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**Phosphorus bioavailability of fertilizers recycled from sewage
sludge and their suitability for organic crop production**

Dissertation

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Abbreviations

BNF	biological nitrogen fixation
DM	dry mass
EBPR	enhanced biological P removal
MCP	monocalciumphosphate, $\text{Ca}(\text{H}_2\text{PO}_4)_2$
MEPHREC®	metallurgisches Phosphor-Recycling
Mg-SSA	sewage sludge ash, treated with magnesium in the ASH DEC® process
Na-SSA	sewage sludge ash, treated with sodium in the ASH DEC® process
PGPR	plant growth promoting rhizobacteria
PR	phosphate rock
P-RoC	phosphorus recovery from waste water by crystallization
PSR	phosphate solubilizing rhizobacteria
PTE	potentially toxic elements
PUE	phosphorus use efficiency
SE	standard error
SEM	standard error of the mean
SSA	sewage sludge ash
Struvite	magnesium ammonium phosphate, $\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$

List of abbreviations

Struvite AirPrex® Struvite produced in the AirPrex® process by Berliner Wasserbetriebe

Struvite SSL Struvite produced in the Stuttgart (Sludge Leaching) process by Stuttgart University

1 General Introduction

1.1 Introduction

In 1669, when phosphorus (P) was first discovered by alchemist Hennig Brand, it was thought of nothing less than an additive for the production of gold. Nowadays, the value of P is even higher, as it is known to be an essential nutrient for all life on earth. In agricultural crop production, P nutrition of plants is a key production factor (Nesme et al., 2014). For centuries, crop production was based on natural reserves of soil P and the application of locally produced manure and human excreta. This ancient nutrient cycle was disrupted with the intensification of agriculture in the 20th century, when higher P inputs were required to secure increased crop yields for a fast growing world population. Phosphorus fertilizers based on Guano and Phosphate Rock (PR) were applied in agriculture, while untreated human excreta were no longer returned to the fields, but, with the introduction of flush toilets, discharged into water bodies (Cordell et al., 2009). Today, approximately 1.71×10^7 Mg P from mineral reserves are applied to soil globally every year (Yuan et al., 2018). On a global scale, only approximately 20% of P contained in human excreta are directly recycled to agriculture (Yuan et al., 2018). In fear of contaminating soils with potentially toxic elements (PTE) and organic pollutants, the application of sewage sludge to land is strictly limited by law in European countries. In organic farming systems of the European Union, direct sludge applications are completely forbidden (Council Regulation EC No. 889/2008) and P management represents a particular challenge. The only permitted mineral P fertilizer in organic farming is PR, which shows poor bioavailability on many soils (Guppy and McLaughlin, 2009; Nesme et al., 2012; Möller et al., 2018). Phosphate Rock is mined

from deposits all over the world. More than 80% of P from PR is used for fertilizer production (Smil, 2000). However, PR is a limited resource, decreasing in grade, accessibility, and purity (Schröder et al., 2011), and future availability of the world's main source of P is debated as uncertain (Cordell and White, 2011). Also, due to the geographical concentration of PR ores, western Europe, which has very low domestic supply, is heavily dependent on imports. Thus, the debate on reclosing the P cycle has gained importance for governmental institutions, researchers and farmers. Of the P contained in animal manure, up to 82% (approximately 1.56×10^7 Mg yr⁻¹) is already recycled to soil in Western European countries (Yuan et al., 2018). However, there are great spatial differences due to the unbalanced distribution of livestock density and high transportation costs of manure. Approximately one third of the P applied to soil as mineral fertilizer ends up in human consumption, most of which within processed food (Yuan et al., 2018). Of the daily ingested P, 98% is excreted (Yuan et al., 2018), thus, municipal waste water is the largest non-agricultural source for the recovery of P (Oelofse et al., 2013; Egle et al., 2015; van Dijk et al., 2015). Nowadays, the majority of P in sewage sludge is disposed of in landfills and becomes unavailable for further use. However, new technologies have been developed, with the aim to recycle P from urban areas back to agriculture (Egle et al., 2016) in order to close this essential gap of the P cycle.

1.2 Phosphorus recycling from waste water

In the European Union, 11 million Mg of sewage sludge dry mass (DM) is produced every year (Gendebien 2010; Stemmann et al. 2014), providing more than 300,000 Mg P per year, which corresponds to around 20% of the mineral fertiliser input per year

(EU-27, 2005) (Stemann et al. 2014; van Dijk et al., 2016). The use and treatment of sewage sludge hugely differs between member states. National limit values for potentially toxic elements, pathogens and organic micropollutants in sewage sludge which is used in agriculture, widely differentiate among countries (Kelessidis and Stasinakis, 2012) and practically determine their way of sludge disposal. In Germany, direct sludge application to agriculture is strictly regulated. In 2016, 1.8 million Mg of sewage sludge DM were produced (containing approximately 49,000 Mg of P), of which only 24% were directly recycled to agriculture (Statistisches Bundesamt, 2018). This leaves a P recycling potential of approximately 38,000 Mg every year, which corresponds to 38% of the domestic P sales in Germany in 2016/2017 (Statistisches Bundesamt, 2018). In the coalition agreement 2013, the provision is made for phasing out direct sludge application until year 2025, being replaced by a P recycling from sewage sludge.

There are several technologies to recover P from municipal wastewater, producing materials that contain varying quantities of P in different chemical forms, while often containing reduced amounts of potentially harmful substances. Depending on that, they can be used as raw material for P fertilizer production or might directly be used as P fertilizers in agriculture. Phosphorus recovery can take place from the liquid phase of sewage sludge by chemical precipitation processes, or from the sludge ash after a mono-incineration (Cornel and Schaum, 2009; Egle et al., 2016). In dependency of the production process, different P fertilizers recycled from sewage sludge hugely differ in their P availability for plants. For example, P in sewage sludge ash (SSA) is usually in the form of aluminum and calcium phosphates and its bioavailability strongly depends

on soil pH (Cabeza et al., 2011; Nanzer et al., 2014a; Möller et al., 2018). Thus, for most ashes, an additional treatment is necessary to increase its bioavailability and to reduce contents of potentially toxic elements (PTE) (Krüger et al., 2014). Other recycled fertilizers, such as e.g. Struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6 \text{H}_2\text{O}$), have been shown to be effective P fertilizers under a wide range of soil pH (Cabeza et al., 2011).

