Brandenburg (0.40 kg P per capita). Nevertheless, the difference in potential P supply between the lowest (Canary Islands with 3×10^{-4} kg P per capita) and the highest (Region of Murcia with 0.42 kg P per capita) regions in Spain was significantly greater than in the other two countries (Fig. 3a). In fact, 13 out of the 17 regions studied from Spain presented a potential P supply inferior to the lowest region in both Germany and Denmark (0.18 kg P per capita, Saxony, DE).

This observed low potential P supply can be explained by the fact that in 2018, the P removal reported from wastewater in Spain was around 34.8%, whereas in Germany and Denmark was higher (95.9% and 85.2%, respectively). According to the data reported in 2017, the percentage of WWTPs that complied with tertiary treatment (described with P and nitrogen removal) was 90.4%, 93.8% and 65.0% for Denmark, Germany and Spain, respectively (Urban Waste Water Treatment Directive, 2021). Moreover, it was estimated that in 2019 Spain had less percentage of the population in small agglomerations (<2000 inhabitants) from the total population (~30%) than Denmark (~50%) and Germany (~50%) (Pistocchi et al., 2022). Therefore, the potential P supply was directly affected by the number of WWTPs with P removal and not by the amount of P lost in the discharge of small agglomerations as permitted by the UWWTD.

The potential P supply obtained from the countries studied depended on the wastewater collection infrastructure and the sludge disposal route. The 2017 amendment of the *Sewage Sludge Ordinance* in Germany prohibits from 2029 onwards sludge (produced in >100,000 p.e. plants) reuse in agriculture and also requires P recycling (AbfKlärV, 2017). Moreover, in 2032, this prohibition will extend to >50,000 p.e. plants. As a consequence, there is a higher potential P supply by sludge ash-based recycled P products in Germany (Table A5). As opposed in Denmark and Spain, there is a tendency towards sludge reuse in agriculture instead of incineration, which leads to a higher potential P supply from sludge and centrate-based recycled P products (struvite and vivianite both Fe–P and Ca–P) than ash-based products (Ca–P). This way, most of the P content is removed from the sludge, thus leaving its reuse in agriculture less attractive for crop nutrition, albeit the value of

