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RESEARCH ARTICLE

Agronomic efficiency of selected phosphorus fertilisers derived from secondary raw materials for European agriculture. A meta-analysis

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Abstract

Phosphorus (P) is a macronutrient essential for all living organisms. Food production has become highly dependent on mineral P-fertilisers derived from phosphate rock, a non-renewable and finite resource. Based on supply risk and economic importance for the European Union, phosphate rock and elemental P have been identified as critical raw materials. Moreover, P dissipation can lead to adverse impacts on the aquatic environment. The production and use of P-fertilisers derived from secondary raw materials could possibly contribute to a more sustainable agriculture in line with a circular economy. Biogenic and industrial resources and waste streams can be converted into value added materials, such as precipitated phosphate salts, thermal oxidation materials and derivates, and pyrolysis and gasification materials. A condition is, however, that the P must be recovered in a plant-available form and that the recovered P-fertiliser supports plant growth and nutrient uptake in European agroecosystems. Here, we review the agronomic efficiency of selected P-fertilisers derived from secondary raw materials by comparing plant responses relative to those after mined and synthetic P-fertiliser application in settings relevant for European agriculture, using meta-analyses. The major points are the following: (1) precipitated phosphate salts show similar agronomic efficiency to mined and synthetic P-fertilisers, with results that are consistent and generalisable across soil and crop types relevant for European agriculture; (2) thermal oxidation materials and derivates can deliver an effective alternative for mined and synthetic P-fertilisers, but the relative agronomic efficiency is dependent on the feedstock applied, possible post-combustion manufacturing processes, and the length of the plant growing season; (3) the agronomic efficiency of pyrolysis and gasification materials remains indeterminate due to a lack of available data for European settings. It is concluded that the agronomic efficiency of selected P-fertilisers derived from secondary raw materials supports their use in conventional and organic European agricultural sectors.

Keywords Phosphate fertiliser · Phosphate salts · Struvite · Biomass ashes · Biochar · Circular economy · Bioeconomy · Europe

1 Introduction

Present day phosphorus (P) nutrient use in the European agricultural sector can be characterised as predominantly linear, with significant P quantities accumulating in agricultural soils or being lost from the biogeochemical cycle and replenished by mineral fertilisers (Schoumans et al. 2015; van Dijk et al. 2016). At the same time, important phosphorus-rich waste streams are being produced, originating from effluents of municipal and industrial wastewater treatment systems, slaughter refuse, or manure from livestock production (van Dijk et al. 2016). Whereas a share of this organic P is recycled directly on agricultural land, a number of concerns are associated to the landspreading of unprocessed biogenic materials. At first, specific organic wastes may contain a broad set of pollutants, which could be hazardous for the environment and may pose a risk to human health (Alvarenga et al. 2016; Charlton et al. 2016a; Charlton et al. 2016b; Harrison et al. 2006; Lowman et al. 2013; McBride 2003). This relates in particular to the presence of potentially toxic metals and metalloids or pathogens, as well as emerging concerns over a wide range of organic bioactive substances, such as antibiotics, organo-metalloids, and endocrine-disrupting substances. As a consequence,

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these materials are increasingly being incinerated and the resulting ashes are transferred to landfills and construction materials (Buckwell and Nadeu 2016; Eurostat 2016), thus removing a significant P portion from the biogeochemical P cycle. Hence, valuable P in organic wastes and similar materials is currently being discarded for the sake of environmental and human health protection and improving public acceptance. Secondly, the unbalanced nutrient stoichiometry and spatial constraints linked to high transport costs of large volumes of material with low nutrient levels often hamper sustainable circular nutrient management and enhance P accumulation in soils (Buckwell and Nadeu 2016; Schoumans et al. 2010). A more efficient recycling of P may also contribute to providing alternative P sources for the European agricultural sector because phosphate rock, the primary material used for production of mineral P-fertilisers, is a finite resource and P demand may further increase over time (Cordell et al. 2009; MacDonald et al. 2011; Sattari et al. 2016). The concentration of P mines outside the continent makes the European Union highly vulnerable on imports, fluctuating prices of raw materials, as well as the political situation in supplying countries (George et al. 2016; Schröder et al. 2010). In any case, in order for recovered P-fertilisers to present a viable alternative to existing mineral P-fertilisers and to avoid long-term P accretion in soils, the P must be recovered in a plant-available form (Schröder et al. 2010). Hence, sustainable nutrient management in Europe will require to shift away from the current handling scenarios for biogenic P-rich materials and to promote efficient P-recycling within the agricultural sector.

The scope of the present study is on processed P-fertilisers derived from secondary raw materials that enable a decoupling of their nutrient value from the undesired properties, such as the low nutrient-to-volume ratio or the presence of specific contaminants (Fig. 1). Explicitly, this work focuses on three distinct P-recovery pathways for which the end-materials could possibly provide an alternative to mined and synthetic P-fertilisers:

Fig. 1 The agricultural valorisation of recovered P from secondary raw materials in high-quality fertilisers provides unique opportunities for nutrient recycling, and can possibly provide an alternative to mined and synthetic P-fertilisers in line with the circular economy framework (A farmer broadcasting fertilisers on arable land; ©oticki—stock.adobe.com)

i. *Precipitated phosphate salts* crystallised out of liquid and liquefied waste streams in the form of phosphate salts (e.g. struvite, $MgNH_4PO_4.6H_2O$). In practice, the recovered materials are not pure salts and, depending on the input material and recovery process applied, the co-precipitation of some organic matter, salts, and hydroxides of some metals present in the waste water end-products (Ca, K, Fe, etc.) typically occurs (Hao et al. 2013);

ii. P-rich ashes and slags obtained after thermal oxidation under non-oxygen limiting conditions. This material group includes raw incineration ashes (e.g. poultry litter ash) as well as derivates from the ashes formed through wet-chemical or thermal manufacturing processes aiming at the removal of contaminants and the increase in plant P-availability (hereafter *thermal oxidation materials and derivates*);

iii. P-rich pyrolysis and gasification materials obtained from production processes in a zero or low oxygen environment that form part of the pyrolysis spectrum techniques, including hydrothermal carbonisation, pyrolysis, and gasification (hereafter *pyrolysis and gasification materials*). This material group is often referred to as "biochar" and "gasification biochar" in scientific publications.