An overview over a broad range of different chemical and thermal P recycling technologies has been published by Egle et al. (2015). Fertilizer efficacy of different recycled fertilizers have been studied in few experiments (Römer, 2006; Cabeza et al., 2011; Antonini et al., 2012; Vogel et al., 2015; Talboys et al., 2016), all of them focusing on their P fertilization effect on single crops, mostly under controlled conditions in the greenhouse. There is no data available, investigating the effects of recycled P fertilizers in a cropping system approach, addressing their consecutive effects on several and diverse crops of a representative cropping sequence. Also, there is very little data available on the efficacy of recycled P fertilizers under field conditions, which is needed to evaluate their usability in agricultural crop production.

1.3 Phosphorus in soil – bioavailability and mobilization

Although total soil P content can be very high, the bioavailability of this essential nutrient is low in many soils. Among all macronutrients taken up by plant roots, P has the lowest solubility (Gerke, 1995). The soil solution in agricultural soils, which is the main source of P for plant roots, contains between 0.01 and 3.0 mg P L⁻¹ (Frossard et al., 2000), which corresponds to less than 1% of total soil P (Richardson et al., 2009b). Of all the different P forms present in soil, solely orthophosphate (HPO_4^{2-} and H_2PO_4^-

) can be taken up by plants. However, when applied to soil, (reactive) fertilizer P undergoes a rapid interchange with the soil matrix. Phosphate is inexorably drawn to sorption surfaces or precipitated, and is removed from the readily plant available P pool (Guppy and McLaughlin, 2009), a process referred to as “fixation”, “sorption” or “retention” in literature. Phosphate is then predominantly associated with calcium (Ca) in alkaline soils and with iron (Fe) or aluminum (Al) in acidic soils, and P anions are adsorbed to soil constituents, such as sesquioxides, Al-silicates, Ca-carbonates, clay minerals and organic matter, through charge-related associations (Sanyal and De Datta, 1991; Richardson, 2001; Richardson et al., 2009b). Once adsorbed or precipitated, P forms show varying, but poor solubility. Thus, the use efficiency of applied mineral fertilizer P is conceivably low, reaching 10 – 20% of applied P (from superphosphate) in the year of application, and a maximum of only 50% over time (Holford, 1997; Richardson et al., 2009b), which inevitably leads to a P accumulation in managed soils (George et al., 2016).

Other P pools besides mineral P (adsorbed or in soil solution) are organic and microbial biomass P. In the upper layer of arable soils, organic P accounts for 20 – 80% of total soil P (Dalal, 1977) and mainly comprises inositol phosphates, notably hexakisphosphate (phytate), phospho-lipids and nucleic acids (Nesme et al., 2014). A mobilization of organic P for plant uptake can take place through hydrolysis by phosphatase-like enzymes, which are produced by plants and, more so, by many soil microorganisms (Nesme et al., 2014). The P contained in soil microorganisms is referred to as microbial biomass P and amounts to 0.4 – 2.5% of total P in arable soils (Bünemann et al., 2011). Although microbial P constitutes to a small amount of total P

only, it can play a major role in the P cycle due to its often very short turnover time (Achat et al., 2010; Bünemann et al., 2011).

As orthophosphate is taken up from the soil solution, a steady interchange between the different P pools takes place, transferring P between the solid phase and soil solution by sorption-desorption processes, and between organic and inorganic P forms by immobilization-mineralization processes (Frossard et al., 2000). These processes are strongly influenced by soil pH, which governs the adsorption properties of major P fixing minerals and the solubility and dissolution kinetics of a variety of P containing minerals (e.g. apatite), as summarized by Devau et al. (2011). However, diffusion and mass flow of P from bulk soil to the rhizosphere is low (< 1 mm over a few days (Hinsinger et al., 2005)), and generally, the replenishment of the soil solution is not sufficient to meet plant requirements (Richardson et al., 2009b). Therefore, biological P mobilization mechanisms in the rhizosphere play a major role in the P supply of plants.

There are several natural, physiological and morphological responses of plants to P deficiency, such as the ability of roots to effectively explore soil volume, the release of exudates from roots that influence soil P availability, and the association of roots with soil microorganisms such as mycorrhizal fungi (Richardson et al., 2009b). In addition, there are agricultural practices to optimize the P efficacy of fertilizers, including the design of fertilizer granules to hinder P fixation, the strategic placement and banding of P fertilizers within the soil (Richardson et al., 2009b), and an application of

rhizobacteria to improve P availability and accessibility by plants (Rodríguez and Fraga, 1999; Rodríguez et al., 2006).

The potential beneficial effects of rhizobacteria for growth and P acquisition of plants have been described by e.g. Rodríguez and Fraga (1999), Richardson (2001), Richardson et al. (2009a), Miransari (2014) and Weinmann (2017). Few studies have shown the efficacies of different rhizobacteria in P solubilization (Achal et al., 2007; Pérez et al., 2007; Hameeda et al., 2008; Collavino et al., 2010; Yu et al., 2011; Kaur and Reddy, 2014; Liu et al., 2014; Pereira and Castro, 2014; Nkebiwe et al., 2017). Among them, *Bacillus*, *Pseudomonas* and *Rhizobium* strains are the most promising in terms of P solubilization (Rodríguez and Fraga, 1999; Nkebiwe et al., 2017). Different *Pseudomonas* strains have been shown to increase grain yield of maize by up to 20%, and total P in shoot (+ 22%), root (+ 42%) and grain (+ 26%) of maize compared to unamended soil in a pot experiment (Pereira and Castro, 2014) and in the field (Kaur and Reddy, 2013). In the latter, phytase activity in the rhizosphere of maize treated with *Pseudomonas* was almost doubled, compared to unamended soil (Kaur and Reddy, 2013). Many plant growth promoting rhizobacteria (PGPR) and fungi have been isolated to develop commercially available products as inoculants (Glick, 2012; du Jardin, 2015), afterwards referred to as “bioeffectors”. However, although a range of studies on bioeffectors showed increased growth and P content of plants, variability of the results is large and has often shown poor reproducibility (Thonar et al., 2017). As a consequence, commercial applications of bioeffectors have been limited (Glick, 2012). Still, they could play a key role in enhancing P availability of less labile P forms, such as those obtained in different P fertilizers recycled from sewage

sludge. This has, apart from few studies (Lekfeldt et al., 2016; Meyer et al., 2017) rarely been tested so far.