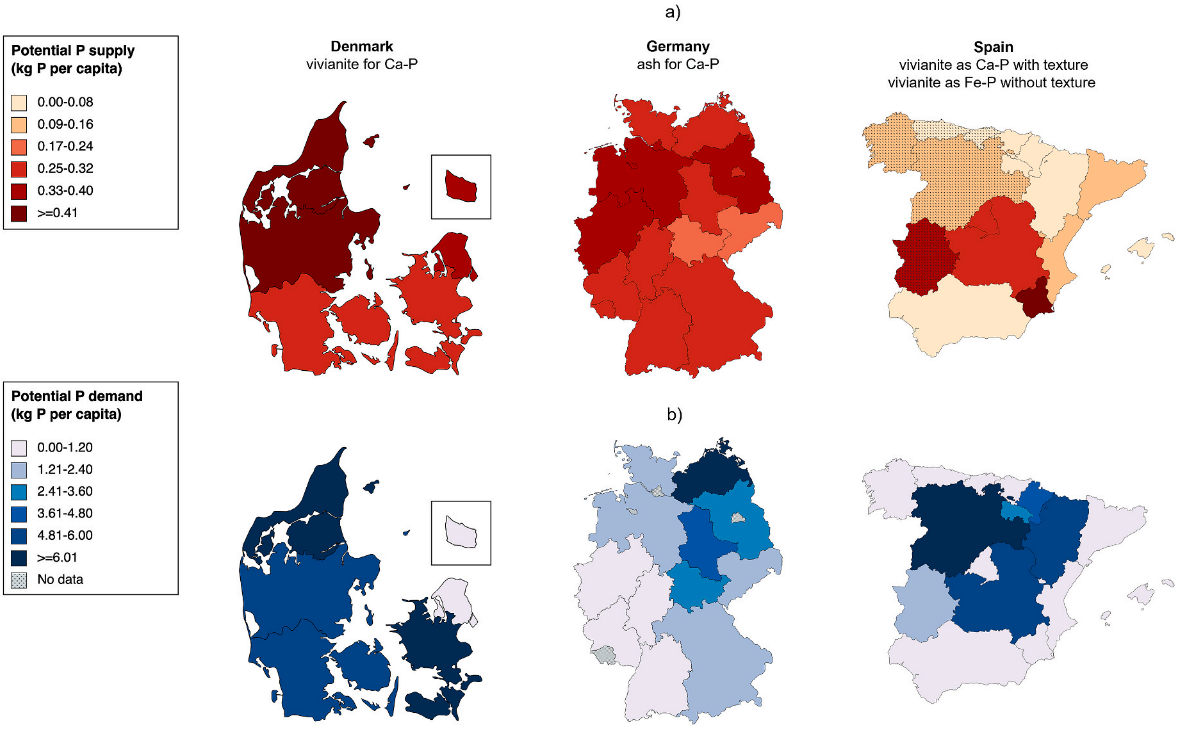


Fig. 3. Regional potential P supply (a) and demand (b) in 2018 in case study country regions. Values are expressed as kilograms of P per person (kg P per capita) and each county's potential supply is associated with the highest supply scenario: Denmark as vivianite as Ca–P, Germany as ash for Ca–P, and Spain as vivianite as Ca–P with texture and vivianite as Fe–P without texture – see Table A6 for all values. In the Canary Islands (Spain), the potential supply was 3×10^{-4} kg P per capita. No soil data was provided in grey regions and was not possible to calculate the potential P demand (including Canary Islands, Spain).

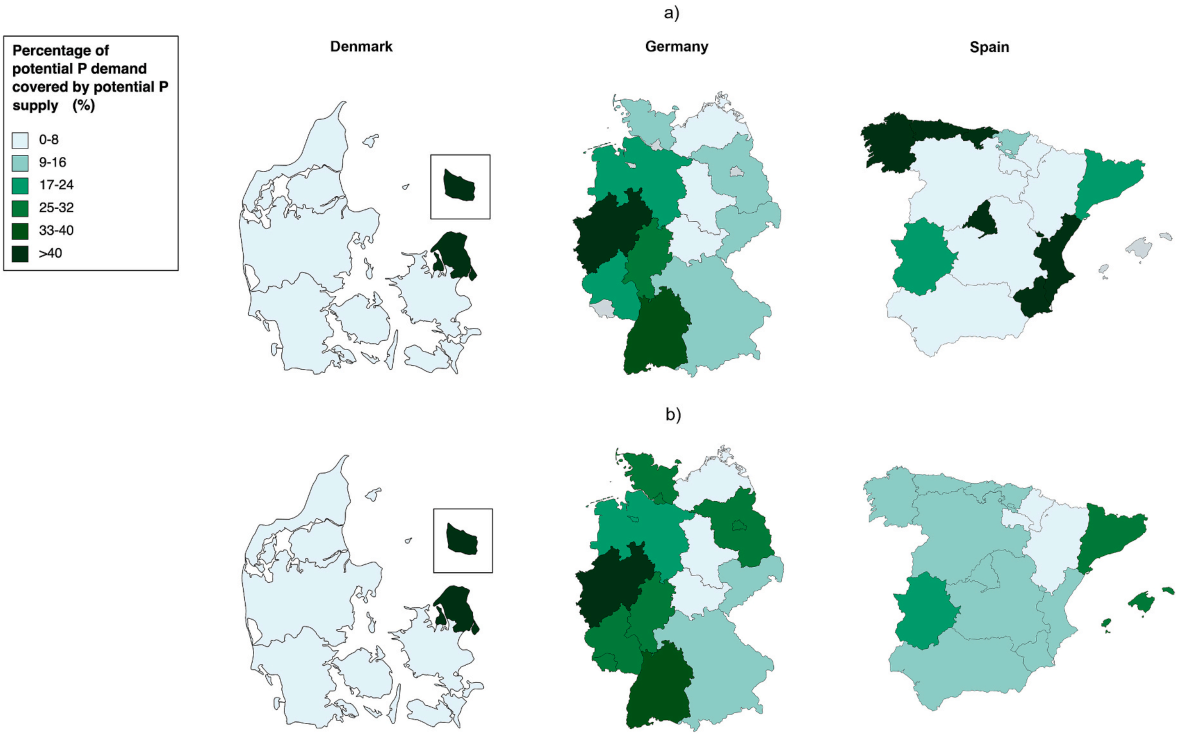


Fig. 4. Percentage of regional potential demand covered by potential P supply (a) and percentage of potential P demand covered if regions with a surplus (>100% coverage) combine their potential with neighbouring region (b) in 2018 for Denmark, Germany and Spain. Values of each county's potential supply are associated with the highest supply scenario: Denmark as vivianite as Ca–P, Germany as ash for Ca–P, and Spain as vivianite as Fe–P (except Galicia, Principality of Asturias, Cantabria, Castile-Leon and Extremadura as vivianite as Ca-P) – see Table A7 for all trade values.

organic carbon supply remains. Although, in Denmark and Spain both vivianite scenarios were higher than the ash-based scenario, struvite was significantly lower than both vivianite scenarios, mainly due to low P recycling efficiency from WWTP influent compared with other technologies. In previous studies (Santos et al., 2021), struvite is considered the most promising recycled P product from wastewater or sludge ashes. Our study indicates, however, that with low sludge incineration rates (e.g., <15% in Denmark) struvite from centrate has approximately the same potential P supply (0.06 kg P per capita) as Ca–P.