After setting minimum product quality requirements, these materials might show negligible risks for the environment and human health and can provide a cost- and carbon-efficient transport pathway for dissipated P from nutrient-excess to nutrient-poor regions in Europe (Buckwell and Nadeu 2016; Huygens et al. 2016; Schoumans et al. 2015). A comprehensive overview of the different production processes for the selected P-fertilisers derived from secondary raw materials, the characteristics of these end-materials, and possible quality requirements for their use as fertilisers are given in Huygens et al. (2016, 2017). It is noted that thermal oxidation materials and derivates and pyrolysis and gasification materials can serve other fertilising functions (e.g. soil improver, liming material, growing media, and plant biostimulant), but evaluating the potential of such fertilising applications falls beyond the scope of this study.

This study aims at assessing if the materials can fulfil the technical requirements for fertilising purposes. This is a relevant question because of the specific nature of such fertilisers; they typically have a reduced water-soluble fraction, but are highly soluble in acid media (Lehmann and Joseph 2015; Wilken et al. 2015). A quantitative review based on meta-analysis techniques is undertaken that compares dry matter yields and P uptake efficiencies for plants grown with P-fertilisers derived from primary and secondary raw materials. Mathematically combining data from a series of well-conducted primary studies provides a more precise estimate that reduces the size of the confidence interval of the underlying "true effect" than any individual study (Garg et al. 2008; Pogue and Yusuf 1998). Meta-analysis techniques enable establishing whether the scientific findings are consistent and generalisable across European settings and facilitate understanding reasons why some studies differ in their results. For these reasons, a meta-analysis of similar, well-conducted, randomised, controlled trials has been considered one of the highest levels of evidence (Garg et al. 2008).

2 Materials and methods

2.1 Data sources

The literature search was initiated using the ISI Web of Science with the topic search terms "phosphorus AND plant AND fertili*er AND (recovery OR waste OR struvite OR calcium phosphate OR ash OR combustion OR biochar OR pyrochar OR hydrochar)." Searches were also undertaken with Google Scholar in order to pick up publications that were not indexed in the Web of Science. The inclusion of grey literature in meta-analysis studies is generally regarded as reducing publication bias and, therefore, preferable (McAuley et al. 2000). The cut-off date for data collection was 1 December 2016.

Studies that quantitatively reported dry matter yield and/or plant P uptake after the application of recovered P and mineral P-fertiliser treatments during one plant growing season were selected. Only processed P-fertilisers with a minimum P_2O_5 content of 2% were selected. Studies with less than three experimental replicates were discarded. When studies did not report measures of variance, the corresponding author was contacted to provide the raw data for the calculation of the standard deviation. When measures of variance were not documented and could not be retrieved, uncertainty of these missing effect sizes was drawn from a multiple imputation algorithm based on the assumption of a common underlying variance, after which Rubin's rules were applied to get the point estimates and standard errors of the meta-analysis results (Schwarzer et al. 2015). Only assessments that there were performed on soils and plant species from boreal, temperate, and Mediterranean climate regions—within or outside Europe—were retained in order to provide an assessment that is relevant for the EU-27 (i.e. latitudes > 35° N/S). If not directly reported, P uptake was derived from the dry matter yield and plant P concentrations, and concomitant standard deviations were calculated assuming error propagation rules for normal distributions. When data were only provided in graphical format, the corresponding authors of the studies were contacted to obtain the raw numerical data. If not successful, relevant data points were extracted from the figures in the paper.

More studies were available for precipitated phosphate salts (26 for the relative agronomic efficiency for the response variable dry matter yield), and thermal oxidation materials and derivates (16 for the relative agronomic efficiency for the response variable dry matter yield), than for pyrolysis and gasification materials (eight for the relative agronomic efficiency for the response variable dry matter yield) (Table 1). Therefore, the results should be interpreted with the necessary caution and it should be clear that the conclusions with regard to agronomic efficiency differ in strength for each of the three fertiliser groups. Following studies were included in the assessment:

Precipitated phosphate salts: Achat et al. 2014b; Ackerman et al. 2013; Antonini et al. 2012; Bonvin et al. 2015; Cabeza et al. 2011; Cerrillo et al. 2015; Degryse et al. 2017; Gell et al. 2011; Gonzalez Ponce and Garcia Lopez De Sa 2007; Hammond and White 2005; Hilt et al. 2016; Johnston and Richards 2003; Katanda et al. 2016; Liu et al. 2016; Liu et al. 2011; Massey et al. 2009; Plaza et al. 2007; Ruiz Diaz et al. 2010; Sigurnjak et al. 2016; STOWA 2016; Talboys et al. 2016; Thompson 2013; Uysal et al. 2014; Vaneeckhaute et al. 2016; Vogel et al. 2015; Weinfurtner et al. 2009; Wilken et al. 2015.

Thermal oxidation materials and derivates: Brod et al. 2016; Cabeza et al. 2011; Codling et al. 2002; Delin 2016; Franz 2008; Komiyama et al. 2013; Kuligowski et al. 2010; Nanzer et al. 2014; Reiter and Middleton 2016; Rex et al. 2013; Schiemenz and Eichler-Löbermann 2010; Schiemenz et al. 2011; Severin et al. 2014; Vogel et al. 2015; Weigand et al. 2013; Wells 2013; Wilken et al. 2015.

Pyrolysis and gasification materials: Alotaibi et al. 2013; Codling et al. 2002; Collins et al. 2013; Kuligowski et al. 2010; Ma and Matsunaka 2013; Müller-Stöver et al. 2012; Reiter and Middleton 2016; Siebers et al. 2014.

2.2 Effect size

Plant dry matter yield and plant P use efficiency were used as the common statistical measures, or response variables, that are shared among studies. Plant P use efficiency was calculated as the difference in P uptake between fertilised (PU_F) and unfertilised plants (PU_C), expressed relative to the fertiliser P applied ($P_{applied}$, kg P ha^{-1}):

$$\text{P use efficiency} = (PU_F - PU_C)/P_{applied}. \qquad (1)$$

Table 1 Number of studies and cases included in the meta-analyses on the agronomic efficiency of P-fertilisers derived from precipitated phosphate salts, thermal oxidation materials and derivates, and pyrolysis and gasification materials relative to P-fertilisers derived from primary raw materials (RAE_{DMY} and RAE_{PUE} indicate the relative agronomic efficiency for the response variables dry matter yield and phosphorus use efficiency, respectively)

	Precipitated phosphate salts		Thermal oxidation materials and derivates		Pyrolysis and gasification materials	
	RAE_{DMY}	RAE_{PUE}	RAE_{DMY}	RAE_{PUE}	RAE_{DMY}	RAE_{PUE}
Studies	26	19	16	14	8	6
Cases	173	103	113	94	31	16