Another strategy to improve P efficiency is, to increase the pool of plant available P within a farming system through the use of P efficient crops in the crop rotation (Kamh et al., 1999; Richardson et al., 2009b). Some legume species have been shown to mobilize non-reactive soil P from less available soil P sources (Hauter and Steffens, 1985; Nuruzzaman et al., 2005a). Thereby, they contribute to an increased cycling of reactive P (Horst et al., 2001; Guppy and McLaughlin, 2009; Güldner and Krausmann, 2017) and an improved P supply of a following crop in the rotation (Nuruzzaman et al., 2005b). Underlying mobilization mechanisms of legume species are root excretions of carboxylates or acid phosphatases (e.g. white lupin, field pea, faba bean) (Nuruzzaman et al., 2005b, 2006; Sugihara et al., 2016). In addition, in soils with pH > 7.0, P availability might be enhanced through a rhizosphere acidification, resulting from a proton release during biological N₂-fixation of legumes (Hauter and Steffens, 1985; Hinsinger et al., 2003). There is a lack of data investigating P fertilizers recycled from sewage sludge in combination with biological means of P mobilization in order to improve their bioavailability to plants.

1.4 Phosphorus management in organic farming

The challenge of securing an adequate P supply for plants in agricultural production is even aggravated in organically managed systems, where highly soluble P fertilizers, such as monocalciumphosphate (MCP), are prohibited. However, P supply of crops is especially important for legumes, which are the main pillar of fertility management in

organic systems, due to their ability to fix atmospheric N₂ (Römer and Lehne, 2004). Except for some P containing organic amendments, such as animal manure, organic residues (composted or digestated) or meat and bone meal (MBM), the only external mineral P source permitted in the EU to balance P offtakes in organic systems is PR (Løes et al., 2017). Phosphate Rock has not only poor solubility in all soils with pH > 6.0, making them poorly efficient in neutral and alkaline soils that are common in European agricultural regions (Nesme et al., 2014), but may also contain relevant loads of potentially toxic elements (PTE) (Weissengruber et al., 2018). Thus, it has been reported that yields in organic systems have so far partly been attained at the expense of soil P reserves or residual P from earlier fertilizer applications (Oehl et al., 2002; Nesme et al., 2012; Ohm et al., 2017), which questions their sustainability in the long term. Hence, to maintain current yields, there is a need for plant available P fertilizers for organic crop production.

Today, 80 – 90% of societal recoverable P sources for recycling are banned in organic systems in order to reduce the risk of importing contaminants (Løes et al., 2017). This includes all P fertilizers recycled from human excreta. However, a serious soil contamination with PTE through the use of recycled P fertilizers in agriculture is not likely (Magid, 2012; López-Rayó et al., 2016; Weissengruber et al., 2018). According to the principles of IFOAM (International Federation of Organic Agricultural Movements), farm inputs should be reduced by reuse and recycling in order to conserve natural resources (IFOAM, 2017). Mainly in stockless organic systems though, off-farm inputs are required to balance P exports via sold products, to maintain a sustainable production in the long term (Nesme et al., 2014). That is why some

scientists suggest that organic production regulations should be adapted to this new farming type or should reassess allowable farm P inputs (Cornish and Oberson, 2008; Nesme et al., 2014; Cooper et al., 2018), and that nutrient cycling should be addressed and assessed at a larger, e.g., district, scale (Nesme et al., 2012). Today, there are only few studies available, assessing the suitability of mineral P fertilizers recycled from sewage sludge for their use in organic crop production (Løes, 2016; Möller, 2016; Løes et al., 2017; Weissengruber et al., 2018).

1.5 Aims and objectives

Within the EU project “**IM**proved **P**hosphorus **R**esource efficiency in **O**rganic agriculture **V**ia recycling and **E**nhanced biological mobilization” (IMPROVE-P), P fertilizers recycled from different waste streams were assessed for their suitability to be used in organic farming systems. Examined aspects in the project included the P bioavailability and the load of potentially toxic elements in recycled fertilizers, the test of different P mobilization techniques as well as a survey of the acceptance of recycled P fertilizers among organic stakeholders and farmers. The research presented here was carried out within the framework of IMPROVE-P, with the aim to evaluate mineral P fertilizers recycled from sewage sludge with regard to their fertilizer efficacy and suitability for organic crop production, combined with different biological P mobilization approaches. The objectives of this research were:

- (1) to evaluate the agronomic efficacy of P fertilizers recycled from sewage sludge for maize, in pot experiments and on a field scale, compared to PR and MCP fertilization,

- (2) to assess the agronomic efficacy of such P fertilizers for several crops of a representative cropping sequence of organic agriculture, including a legume species,
- (3) to investigate the P mobilization potential of red clover within a crop rotation,
- (4) to investigate different bacteria strains for their ability to increase plant P availability of recycled P fertilizers in pot experiments and on a field scale, and
- (5) to evaluate the suitability of P fertilizers recycled from sewage sludge for organic crop production, from an agronomic point of view.

1.6 Structure of the thesis

Apart from the General Introduction and Discussion, this thesis includes four chapters that attend to the aims and objectives stated above. These chapters comprise three original research manuscripts that have been published in international peer-reviewed journals (chapter 2, 3, 5) as well as one original research paper that was prepared for submission (chapter 4).

Chapter 2: **Phosphorus bioavailability of sewage sludge-based recycled fertilizers.** Wollmann, I., Gauro, A., Müller, T., Möller, K. (2018) Reprinted from the Journal of Plant Nutrition and Soil Science, 181, 158 – 166. With permission from Wiley.

Chapter 3: **Phosphorus bioavailability of sewage sludge-based recycled fertilizers in an organically managed field experiment.** Wollmann, I., Möller, K. (2018) Reprinted from the Journal of Plant Nutrition and Soil Science, 181, 760 – 767. With permission from Wiley.

Chapter 4: **Red clover (*Trifolium pratense* L.) increases plant growth and P offtake from sewage sludge ashes for a subsequent maize.** Wollmann, I., Möller, K.

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Chapter 5: **Improved Phosphorus Recycling in Organic Farming: Navigating Between Constraints.** Möller, K., Oberson, A., Bünemann, E.K., Cooper, J., Friedel, J., Glæsner, N., Hörtenhuber, S., Løes, A.-K., Mäder, P., Meyer, G., Müller, T., Symanczik, S., Weissengruber, L., Wollmann, I., Magid, J. (2018) Reprinted from *Advances in Agronomy*, 147, 159 – 237. With permission from Elsevier.