3.3. Regional potential phosphorus demand

From the regions that reported soil P content and pH in the LUCAS topsoil database, the potential P demand per capita average potential P demand was 4.93, 2.38 and 1.18 kg P per capita for Denmark, Germany and Spain, respectively, with a lowest of 9×10^{-5} kg P per capita (Principality of Asturias, Spain) and a highest of 9.94 kg P per capita (Castile-Leon, Spain) (Fig. 3b). In Denmark, except for Capital Region, the potential demand was over 5 kg P per capita, whereas only in German Mecklenburg-Vorpommern Region (6.58 kg P per capita) and Spanish Castile-Leon and Aragon (6.47 and 5.07 kg per capita, respectively) was surpassed. German regions such as Berlin, Bremen, Hamburg and Saarland, as well as the Canary Islands in Spain, did not present soil data to evaluate their potential P demand and were marked in grey.

The fertilisation strategy depends on the P exported component, which is based on crop species and yield. In the year 2018, the wheat grain yield was similar for Denmark and Germany (6.36 and 6.67 t ha^{-1}), but considerably lower in Spain (3.90 t ha^{-1}) (Baruth et al., 2019). For barley, Germany had the highest grain yield in the same year (5.77 t ha^{-1}), being higher than Denmark (4.53 t ha^{-1}), which in turn, was also higher than Spain (3.51 t ha^{-1}) (Baruth et al., 2019). Grain yields of rye were 5.50 t ha^{-1} in Denmark, 4.30 t ha^{-1} in Germany, and 2.85 t ha^{-1} in Spain. Although Spain presented a lower crop yield in all regions than the other two countries, Castile-Leon region had the highest cereal cropping area (0.88 million ha) (Baruth et al., 2019).

In the three countries, the $P_{Olsen,S}$ was higher than $P_{Olsen,T}$, which implied that the P fertiliser requirements for the production of these crops were only for maintenance or compensation of P exported (or P taken away when harvesting). Similar results were also observed (Panagos et al., 2022b; Recena et al., 2022) when assessing the magnitude of the build-up component for cropland in the EU on a NUTS 3 scale. However, the maintenance strategy could be subjected to further reduction of fertilisation quantities when the ratio $P_{Olsen,S} > 2P_{Olsen,T}$ (Delgado et al., 2016). The main reason is that $P_{Olsen,T}$ is a factor that relies on crop type and soil conditions, and depending on the case, if that threshold is surpassed it could lead to higher losses due to erosion (Delgado et al., 2016). As estimated by (Panagos et al., 2022a), in the agricultural land of the EU around 374 kt P per year is displaced (or 2 kg P ha^{-1} $year^{-1}$) and on average 18% of it is displaced to the riverine system and the sea. All regions in Denmark and Germany surpassed the ratio 1:2, while in Spain regions like Basque Community (1:1.4), Aragon (1:1.9), Madrid (1:1.5), Castile-La Mancha (1:1.8), Andalusia (1:1.4) and Murcia (1:1.8) were below (Table A5). Therefore, the potential P demand (Fig. 3b) considering the maintenance strategy, could be lower in those cases where $P_{Olsen,S} > 2P_{Olsen,T}$, thus making a more efficient use of P considering crop type and soil conditions.

3.4. Percentage of potential phosphorus demand covered by potential phosphorus supply

The percentage of potential P demand covered only by the potential P supply from P-recycling technologies in wheat, barley, and rye production was on average 9.1%, 21.7% and 10.0% in Denmark, Germany and Spain (Fig. 4a), respectively. In Spain alone, six regions (Galicia, Principality of Asturias, Cantabria, Madrid, Valencian Community and Murcia) could supply more than their full P demand (Table A6). In contrast, only Capital Region (Denmark) and North Rhine-Westphalia

(Germany) could cover over 40% (74% and 59%, respectively) in both Denmark and Germany. Nonetheless, the Spanish regions capable of supplying the full P demand for selected crops presented a potential P demand lower than 0.15 kg P per capita (Table A6). Also, Capital Region (Denmark) and North Rhine-Westphalia (Germany) had lower potential P demand (0.50 and 0.65 kg P per capita, respectively) than the other regions in both countries.