Standardisation of the literature results was undertaken through calculation of the effect size. This allows quantitative statistical information to be pooled from and robust statistical comparisons to be made between effects from a range of studies that reported results based on different experimental variables. The effect size was calculated as the natural logarithm of the response ratio R by using the following equation (Borenstein et al. 2009):

$$\ln R = \ln\left(\bar{X}_{\text{P-fertilisers derived from secondary raw materials}}/\bar{X}_{\text{mined and synthetic P-fertilisers}}\right). \tag{2}$$

where $\bar{X}_{\text{P-fertilisers derived from secondary raw materials}}$: mean dry matter yield or mean P use efficiency after the application of P-fertilisers derived from secondary raw materials, and $\bar{X}_{\text{mined and synthetic P-fertilisers}}$: mean dry matter yield or mean P use efficiency after the application of mined and synthetic P-fertilisers.

The response ratio was then calculated for a number of pairwise comparisons or "cases" where all grouping variables are identical for both fertiliser treatments. These variables include soil and crop used, crop harvest time, P application rate, etc. (see Sect. 2.3). We used the log response ratio and its variance in the analysis to yield summary effects and confidence limits in log units during the different meta-analysis steps. Each of these values was then converted back to response ratios to report the final results (Borenstein et al. 2009) (see Sect. 2.4.).

When P uptake is lower for fertilised than for the control unfertilised treatments, a negative P use efficiency value is produced that limits further calculations. Therefore, only cases were retained when the P uptake after the application of mined and synthetic P-fertilisers (PU_{Fprim}) is significantly different from the unfertilised treatment (PU_C) at the 95% level, corresponding to the cases when the application of mined and synthetic P-fertilisers effectively increased plant P uptake. The selective removal of all such cases, however, penalised treatments assessing the plant P uptake responses to P-fertilisers derived from secondary raw materials (PU_{Fsec}) as it also removed some cases for which exclusively those treatment resulted in a significantly greater plant P uptake relative to the unfertilised treatment. Therefore, the number of cases when $PU_{Fsec} > PU_C$ and $PU_{Fprim} = PU_C$ was calculated and an equal number of cases for which $PU_{Fsec} = PU_C$ and $F_{prim} > PU_C$ were removed from the analyses. This was done by cumulatively removing the F_{sec} treatments that were least different from PU_C as indicated by the P value of a t test between PU_{Fsec} and PU_C. Ultimately, this procedure generated a dataset in which only positive P use efficiency values were retained.

P-fertilisers derived from secondary raw materials are fertilisers resulting from of a nutrient recovery operation of secondary raw materials through crystallisation processes (e.g. struvite and calcium phosphates; precipitated phosphate salts) or thermo-chemical processes (i.e. ashes, ash-derivates, slags, and chars as obtained by thermal oxidation and gasification/pyrolysis; thermal oxidation materials and derivates and pyrolysis and gasification materials, respectively). Mined and synthetic P-fertiliser treatments included different P fertilising substances, such as triple superphosphate, monoammonium phosphate, diammonium phosphate, calcium super phosphate, single superphosphate, and potassium phosphate. Dry matter yield and plant P uptake were mostly measured for aboveground plant biomass yield, but some studies assessed whole plant biomass or specific plant organs. The control was defined as being identical to the experimental treatment with regard to all variables apart from the type of fertiliser applied.

2.3 Grouping variables

For all selected studies, quantitative information on following grouping variables was recorded: application rate, application form, harvest time after fertiliser application, soil pH, soil texture, soil P fertility, sowed plant species, experiment type, and geographic latitude of the collected experimental soils. When specific parameters were not documented in the publication, the corresponding author was requested to provide the information. In case the data was not available, the respective cases were not included in the statistical assessment for the grouping variable.

Data were grouped prior to meta-analysis to enable a broad ranging assessment of fertilising effectiveness of P-fertilisers derived from secondary raw materials as a function of soil type, plant group, and management option. *Soil pH* was classified as acidic for soils with a pH value less or equal than 6.0 and as neutral/basic for soils of pH greater than 6.0. *Soil texture* was classified as coarse (sand, loamy sand, and sandy loam), medium (loam, silt loam, and silt), or fine (sandy clay, sandy clay loam, clay loam, silty sandy clay loam, silty clay, and clay). *Feedstock* indicated the input materials from which the P-fertiliser was derived (e.g. sewage sludge, manure). For thermal oxidation materials and derivates, *post-processing* refers to the production of ash-derivates through wet-chemical or thermal manufacturing steps applied. *Plant groups* involved grasses (both annual and perennial species), oilseeds, cereals, legumes, and others (e.g. leaf vegetable, cormous flowering plants, fruit vegetable). *Application form* distinguished fertilisers that were applied as a powder or as granules. *Assessment time* was categorised as either short or long for studies that harvested plants within and posterior to a period of 65 days of fertiliser application, respectively. In case of assessments on grasses, only the cumulative biomass and P uptake at the end of the experiment was considered. *Soil P status* was categorised as P-poor and P-rich, with a cutoff value of extractable Olsen-P content of 12.4 mg P kg^{-1}. The cutoff value was based on the average limit value for the "very low" P fertility category for a single soil within a number of European countries (Jordan-Meille et al. 2012). When other extractable P methods were applied, transfer functions and comparative relationships as given in Jordan-Meille et al. (2012), Neyroud and Lischer (2003) McLaughlin (2002), and Prasad et al. (1988) were applied. A P-poor status was assumed for studies that used Rhine sand as potting medium. The approach applied based on a single cutoff value and transfer functions to discern soil P fertility for all soil-plant combinations is a simplification of a complex scientific matter (Jordan-Meille et al. 2012), but we are confident that it meets the objective of generally discerning soil P status in this meta-analysis study. *Experimental setting* separated pot from field studies. *Experimental design* assessed if the experimental study design involved the addition of plant nutrients, other than P, present in P-fertilisers derived from secondary raw materials were also added in the treatment that applied mined and synthetic P-fertilisers; "Fully balanced" corresponds to cases where all micro- and macronutrients were balanced between treatments. "Deficient" refers to designs where primary and secondary macronutrients present in P-fertilisers derived from secondary raw materials were not added in the treatment that applied mined and synthetic P-fertilisers (e.g. struvite as P-fertiliser derived from secondary raw materials, but no addition of Mg in the mined and synthetic P-fertiliser treatment). The effect of the different groups was assessed in the meta-analysis. The geographic latitudes of the collected soils were plotted against the relative agronomic efficiency for the response variable P use efficiency, and the significance of the regression slope was assessed.