2 Phosphorus bioavailability of sewage sludge-based recycled fertilizers

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Phosphorus bioavailability of sewage sludge-based recycled fertilizers

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Abstract

Six phosphorus (P) fertilizers recycled from sewage sludge [Struvite SSL, Struvite AirPrex[®], P-RoC[®], Mephrec[®], Pyrolysis coal and Ash (Mg-SSA)] were tested for their plant availability in potted soil of pH 7.2 under greenhouse conditions. The crop sequence simulated a rotation of red clover (*Trifolium pratense* L.), maize (*Zea mays* L.), and ryegrass (*Lolium perenne* L.). Other P fertilizer treatments included: Phosphate Rock (PR), Calcium dihydrogen phosphate [$\text{Ca}(\text{H}_2\text{PO}_4)_2$], and an unfertilized control. Additionally, soil was regularly inoculated with two strains of plant growth-promoting rhizobacteria (PGPR; *Pseudomonas* sp. Proradix, and *Bacillus amyloliquefaciens*) to test their ability to increase P availability to plants. Sequential P fractionation was conducted to link the amount of readily available P in fertilizers to plant P acquisition. Shoot P content and dry matter of maize decreased in the following order: Struvite SSL \geq $\text{Ca}(\text{H}_2\text{PO}_4)_2$ $>$ P-RoC[®] \geq Struvite AirPrex[®] \geq Mephrec[®] $>$ Pyrolysis coal \geq Mg-SSA \geq PR \geq unfertilized. Rhizobacteria did not affect shoot biomass or P content. The results show that red clover might have mobilized substantial amounts of P. Sequential P fractionation was not suitable to predict the efficacy of the fertilizers. Generally, the sewage sludge-based fertilizers tested proved to be suitable alternative P sources relevant to organic farming systems. However, the efficacy of recycled fertilizers is strongly dependent on their specific production conditions.

Key words: crop rotation / phosphorus fertilization / phosphorus mobilization / red clover / rhizobacteria / struvite

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1 Introduction

Phosphorus (P) is a key production factor in agriculture. In certified organic arable farming systems, soil P management is a challenge (Guppy and McLaughlin, 2009) due to the self-imposed regulation that prohibits P fertilization with water-soluble mineral P fertilizers, thus, leading to P imbalances (Friedel et al., 2014; Kolbe, 2015; Tittarelli et al., 2016; Zikeli et al., 2017). The main mineral P source obtained externally and applied to balance field P outputs in organic farming is phosphate rock (PR) (Berry et al., 2003; Council Regulation EC, 2007). However, PR, a non-renewable resource, shows poor plant availability in slightly acidic to alkaline soils (Boland et al., 1997; Möller et al., 2017) that are common in European agricultural regions. Therefore, it is crucial to recycle P from the human food chain back into crop production. In the European Union (EU), 11⁶ t of sewage sludge dry mass are produced every year (Gendebien, 2010). This provides more than 300,000 t of P per year which corresponds to around 20% of the mineral fertilizer input of the EU-27 in 2005 (Stemann et al., 2014; van Dijk et al., 2015). Direct use of sewage sludge in organic agriculture is forbidden in the EU (Commission Regulation EC, 2008) assuming a risk of contamination by harmful substances (Geier et al., 2007). Nevertheless, organic farming principles postulate efficient utilization and recycling of nutrients (Möller et al., 2017). Several

chemical and thermal technologies have been developed to recover P from sewage sludge, thus, generating nutrient-rich fertilizers with low levels of harmful substances (Egle et al., 2015; Schoumans et al., 2015).

Besides recycling P, efficient P use comprises effective utilization of less labile soil and fertilizer P sources through P mobilizing approaches. Soil microorganisms influence plant P availability by mineralization and solubilization of soil P (Rodríguez and Fraga, 1999; Lavakush et al., 2013) and a stimulation of root growth to expand accessible P pools (Jakobsen et al., 2005). Various plant growth-promoting rhizobacteria (PGPR) have been isolated to develop bioeffector products as inoculants to improve P acquisition by plants (Glick, 2012). Among them, *Bacillus* and *Pseudomonas* strains are most promising in terms of P solubilization (Rodríguez and Fraga, 1999; Zabihi et al., 2011). These kinds of bioeffectors have not yet been investigated for their P solubilization ability of recycled P fertilizers from sewage sludge.

For nutrient management in organic farming, crop rotations are of particular importance due to their use of N₂-fixing legumes (Römer and Lehne, 2004). Some legume species are adapted to mobilize sparingly available soil P sources (Hauter and Steffens, 1985; Hinsinger and Gilkes, 1996; Nuruzzaman et al., 2005a) and improve the P supply of a fol-



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lowing crop in the rotation (Nuruzzaman et al., 2005b). Thus, with regard to the high share of legumes in typical crop rotations in organic farming systems, there is a potential for using recycled P fertilizers from sewage sludge including those with low P bioavailability.

In the present study, P availability of six fertilizers recycled from sewage sludge was investigated in two pot experiments. One of them represented a typical cropping sequence of organic farming. In addition, two bioeffector products were tested for their ability to increase bioavailable P from soil and fertilizers. Recycled fertilizers were investigated for their solubility in water and citric acid and sequentially extracted in NaHCO_3 , NaOH , and H_2SO_4 . The amount of NaHCO_3 -extractable P in fertilizers was assumed as the plant-available (labile) P fraction (Hedley et al., 1982; Vogel et al., 2015), and linked to the bioavailability of fertilizers tested in pot experiments. We hypothesized that (1) the tested recycled P fertilizers have a higher P bioavailability than PR, (2) P bioavailability of the tested recycled fertilizers in soil can be linked to their amount of NaHCO_3 -extractable P, (3) P bioavailability of applied fertilizers is enhanced when tested in a crop rotation including legumes, and (4) the application of the tested bioeffector products leads to higher plant P availability compared to treatments without bioeffectors.

2 Material and methods

2.1 Tested P fertilizers and bioeffector strains

Six P fertilizers recycled from sewage sludge were tested for their P availability in two pot experiments: Struvite SSL was produced in a pilot plant by the Department of Waste Water Management (ISWA, Stuttgart University, Germany). In the SSL process, a chemical re-dissolution of Ca-, Mg-, Fe-, Al-, and other metal phosphates from digested sewage sludge is achieved by a pH decrease (pH 4) through the addition of sulfuric acid. Afterwards, a solid-liquid separation takes place. Citric acid is added as a chelating agent to the liquid fraction in order to keep potentially toxic elements and other elements interfering with P precipitation (e.g., Ca, Fe, Al) in solution. Then struvite crystallization is triggered by the addition of magnesium oxide and the increase of pH to 7.0–8.5 through the addition of sodium hydroxide (Steinmetz et al., 2014; Herzel et al., 2015). Struvite AirPrex® is commercially available and produced by Berliner Wasserbetriebe (BWB Berlin, Germany). In the AirPrex® process, struvite precipitation takes place directly from digested sewage sludge without a previous chemical re-dissolution step. An increase of pH to 8 is achieved by CO_2 -stripping and magnesium chloride is added to trigger struvite crystallization (Kern et al., 2008; Ewert and Wagenbach, 2014; Herzel et al., 2015). Besides precipitation of struvite, calcium phosphates (Ca-P) are formed (Kern et al., 2008). In the P-RoC® process (Phosphorus Recovery from Wastewater by Crystallization), developed by Karlsruhe Institute of Technology (KIT, Karlsruhe, Germany), an increase of the pH of process water to 9–10 is achieved by the addition of calcium silicate hydrate (CSH), which releases hydroxide ions. Struvite and water-insoluble Ca phosphates (including hydroxyapatite or brushite crystals)