In some cases, the surplus regions were located inside the receiving regions (e.g., Berlin-Brandenburg and Bremen-Lower Saxony, Germany). It was possible to observe more than 10% increase in the potential P demand covered by potential P supply in German regions such as Brandenburg and Schleswig-Holstein, as well as in Andalusia, Spain (see Fig. 4b). Although receiving regions such as Castile-Leon (Spain) presented a lower increase, it also has one of the highest potential P demands from all regions in the 3 countries studied (9.94 kg P per capita). Therefore, the market prospect of P recycled products could be analysed in terms of spatial distribution among regions or countries with high potential P supply and low potential P demand and those with opposite conditions.

Spatial distribution analyses, connecting human and animal excreta P supply and crop demand regions, have been done in some countries, demonstrating that logistics are costly even if there are multiple benefits beyond P circularity (e.g. the Netherlands in Lessmann et al. (2023) and Sweden in Metson et al. (2020)). High transport costs associated with the bulky and low nutrient concentrations in sludges and application restrictions due to variable stoichiometric ratios between nutrients can make complete recycling of excreta challenging (Kleinman et al., 2022). In this study, we estimated the potential of recycled P products that have similar characteristics to mineral P fertilisers as an alternative and sustainable P supply for agricultural use. These recycled products could partly cover the P demand for crops with potentially lower logistic costs, but to determine the true potential requires further analysis.

The manufacturing and use of recycled P products evaluated in this manuscript could partially substitute mineral P fertilisers, offering not only benefits in terms of P security via circularity, but also potential environmental benefits. For example, substitution could reduce the risk of Cd accumulation from ~25% to less than 15% (Weissengruber et al., 2018). In countries with high sewage sludge direct reuse in agriculture, the risk of soil pollution with microplastics, estimated approximately in 6 kt per year in the EU at current reuse rates, could be reduced in the same proportion of substitution with recycled P products. Still, quantitatively comparing resource use and pollution risks between mined and recycled products remains challenging, given that studies use different system boundaries (Manoukian et al., 2023).

4. Conclusions

Most P recycling technologies from wastewater either do not provide the product information required or currently do not meet the majority of legislative guidelines to be used across multiple EU countries. Five out of the 22 technologies we reviewed were in full compliance with EU and national legislations in Denmark, Germany, and Spain. We conclude that there is still a need to provide complete information on product characteristics by technology providers, as it is the first step to comparing recycled P products with conventional mineral P fertilisers.

Still, for those technologies that were compliant, we observed that a notable proportion of P demand of selected crops (wheat, barley, and rye, which are produced in around 80% of the total cropland in the selected countries) could be covered by the potential P supply from P recycling technologies best suited for different countries. The most suitable product identified for Denmark was vivianite for calcium phosphate precipitation (0.38 kg P per capita), whereas in Germany ashes for calcium phosphate (0.29 kg P per capita) were the most suitable technology. Lastly, in the case of Spain, vivianite as iron supply was the most suitable product, with 0.05 kg P per capita. These represented 9.1, 21.7 and 10.0 percent (Denmark, Germany and Spain, respectively) of the total potential P demand estimated for wheat, barley and rye production in the studied countries.

Our estimates present a useful and systematically compiled figure, but do not represent the full variability of crop requirements on the landscape. For instance, provided that in Denmark and Spain the digested sludge reuse in agriculture is still commonly practised, there is more potential for sludge-based recycled P products than for ash-based. In Germany, in turn, the potential is higher for ash-based products and it is expected to increase due to an increase in sludge incineration rates as a consequence of legislation changes in 2017 that restrict sludge reuse in agriculture. Nonetheless, the estimation of the potential P supply was restricted due to a lack of information not only on sludge disposal routes at a regional level but also on the wastewater P inflow reported. Moreover, detailed information on soil properties which affect crop P uptake from a given product is necessary to accurately estimate potential substitution of mineral P.

From our estimations of potential supply and demand of recycled P, we evaluated possible trade of P among regions with low agricultural P requirements and regions that did not report crop production information or covered over 100% of their potential P demand. As a result, the percentage of demand covered by recycled P products increased by over 5% in most cases, where trade with neighbouring regions was assessed. Consequently, the analysis of the spatial distribution of potential P supply and potential P demand between regions is crucial to improve the P use efficiency in agriculture, thus reducing mineral P dependency and the environmental impact of directly using untreated sewage sludge in agricultural soil and water pollution.

CRediT authorship contribution statement

Juan Serrano-Gomez: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Visualization. **Geneviève S. Metson:** Conceptualization, Methodology, Writing – review & editing. **Tina-Simone Neset:** Conceptualization, Methodology, Writing – review & editing. **Jakob Santner:** Methodology, Writing – review & editing. **Ludwig Hermann:** Writing – review & editing, Supervision. **Matthias Zessner:** Methodology, Writing – review & editing, Supervision.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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