2.4 Presentation of meta-analysis results

The response ratio can be interpreted as the agronomic efficiency of P-fertilisers derived from secondary P sources relative to mined and synthetic P-fertilisers. Response ratios were plotted for the different grouping variables with squares indicating the weighted mean of the effect and error bars showing 95% confidence intervals. A relative agronomic efficiency value below 1 indicates that that the P-fertiliser derived from secondary P sources is a less effective plant P-source than a synthetic P-fertiliser derived from mined phosphate rock; a value above 1 indicates the opposite. The error bars that cross the vertical 1 line indicate that the agronomic efficiency of F_{sec} is not significantly different from F_{prim}. Meta-analyses were performed using the "meta" package (Schwarzer 2007) in R version 3.3.0 (R Development Core Team 2008).

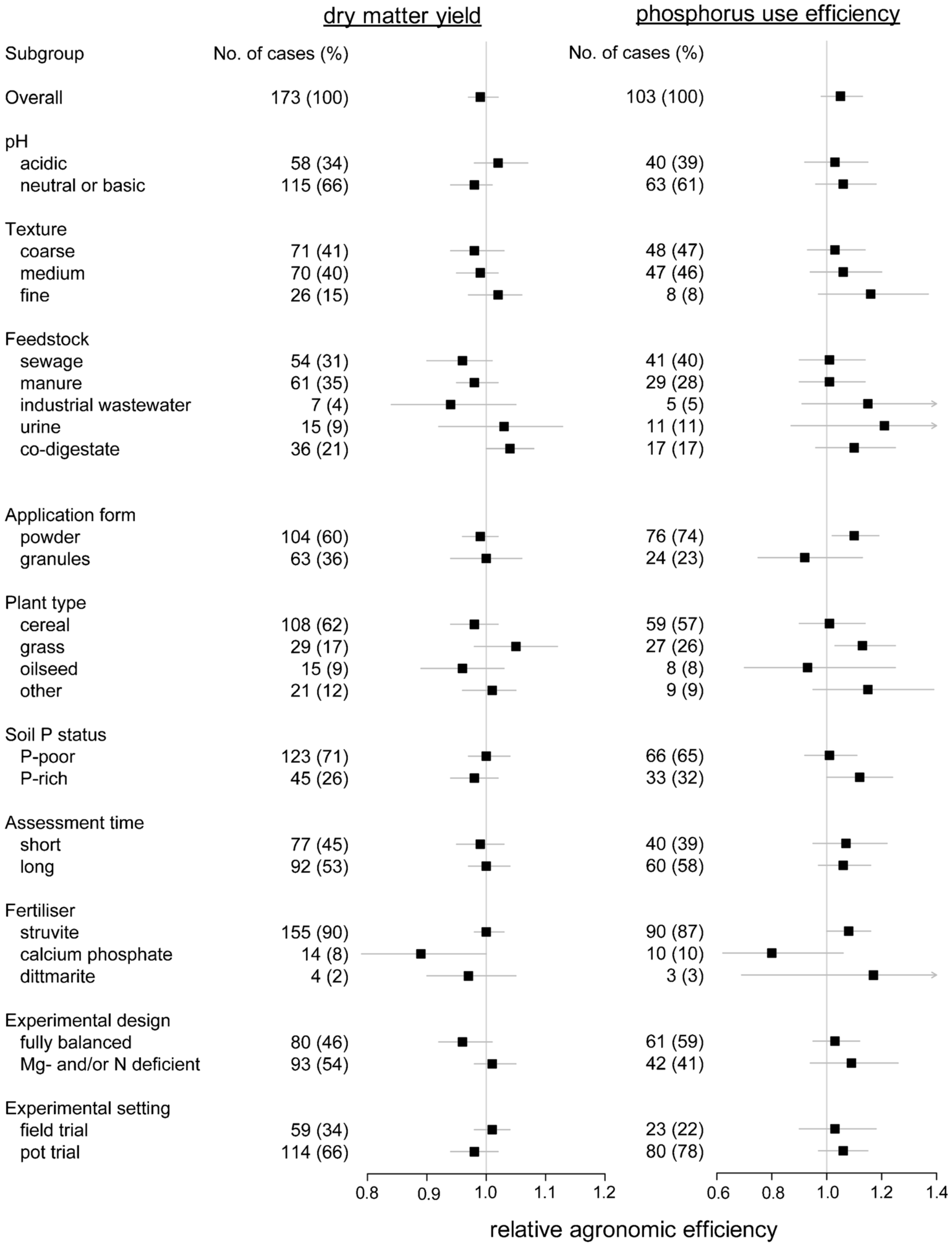
Data availability statement The datasets analysed during the current study are not publicly available because the authors obtained some primary data from original works based on an agreement that the data would be presented as "aggregated results of the full database as a mean value plus a standard deviation." Data are available from the corresponding author on reasonable request, and on condition that the author of the primary data approves the request.

3 Results and discussion

3.1 Precipitated phosphate salts

The overall results indicated a similar agronomic efficiency for precipitated phosphate salts compared to mined and synthetic P-fertilisers. The mean relative agronomic efficiency values equal 0.99 and 1.05 for dry matter yield and P use efficiency, respectively (Fig. 2), with the corresponding 95% confidence intervals overlapping the 1 value for both parameters. These observations hold true for groups varying in soil pH, soil texture, feedstock, application form, plant type, soil P status, assessment time, and experimental design and setting. The agronomic efficiency of precipitated phosphate salts is thus consistent and generalisable across different settings, including soil and crop types, relevant for the European agricultural sector. Although multi-year assessments fall beyond the scope of this meta-analysis, the results of Thompson (2013) and Wilken et al. (2015) confirm the sustained long-term efficiency of precipitated phosphate salts as a P-fertiliser.