are then formed on the surface of the CSH pellets (Berg et al., 2007; Petzet and Cornel, 2009). Mephrec® is a P-containing slag produced from dewatered, dried and compressed sewage sludge in a melt-gassing process at 2000°C under the addition of lime, based on the MEPHREC® process (INGI-TEC GmbH, Leipzig, Germany; Scheidig et al., 2009). Quartz is added to influence the P speciation (Möller, 2016) and phosphates contained in sewage sludge are transformed into silico-phosphates comparable with the so-called Thomas phosphate stemming from the Thomas process. Pyrolysis coal was produced from sewage sludge in an oxygen-free atmosphere at 500°C by Pyreg GmbH (Dörth, Germany). Prior to thermal treatment, calcium chloride (15%, based on the ash content) was added to the sewage sludge in order to increase the plant P availability of the produced coal (Peplinski et al., 2009). The production of Mg-containing sewage sludge ash (Mg-SSA) from mono-incineration of sewage sludge was conducted based on the ASH DEC® process (Outotec, Finland). Sewage sludge ash is thermochemically treated with MgCl_2 in a rotary kiln for 15 min at 950°C (Hermann and Schaaf, 2016) in order to remove potentially toxic elements as chlorides via the gaseous phase while producing calcium-magnesium-phosphates in the final ash (Adam et al., 2009). The total P concentration contained in recycled fertilizers is listed in Tab. 1.

Two commercially available bioeffector products were assessed in the experiments for their ability to increase plant availability of P from soil and fertilizers. The products were the bacteria *Bacillus amyloliquefaciens* strain FZB42 contained in RhizoVital® 42 fl. (ABITEP Berlin, Germany) and *Pseudomonas* sp. “Proradix” (Proradix, Sourcon Padena, Tübingen, Germany). Both products were prepared and applied as a suspension in 0.025 M CaSO_4 . Bioeffector stock suspensions were freshly prepared before each application. The application rate of bacteria was based on results of an internal preliminary test. An inoculum containing 2×10^8 colony-forming units (cfu) was applied fortnightly to clover and maize in Experiment 1, and to maize in Experiment 2. An amount of 15 mL of bioeffector suspension was pipetted onto the soil surface and washed into the soil with 15 mL deionized water. The first application date of bioeffectors was 1 d after sowing. On each application date, treatments without bioeffector received 15 mL of 0.025 M CaSO_4 solution. For the ryegrass in Experiment 1, the application strategy was changed towards a less frequent, but higher concentrated application rate of 2×10^9 cfu in three single applications 0, 2, and 4 weeks after sowing, respectively.

2.2 Experimental setup

The substrate used in both experiments was a 2 : 1 mixture of a silty sandy loam soil and silica sand. The soil used was a fresh top soil obtained from grassland. It was sieved and stored at 15°C for 3 and 8 months before the start of the first and second experiment, respectively. Soil chemical and physical properties included: pH: 7.2 (CaCl_2 ; VDLUFA, 1991), total P: 55.4 mg P 100 g⁻¹ DM, P_{CAL} : 2.20 mg P 100 g⁻¹ DM, carbonates: 4.7%, organic carbon: 1.8%, clay: 19.2%, silt: 48.8%, sand: 32.0%. Experiments were performed in 1.8 L pots, containing 1.08 kg dry soil and 0.54 kg dry sand. During

Table 1: Total P content (mg P g DM⁻¹) and P contained in different fractions of sequentially extracted P (NaHCO₃, NaOH, H₂SO₄), indicated as total amounts (mg P g DM⁻¹) and relative quantities (% of total P, bold numbers in parentheses) and solubility of P in water (H₂O) and 2% citric acid (% of total P) of investigated P fertilizers (PR = Phosphate Rock, Struvite SSL = struvite obtained from Stuttgart Sewage Leaching Process, Struvite AirPrex[®] = struvite obtained from the AirPrex[®] process, P-RoC[®] = Ca-Phosphates recovered in the P-RoC[®] process, Mephrec[®] = P recovered in a melt-gassing process (MEPHREC), Pyrolysis coal = P recovered through pyrolytic treatment of sewage sludge, Mg-SSA = Mg-treated sewage sludge ash).

Tested fertilizers	Total P ^a (mg P g DM ⁻¹)	Sequentially extracted P			P _{H₂O} (% of total P)	P _{2% citric acid}
		NaHCO ₃ (mg P g DM ⁻¹) and (% of total P)	NaOH (mg P g DM ⁻¹) and (% of total P)	H ₂ SO ₄ (mg P g DM ⁻¹) and (% of total P)		
Struvite SSL	233	3.89 (2.98)	20.7 (15.8)	106 (81.2)	6.56	56.1
Struvite AirPrex [®]	223	3.09 (2.63)	20.7 (17.6)	93.6 (79.7)	0.89	37.1
P-RoC [®]	49.8	2.83 (7.29)	0.58 (1.50)	35.5 (91.2)	0.14	67.8
Mephrec [®]	43.1	0.31 (0.65)	0.18 (0.38)	47.6 (98.9)	0.08	53.9
Pyrolysis coal	56.0	0.69 (1.31)	11.8 (22.5)	40.0 (76.2)	0.07	43.1
Mg-SSA	62.8	0.68 (0.98)	1.45 (2.09)	67.3 (96.9)	0.21	67.9
PR	136	0.00 (0.00)	0.00 (0.00)	129 (100)	0.01	21.6

^aDetermined after drying of fertilizers and aqua regia extraction, according to VDLUFA (2000) and measured with ICP-OES.

the duration of the experiments, temperature conditions in the greenhouse ranged between (mean monthly temperature) 17.4°C (January 2015) and 24.8°C (July 2014). Additional light was switched on during the months October to March for 9 h d⁻¹ with an average light intensity of 430 μmol m⁻² s⁻¹, measured at the height of plant shoot-tips. Plants were watered daily with deionized water to maintain 50% water-holding capacity. After each harvest, shoot biomass was dried at 60°C and DM content was determined. Soil and roots from each pot were mixed thoroughly and returned to the pots. When dissected, clover root nodules showing a reddish inside coloration indicated a functioning N₂-fixing symbiosis.

Experiment 1 was performed between November 2013 and April 2015 with a cropping sequence of red clover (*Trifolium pratense* L., cv. Astur, Delley Samen und Pflanzen AG, Switzerland), maize (*Zea mays* L., cv. Colisee, KWS Saat SE, Germany), and perennial ryegrass (*Lolium perenne* L., cv. Arvicola, Feldsaaten Freudenberger GmbH & Co., Germany). Struvite SSL and Mg-SSA were tested as recycled P fertilizers beside PR, Ca(H₂PO₄)₂, and an unfertilized control. Phosphorus fertilizers were ground, homogenized, and applied by mixing with soil prior to sowing at a rate of 32 mg P kg soil⁻¹ DM. Other nutrients were applied in amounts to secure sufficient supply with (per kg soil DM) 100 mg potassium (K) as K₂SO₄, 50 mg Mg as MgSO₄ and 35 mg N as Ca(NO₃)₂. Nitrogen and Mg amounts contained in recycled fertilizers were considered in the N and Mg fertilization. After the low initial N application, N supply to clover was assumed to be covered symbiotically. Clover seeds were inoculated with RADICIN-Trifol[®] (Jost GmbH, Iserlohn, Germany) to ensure rhizobia colonization. Four red clover plants were grown per pot. Above-ground biomass was harvested 4, 6, and 8 months after sowing. After clover harvest, the soil was kept at 50% water-holding capacity during a seven-week fallow period. Subsequently, one maize plant was grown per

pot. During maize growth (September–December 2014), the soil was fertilized twice with 200 mg N kg⁻¹ soil DM as NH₄NO₃, respectively and with 200 mg K kg⁻¹ soil DM as K₂SO₄. In January 2015, ryegrass was sown at a rate of 4 g per pot and cultivated for 9 weeks. In Experiment 1, both bioeffector products were applied regularly throughout the experiment as described above.

In Experiment 2, all recycled P fertilizers (Tab. 1) were tested on maize cv. Colisee beside PR, Ca(H₂PO₄)₂, and an unfertilized control. Application rates of P fertilizers, K and Mg were identical with pot Experiment 1. Taking into account the N amount applied with the two struvite products, 200 mg N kg⁻¹ soil DM were applied as NH₄NO₃. From March 2014 onwards, one maize plant was cultivated per pot for 8 weeks. In Experiment 2, only the bioeffector product containing the *Bacillus* strain was tested.

2.3 Chemical characterization of recycled P fertilizers

Solubility of P in recycled P fertilizers was tested in water and 2% citric acid according to VDLUFA (1995a). Phosphorus concentration was measured photometrically (U-3300, Hitachi, Japan) using the vanadate-molybdate method (Gericke and Kurmies, 1952; VDLUFA, 1995b). Sequential P fractionations of recycled P fertilizers were conducted according to a modified Hedley fractionation method (Hedley et al., 1982; Tiessen and Moir, 1993). From each product, 0.5 g of air-dried material was weighed into 50 mL centrifuge tubes, 30 mL of 0.5 M NaHCO₃ were added, and tubes were shaken at 180 rpm for 16 h. Suspension was centrifuged at 2500 g for 10 min and NaHCO₃ supernatant was decanted through blue-band filter paper. Remaining particles in filter paper were washed back to the tube using 0.1 M NaOH, which was then filled up to 30 mL, shaken, centrifuged, and filtered according

to the previous procedure. The same procedure was conducted with 30 mL of 1 M H_2SO_4 . Total P concentration of the extracts was measured using ICP-OES (VISTA Pro, Varian inc., USA).

2.4 Phosphorus concentration in plant tissue

Dry plant material was ground and extracted in concentrated HNO_3 using the wet chemical digestion method (VDLUFA, 2011). P concentration was measured photometrically (U-3300, Hitachi, Japan) according to Gericke and Kurmies (1952), and shoot P content was calculated as product of shoot DM and shoot P concentration.

2.5 Data analysis

Both experiments were arranged in a completely randomized design with pot rotations. They included 15 treatments with five replicates (Experiment 1), and 18 treatments with four replicates (Experiment 2). Two-way analysis of variance (ANOVA) was performed (SigmaPlot 11.0) to study the effect of different P fertilizers and bioeffectors and their interaction on shoot DM and P content of the test plants in both experiments. All pairwise multiple comparison of the means was conducted using a Tukey test ($P \leq 5\%$).

3 Results

Phosphorus solubility of recycled P fertilizers in H_2O was $< 1\%$ of total P for all fertilizers except Struvite SSL (6.56% of total P). Phosphorus solubility in citric acid (% of total P) ranged between 21.6 (PR) and 67.9 (Mg-SSA) (Tab. 1). All recycled fertilizers contained a small fraction of NaCO_3 -extractable P. Slightly higher amounts of P were obtained by extraction with NaOH. The fraction with the highest share of P in all recycled fertilizers was extracted with H_2SO_4 . PR consisted of 100% H_2SO_4 -extractable P (Tab. 1).

Application of bioeffectors did not influence shoot DM and P content of the test plants in Experiment 1 (Fig. 1) and Experiment 2 (Fig. 2). These results were consistent throughout all harvest dates of clover, maize, and ryegrass in Experiment 1. No significant interactions were found between fertilizer and bioeffector treatments. Thus, results are shown as mean values of shoot DM and P content separately according to P fertilizer or bioeffector treatments. In Experiment 1, fertilization with recycled fertilizers significantly influenced the sum of shoot DM and P content of the test plants (Fig. 1). Fertilization with $\text{Ca}(\text{H}_2\text{PO}_4)_2$ led to highest shoot DM and P content throughout all test plant species, closely followed by Struvite SSL. In contrast, in the Mg-SSA treatment, shoot DM was as low as in the unfertilized control and shoot P content reached an even lower level, comparable to PR.