Fig. 2 The agronomic efficiency of precipitated phosphate salts relative to mined and synthetic P-fertilisers for the response variables dry matter yield and phosphorus use efficiency as a function of grouping variables. Results are presented as weighted mean (square) and 95% confidence intervals (error bars)



Unlike most mined and synthetic P-fertilisers, precipitated phosphate salts are water insoluble, but their solubility is increased in acid solutions (Wilken et al. 2015). Nonetheless, our results indicated that soil pH had no significant effect on the relative agronomic efficiency. Achat et al. (2014a) indicated that isotopically exchangeable P was similar for finely ground struvite as for triple superphosphate, irrespective of pH in the range 5.2–8.1. Talboys et al. (2016) indicated that the short term (< 42 days) dissolution of granulated struvite, the most common precipitated phosphate salt, shows similar dynamics across a wider soil pH range of 5.0–8.0. Degryse et al. (2017) indicated a 60-day granulated struvite dissolution rate of > 80% in an acid soil (pH 5.9), but < 10% dissolution in a basic soil (pH 8.5). Hence, as most European soils have a pH between 5 and 8 (Reuter et al. 2008), soil pH is not expected to exert a major influence over the dissolution patterns of precipitated phosphate salts and the relative agronomic efficiency. Plants also modify the rhizosphere pH as they exudate organic acids from their root biomass in significant quantities that can drastically lower pH in the plant root microenvironment. Talboys et al. (2016) indicated that organic acids have a major impact on the rate of dissolution of P from struvite and that plants with root systems that exude large quantities of organic acids are more effective at taking up P from struvite granules.

The exudates cause the dissolution of the precipitated phosphate salts in the vicinity of the plant root. Grasses exudate significantly more organic acids than common crops; estimates for the total allocation of photosynthates—a proxy for rhizodeposition—to roots are 50–70% higher for grasses than for cereals, such as wheat and barley (Kuzyakov and Domanski 2000). Hence, species-specific patterns of root exudation may explain the variations in relative agronomic efficiencies observed, but the effect of plant type is overall not significant (Fig. 2).

No significant effect of assessment time and application form on the relative agronomic efficiency along a single plant growing season was observed for precipitated phosphate salts (Fig. 2). Although the slower initial P release rate from the granulated fertiliser could possibly reduce plant uptake of P during the very initial plant growth stages (< 36 days; Degryse et al. 2017; Talboys et al. 2016), studies that applied an assessment time between 36 and 65 days showed good performance when precipitated phosphate salts were applied. For crops subject to struvite fertilisation, it is has been suggested that a reduction in number of grain heads due to short-term P deficiency is counterbalanced by the crop root system's capacity to take up P in the later plant growth stages (Talboys et al. 2016). Hence, even for studies with an assessment time < 65 days, the sustained P release from precipitated phosphate salts could possibly compensate their lower initial P-availability and their lower P-dissolution rate relative to water-soluble P-fertilisers (Degryse et al. 2017; Talboys et al. 2016). The relative agronomic efficiencies for dry matter yield and P use efficiency were not significantly different from 1 for struvite and dittmarite, but the 95% confidence interval for calcium phosphates (grouping variable fertiliser) extended to a value marginally below 1 for dry matter yield (0.995; Fig. 2). Struvite is the most common precipitated phosphate salt, but some P-recovery processes target a different end-material such as dittmarite or dicalcium phosphates. The crystallisation of calcium phosphates may involve the formation of metastable precursor phases, such as octocalcium phosphate and hydroxyapatite, which are less available to plants, especially at alkaline pH (Wang and Nancollas 2008). Hence, the relative agronomic efficiency of calcium phosphates can vary depending on the exact composition of the calcium phosphate phases included in the end-material. After application to the soil, calcium phosphates can also transform into more stable forms (Arai and Sparks 2007), potentially further contributing to the wider relative agronomic efficiency ranges observed for calcium phosphates than for struvite and dittmarite.

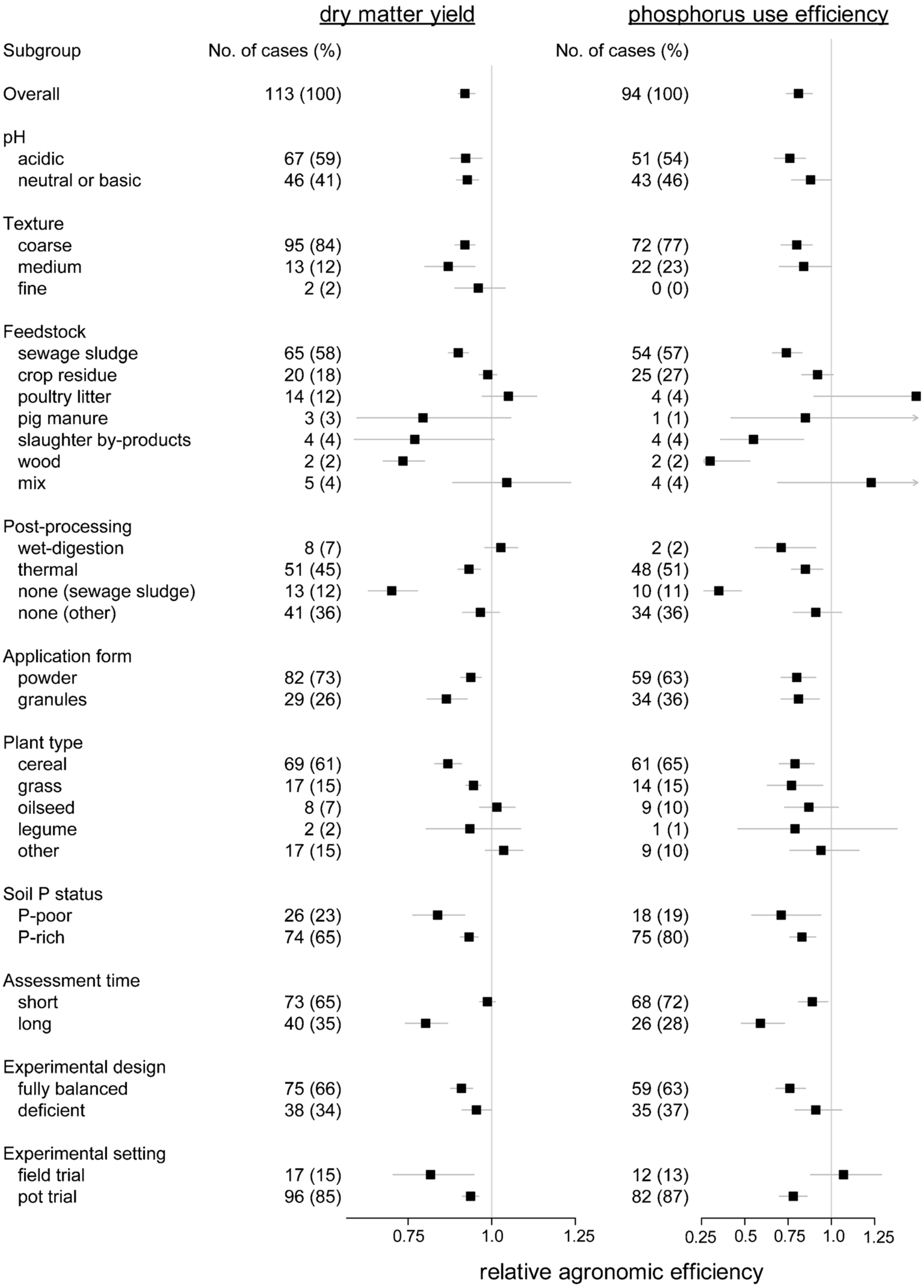
3.2 Thermal oxidation materials and derivates