There were huge differences in shoot DM and P content of maize between Experiments 1 and 2. Despite equal experi-

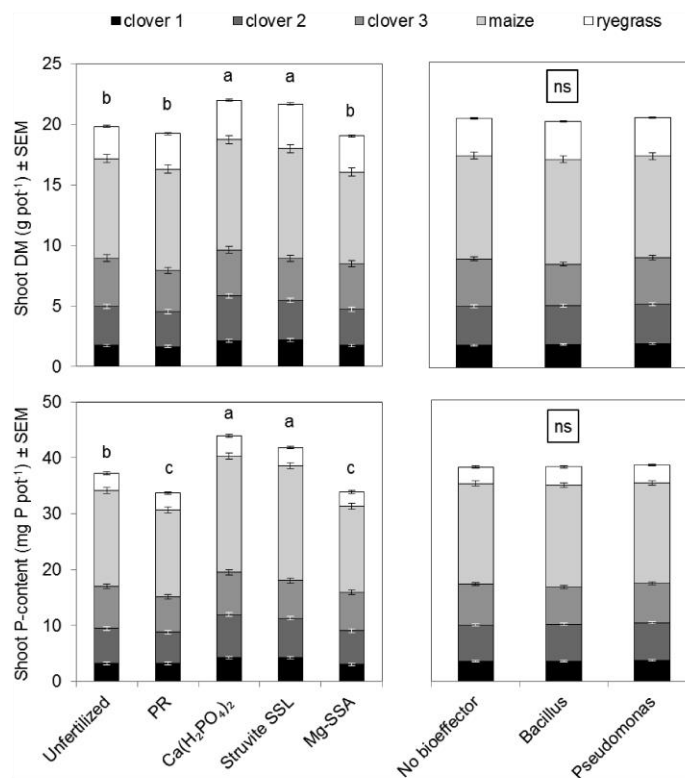


Figure 1: Shoot DM (g pot^{-1}) and shoot P content (mg P pot^{-1}) of clover at harvest dates 1, 2, and 3, maize and ryegrass, grown in Experiment 1, fertilized with recycled P fertilizers and control fertilizers (PR = phosphate rock, Struvite SSL = struvite obtained from Stuttgart Sewage Leaching Process, Mg-SSA = Mg-treated sewage sludge ash), and treated with different bioeffector products. Data are means of 15 replicates (fertilizer treatments) and 25 replicates (bioeffector treatments), SEM = standard error of the mean. Different letters indicate significant differences between treatments for the sum of all harvested plants, ns = not significant (Tukey, $P \leq 5\%$).

mental setup, distinctly higher values of maize biomass and P content were measured in Experiment 1 compared to Experiment 2. Shoot DM of maize ranged from 7.57 g pot^{-1} (Mg-SSA) to 9.11 g pot^{-1} [$\text{Ca}(\text{H}_2\text{PO}_4)_2$] in Experiment 1 (Fig. 1) and 1.48 g pot^{-1} (Mg-SSA)– 3.88 g pot^{-1} (Struvite SSL) in Experiment 2 (Fig. 2). Similarly, shoot P content ranged from $15.4 \text{ mg P pot}^{-1}$ (Mg-SSA)– $20.8 \text{ mg P pot}^{-1}$ [$\text{Ca}(\text{H}_2\text{PO}_4)_2$] in Experiment 1 and only $1.04 \text{ mg P pot}^{-1}$ (unfertilized)– $6.69 \text{ mg P pot}^{-1}$ (Struvite SSL) in Experiment 2 (Figs. 1 and 2).

Fertilization with recycled P fertilizers significantly influenced shoot DM and P content in Experiment 2 (Fig. 2). Shoot DM and P content of the Struvite SSL treatment was on the same level as the $\text{Ca}(\text{H}_2\text{PO}_4)_2$ treatment. Intermediate P content was achieved by fertilization with Struvite AirPrex®, P-RoC®, and Mephrec®. Shoot DM of the treatments pyrolysis coal and Mg-SSA did not differ from the unfertilized control. The shoot P content of plants supplied with pyrolysis coal and Mg-SSA was the lowest out of all recycled fertilizers, and plants supplied with PR did not differ from the unfertilized control.

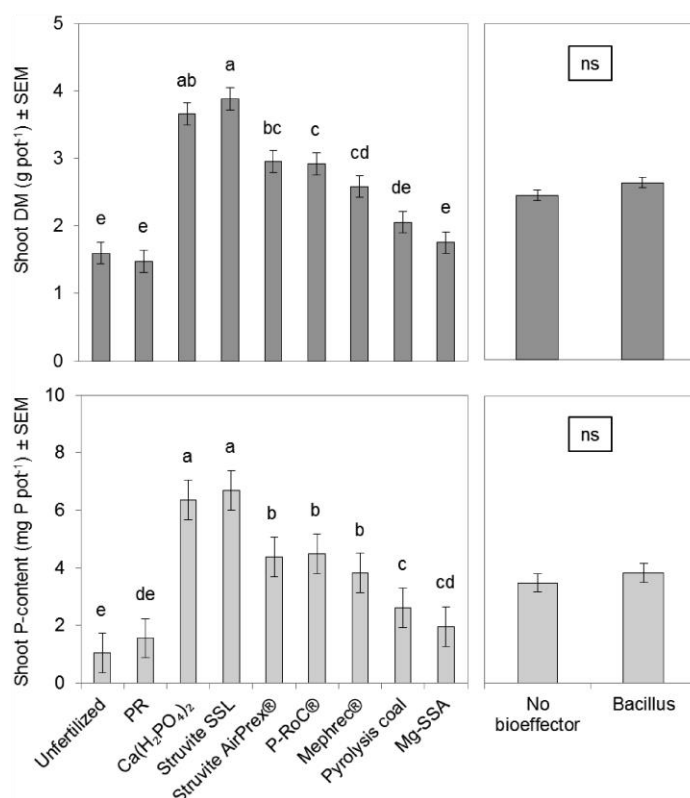


Figure 2: Shoot DM (g pot⁻¹) and shoot P content (mg P pot⁻¹) of maize grown in Experiment 2, fertilized with recycled P fertilizers and control fertilizers (PR = phosphate rock, Struvite SSL = struvite obtained from Stuttgart Sewage Leaching Process, Struvite AirPrex® = struvite obtained from AirPrex® process, P-RoC® = Ca-Phosphates recovered in the P-RoC® process, Mephrec® = Phosphorus recovered in a melt-gassing process (MEPHREC), Pyrolysis coal = Phosphorus recovered through pyrolytic treatment, Mg-SSA = Mg-treated sewage sludge ash) and treated with and without the bacteria strain *Bacillus*. Data are means of eight replicates (fertilizer treatments) and 36 replicates (bioeffector treatments), SEM = pooled standard error of the mean. Different letters indicate significant differences between fertilizer treatments in P content and shoot DM, ns = not significant (Tukey, $P \leq 5\%$).