The mean relative agronomic efficiency values for thermal oxidation materials and derivates equal 0.92 and 0.81 for dry matter yield and P use efficiency, respectively (Fig. 3).

Significant differences in the relative agronomic efficiency of thermal oxidation materials and derivates were observed dependent on the feedstock applied and the possible post-processing steps that were performed (Fig. 3). The agronomic efficiency of thermal oxidation materials and derivates derived from crop residues, poultry litter, and pig manure did not differ from mined and synthetic fertilisers (Fig. 3). Thermal oxidation materials and derivates derived from wood showed a low relative agronomic efficiency, but the results should be interpreted with precaution because of the low number of cases (Fig. 3). Thermal oxidation materials and derivates derived from sewage sludge showed a significantly lower relative agronomic efficiency than for thermal oxidation materials and derivates derived from crop residues and poultry litter (Fig. 3). Nonetheless, it should be considered that thermal oxidation materials and derivates derived from sewage sludge include both raw ashes and ashes that have been further processed after incineration, and that results for crop residues were derived from only three studies that used a similar soil type (Delin 2016; Schiemenz and Eichler-Löbermann 2010; Schiemenz et al. 2011). For sewage sludge ashes, a post-incineration manufacturing step is often applied to increase P-availability and to comply with legislative limit values for metals and metalloids. This analysis confirms that such manufacturing processes starting from sewage sludge mono-incineration ashes clearly improve the plant availability relative to unprocessed sewage sludge ashes and enable the transformation of sewage sludge ashes into efficient P-fertilisers. The relative agronomic efficiency values for dry matter yield were 1.03 and 0.93 for materials subjected to wet-digestion and thermal post-processing steps, respectively (Fig. 3). Relative agronomic efficiencies close to 1 can reasonably be expected for materials resulting from wet-digestion post-processing, especially for these that have an equal chemical composition to that of mined rock phosphate and processed P-fertilisers (e.g. Ecophos® process, ICL RecoPhos® process, acidulation process; see Huygens et al. (2016) and Egle et al. (2016)). Thermal post-processing steps on sewage sludge incineration ashes aim at separating P from other elements and to influence the crystal structure of the materials by isomorphic substitution of the PO_4^{3-} ionic group (by for example SiO_4^{2-} or CO_3^{2-}) and thus affect the reactivity of the final product and therefore plant P availability. The final products show similar characteristics as Thomasphosphate and Rhenaniaphosphate (Huygens et al. 2016) and show overall good fertiliser efficiency.

The observed relative agronomic efficiencies were not affected by soil pH, soil texture, application form, or soil P status (Fig. 3). The impact of pH on the P-dissolution depends on the elemental composition of the P-fertiliser because P is strongly bond to Ca at high pH and to Fe and Al

Fig. 3 The agronomic efficiency of thermal oxidation materials and derivates relative to mined and synthetic P-fertilisers for the response variables dry matter yield and phosphorus use efficiency as a function of grouping variables. Results are presented as weighted mean (square) and 95% confidence intervals (error bars)



at low pH (Hinsinger 2001; Tóth et al. 2014). Nonetheless, the high basic cation contents of some thermal oxidation materials might buffer the acidity effect of the soil micro-environment, thus obscuring the effect of the soil pH. Also, no consistent differences were observed in relative agronomic efficiency across plant types for the response variables, indicating that possible differences in root exudation patterns of organic acids are not meaningfully impacting the P-release patterns from thermal oxidation materials and derivates.

A significant effect of assessment time on relative agronomic efficiency for dry matter yield and P use efficiency was observed ($P < 0.001$; Fig. 3), with values that are 20–40% lower in the long-term (> 65 days) than in the short-term (< 65 days). The plant-availability of the P in thermal oxidation materials and derivates is likely controlled by the coordinated cations of Ca, Mg Al, and Fe to which PO_4^{3-} is bound. All these different ions are abundantly present in thermal oxidation materials, although their relative abundance varies across end-materials. Complexes between phosphate and K, Ca, Mg, and S ions are relatively easily decomposed (Hinsinger 2001; Tóth et al. 2014), and this more labile P-fraction is therefore likely to be released in the short-term. Phosphate may, however, be unavailable to plants when strongly bound to particular trivalent cations in a stable matrix (Barrow 1984; Hinsinger 2001). The release of P from this more stable fraction could be limited, effectively decreasing the long-term P supply from thermal oxidation materials and derivates. This contrasts with mined and synthetic fertilisers that are of a uniform chemical composition; such fertilisers can be expected to release P readily upon physical disintegration. The released P that is not readily taken up by plants can be adsorbed to soil minerals, with the nature of such reactions dependent on the pH and on the concentration of metal cations, such as Ca, Fe, and Al as well as organic and inorganic ligands (Hinsinger 2001; Tóth et al. 2014). At a later time in the plant growing season, desorption of sorbed P can occur via ligand exchange reactions, especially if the P was bound in more labile soil P-complexes (Hinsinger 2001). Possibly, such desorption processes could effectively contribute to a better long-term effect of mined and synthetic P-fertilisers compared to thermal oxidation materials and derivates rich in trivalent cations.

A significant effect of experimental design (*P*: 0.04) and experimental setting (*P*: 0.003) was observed for the relative agronomic efficiency for the response parameter P use efficiency (Fig. 3). Studies that supply primary and secondary macronutrients together with mined and synthetic P-fertilisers to ensure the equal supply of all different plant nutrients present in the thermal oxidation materials and derivates show somewhat reduced relative agronomic efficiency values, especially for the response variable P use efficiency. On the other hand, results for the field studies performed in more realistic settings than those of pot experiments show better results, although this effect was only observed for the response variable P use efficiency. Both effects are potentially related, as field studies often apply a deficient experimental design where the broad range of secondary macronutrients and micronutrients present in thermal oxidation materials and derivates are not added in the mined and synthetic P-fertiliser treatment. Hence, these results indicate the importance of secondary macronutrients and micronutrients in achieving optimal agricultural yields. It is often challenging to evaluate the supplementary fertiliser need for particular plant-limiting elements within the broad spectrum of secondary macronutrients and micronutrients. On condition that the excess application of micronutrients is avoided, the application of thermal oxidation materials and derivates as P-fertilisers could provide the complementary benefit of supplying secondary macronutrients and micronutrients to enhance agronomic yields.

Altogether, these observations validate that thermal oxidation materials and derivates can deliver an effective alternative for mined and synthetic P-fertilisers in the European agriculture, but that the relative agronomic efficiency is dependent on the properties of the produced material.

3.3 Pyrolysis and gasification materials

The mean relative agronomic efficiency values for pyrolysis and gasification materials equal 0.87 and 0.46 for dry matter yield and P use efficiency, respectively (Fig. 4). Due to the low sample size, only a marginal reduction of the size of the confidence interval of the underlying true effect across groups could be achieved, compared to the results from individual studies by applying the meta-analysis techniques. Hence, no general conclusions can be drawn on relative agronomic efficiency across pyrolysis and gasification materials applied to different soil types, feedstocks, application form, and plant types. Figure 4 enables, nevertheless, a standardised visual assessment of the ranges observed for relative agronomic efficiency across selected studies.

The properties of pyrolysis and gasification materials can vary widely, depending on the interactive effects between production process conditions and feedstock applied. Many groups, including pyrolysis and gasification materials derived from slaughter by-products, poultry litter, crop residues, and pig manure, display an agronomic efficiency that is not significantly different from mined and synthetic P-fertilisers (Fig. 4). The significant differences in relative agronomic efficiency between specific groups varying in soil texture (for dry matter yield), feedstock (for P use efficiency), application form (for P use efficiency), plant type (for dry matter yield and P use efficiency), experimental design, and setting (for P use efficiency) should be interpreted with caution because some of the contrasting groups have a low number of cases, often originating from a few studies.

Only the relative agronomic efficiency values for neutral and basic soils and for pyrolysis and gasification materials that were applied in granulated form were derived from a minimum of four different studies and a number of cases greater than 10 for both response variables (Fig. 4). For these groups, the relative agronomic efficiency values pointed towards a significantly lower effectiveness than for mined and synthetic P-fertilisers. Potentially, some of the documented high agronomic efficiencies after the addition of pyrolysis and gasification materials could be the result of a liming effect that increases soil P availability (Hass et al. 2012), or the result of the milling of the pyrolysis and gasification material that increases the P solubility in the otherwise stable pyrolysis matrix (Ma and Matsunaka 2013). Therefore, future studies should focus on assessing the mechanisms that underlie documented potential positive plant responses and evaluate the agronomic efficiency of pyrolysis and gasification materials in the same physical form as it will be applied under actual settings in agriculture.

It is concluded that the current available dataset does not enable a comprehensive assessment of the agricultural efficiency of P-rich pyrolysis and gasification materials in

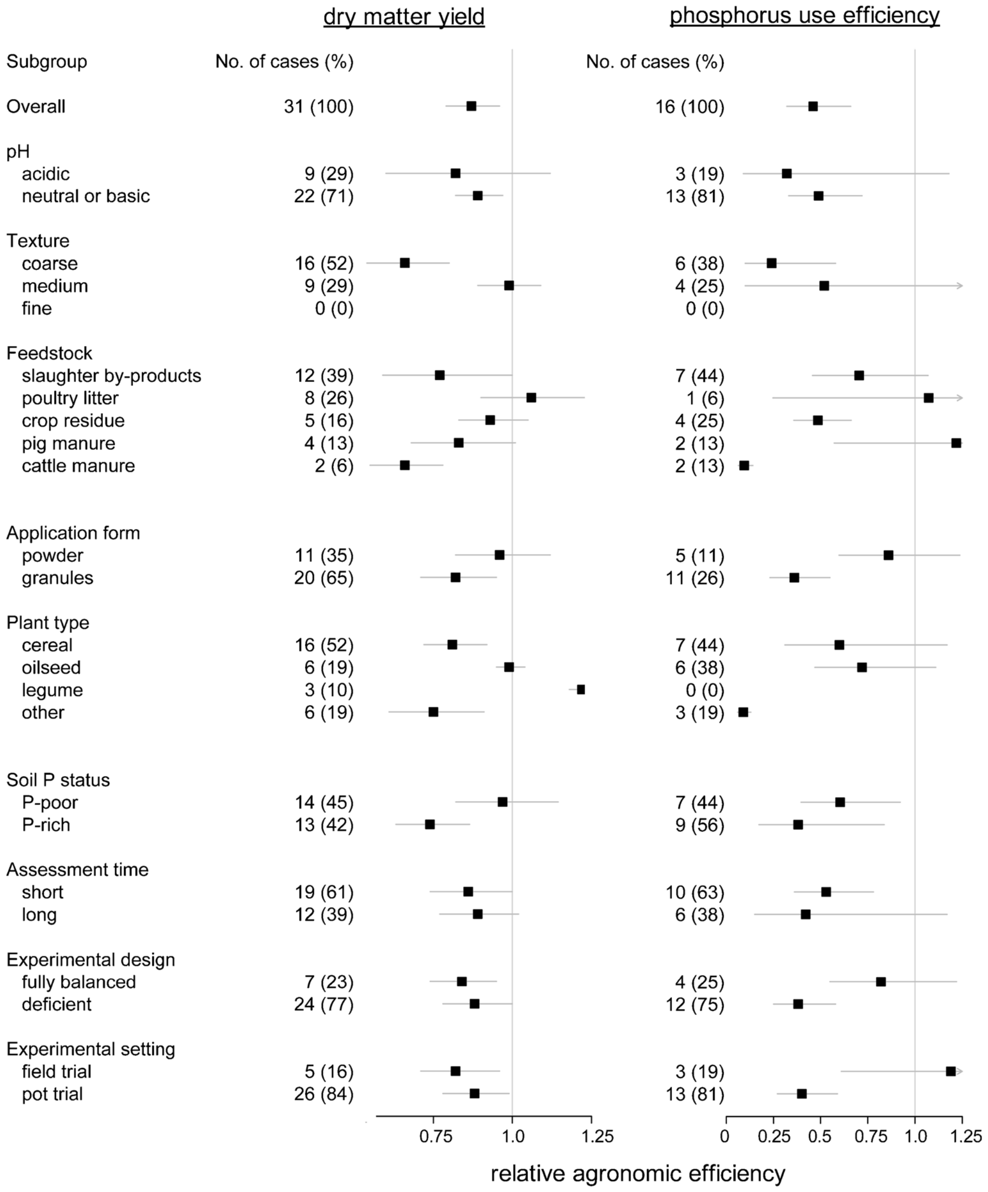


Fig. 4 The agronomic efficiency of pyrolysis and gasification materials relative to mined and synthetic P-fertilisers for the response variables dry matter yield and phosphorus use efficiency as a function of grouping variables. Results are presented as weighted mean (square) and 95% confidence intervals (error bars)

relevant European agricultural settings and that plant responses for P-rich pyrolysis and gasification materials can vary widely depending on the feedstock and production conditions of the pyrolysis and gasification materials, as well as on the soil and plant type under fertilisation.

3.4 Effect of geographic latitude

Sections 3.1–3.3 provide an overview of the relative agronomic efficiency as a function of soil and plant type, but fail to take into consideration the interactions and combinations of those variables that occur in different geographic regions in Europe. Especially the effect of the north–south position (i.e. latitude of the geographic coordinates) is relevant to consider, given that climate conditions (colder and drier soils at higher latitudes), soil texture (sandier at higher latitudes), and soil pH (more basic at lower latitudes) vary significantly across this gradient (Ballabio et al. 2016; Panagos et al. 2012). Concerns related to the effectiveness of water insoluble P-fertilisers in semi-arid and Mediterranean regions may exist because some slow release P-fertilisers, such as phosphate rock and meat and bone meal, do not dissolve readily in such soils (Bolland and Gilkes 1990; Elliott et al. 2007). The results of our work, however, reject such expectations for European settings as the relative agronomic efficiency for the response variable P use efficiency correlated negatively to latitude (Fig. 5). A significant negative correlation between geographic latitude and the relative agronomic efficiency was indicated (*P*: 0.02), with greater values observed in sites of lower latitudes than in higher latitudes (Fig. 5). Latitude explained, however, only a minor share of the total variance

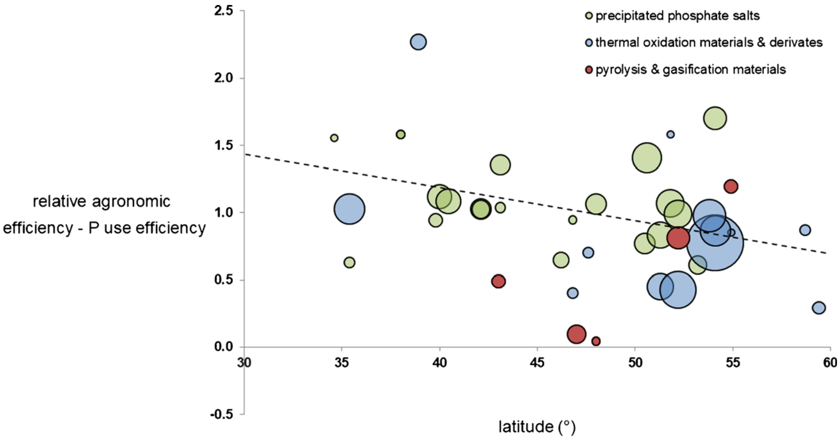


Fig. 5 Bubble plot indicating the relationship between the relative agronomic efficiency for the response variable phosphorus use efficiency and geographic latitude. The size of the bubbles represents the number of cases and relative weight for each data pair. The regression line across all data points was significant (*P* 0.02; R^2_{adj} 0.14)

observed (R^2_{adj}: 0.14) (Fig. 5). It should, however, be noted that the assessment includes both pot and field studies, and that some variables, especially climate conditions, may not be accurately represented in pot experiments. Therefore, the results should be interpreted with the necessary precaution. Nonetheless, our preliminary results suggest effectiveness of P-fertilisers derived from secondary raw materials in semi-arid and Mediterranean European regions. Given their low water-soluble P fraction, the soil moisture patterns probably have a negligible impact on the solubility of P-fertilisers derived from secondary raw materials. The solubility of those fertilisers is mainly determined by the extent of root exudation of the plants grown on the agricultural field. It can, however, be expected that the solubility of mined and synthetic P-fertilisers is increased in the more northern latitudes characterised by more moist soils due to the increased precipitation. Therefore, the agronomic efficiency of mined and synthetic P-fertilisers could be higher for the higher latitudes, resulting in decreased relative agronomic efficiency values in the more northern regions. Other soil properties that vary across latitude, such as soil texture and soil pH, did not have a significant effect on the relative agronomic efficiency for the P-fertilisers under study.

4 Conclusion

This work is important as it reviews for the first time the agricultural efficiency of different P-fertilisers derived from secondary raw materials that show a significant potential to substitute mined rock phosphate and processed P-fertilisers in Europe (Huygens and Saveyn 2017). The meta-analysis estimates suggest that selected P-fertilisers derived from secondary raw materials may compare in agronomic efficiency with mined and synthetic P-fertilisers. Specifically, our results demonstrate that the agronomic efficacy of precipitated phosphate salts and specific thermal oxidation materials and derivates is consistent for different soil and plant types and is thus not restricted to specific agricultural settings within a European context. In spite of their low water solubility, specific P-fertilisers derived from secondary raw materials could be a valuable alternative for mined rock phosphate and processed P-fertilisers in the conventional European agriculture. Applications for all studied P-fertilisers derived from secondary raw materials are also apparent for the expanding organic farming sector in Europe; at present, meat and bone meal and their ashes and low concentrated P-fertilising products, such as manure and compost, are the sole P-rich fertilising materials used in organic farming (Nelson and Janke 2007). Phosphorus-recycling from vastly dissipated P-sources, such as municipal and industrial wastewaters and manure in the form of P-fertilisers, is an apt manner to transport P in a concentrated form over long distances (e.g. from livestock and demographically dense regions in north-west Europe to more southern European regions with increased P-fertiliser needs; Tóth et al. 2014). Based on the assessment of agronomic efficiency, it is concluded that an increased use of selected P-fertilisers derived from secondary raw materials in European agriculture could contribute to decreased P dissipation and more circular nutrient cycles.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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