by a higher mineral purity of Struvite SSL, due to a different manufacturing procedure and source material. Struvite purity has been reported to be affected by an interaction of several production factors (Darwish et al., 2015). For example, the formation of undesired Ca-P beside struvite is governed by the initial concentrations of P, N, and Mg, the Ca : Mg molar ratio in waste water, and the pH in the precipitation tank. However, optimum conditions (pH, reaction time, temperature) for pure struvite precipitation vary in dependency of waste water characteristics and the added Mg source, thus cannot be generally defined (Darwish et al., 2015). Kern et al. (2008) describe that Ca-P precipitation might occur besides struvite precipitation in the Airprex® process. Furthermore, an incomplete dissolution of struvite in soil solution has been reported, suggesting the presence of other P compounds of lower solubility besides struvites (Cabeza et al., 2013). Water-insoluble, amorphous Ca-P has low plant P availability (Johnston and Richards, 2003; Bauer et al., 2007; Römer, 2013). Nevertheless, it can be concluded that struvite is a promising recycled P fertilizer under neutral soil pH. However, attention should be paid to the specific production conditions in order to obtain a product with high struvite purity. Furthermore, it becomes clear that water solubility alone is not an adequate indicator for bioavailability of recycled fertilizers.

P-RoC® precipitates are less effective as P fertilizer than struvite. P-RoC® consists of di-hydrogen phosphates, hydrogen phosphates, orthophosphates, and traces of struvite, whose composition determines its solubility (Berg et al., 2007). A decreased efficacy of P-RoC® under alkaline than under acidic soil conditions has been reported by Cabeza et al. (2011) and Johnston and Richards (2003). Whereas struvite seems suitable to be used both on neutral and acidic soils (Möller et al., 2017), P-RoC® fertilization might be more promising when applied to (moderately) acidic soils.

4 Discussion

4.1 Phosphorus availability of recycled P fertilizers

Fertilization with recycled P fertilizers affected shoot P content of the test plants. High efficacy of Struvite SSL (Figs. 1 and 2) is consistent with findings by Johnston and Richards (2003), Cabeza et al. (2011), Gell et al. (2011), Liu et al. (2011), Vogel et al. (2015), Möller et al. (2017), and contradicts the results of Ackerman et al. (2013) who found struvite less effective than mono-ammonium phosphate. Throughout the experiment, Struvite SSL showed a similar P release pattern as Ca(H₂PO₄)₂ thus performing as a rapid P-release fertilizer.

Bioavailability of Struvite AirPrex® was significantly lower compared to Struvite SSL (Fig. 2). This was possibly caused

The thermally recycled P fertilizers Mg-SSA, Pyrolysis coal and Mephrec® were of low efficacy. Among them, Mephrec® showed the highest plant P availability (Fig. 2), despite of its low solubility in water and citric acid (Tab. 1). During the Mephrec® process, phosphate forms present in sewage sludge are transformed into silico-carnotites which are characterized by a moderate to high plant availability (Möller et al., 2017), possibly due to their structural instability (Gomes et al., 2011).

The apparent low P availability of Pyrolysis coal confirms findings of Schuh et al. (2013), who also tested Pyrolysis coal produced under the addition of CaCl₂. In contrast, when tested at soil pH 6.1, Mindermann et al. (2014) found a high fertilization efficacy of Pyrolysis coal produced at 500°C from sewage sludge that underwent biological or chemical P removal prior to carbonization. Due to lacking information on

the specific production conditions of the Pyrolysis coal tested in our experiments, the reason for its low P availability is not known. It can be assumed that the addition of CaCl_2 prior to the carbonization process does not increase bioavailability of the produced coal (Schuh et al., 2013), as has been reported for ashes (Adam et al., 2009). Solubility of P in thermally recycled fertilizers from sewage sludge seems to be strongly dependent on their production process (Schimmelpfennig and Glaser, 2012), e.g., temperature (Friedrich et al., 2015), retention time in the reactor and by the precipitation agent used in sewage sludge prior to thermal treatment (Ottosen et al., 2013; Mindermann et al., 2014).

Mg-SSA was completely ineffective as P fertilizer (Figs. 1 and 2). In Experiment 1, fertilization with Mg-SSA and PR even reduced plant P offtake. This might be caused by an increase of soil pH through fertilization with the alkaline Mg-SSA and PR (Sinclair et al., 1993; Nanzer, 2013) or by providing Ca ions into the soil solution. Low effectiveness of SSA (Ottosen et al., 2013) on both acidic and neutral soils (Cabeza et al., 2011) contradicts findings of Nanzer (2013) and Nanzer et al. (2014) who reported a relative efficiency of up to 88% compared to a water-soluble P fertilizer (100%) under acidic soil conditions. Phosphorus availability of ashes is mainly determined by their form of chemical bonding and degree of crystallization (Nanzer et al., 2014). Thus, it can be assumed that Mg-SSA, Pyrolysis coal and Mephrec® contain different amounts of sparingly available P-forms (e.g., brushite, stanfieldite, hydroxyl-apatite-like compounds) (Nanzer, 2013), which significantly determine their plant P availability in soil. Hypothesis 1 can be confirmed for all tested recycled fertilizers except Mg-SSA. However, a use of thermally recovered P fertilizers to cover P supply of crops may only make sense when being combined with efficient P mobilization strategies.

Hypothesis 2 has to be rejected for all tested recycled fertilizers. Sequential fractionation of fertilizers was not suitable for providing a reliable estimate of their P fertilizer value, thus, contradicting findings of Brod et al. (2015). This highlights the importance of finding new methods for describing P availability of fertilizers that are obtained from different P recycling processes and adapted to different soils. The relationship between shoot P content of plants and Hedley fractions was analyzed in a simple linear regression (data not shown) that produced weak correlations only ($R^2 \leq 0.22$). Limitations of sequential fractionation methods are widely acknowledged (Negassa and Leinweber, 2009; Negassa et al., 2010; Condrón and Newman, 2011). For a comprehensive P speciation and interpretation of results, a combination of different methods (e.g., solution ^{31}P nuclear magnetic resonance spectroscopy) with sequential fractionations has been suggested (Negassa et al., 2010).

It is well known that struvites have a high P fertilization value while showing low solubility in water (Johnston and Richards, 2003; Cabeza et al., 2011; Gell et al., 2011; Möller et al., 2017). This confirms the conclusion that P availability of recycled fertilizers in soil is not necessarily linked to their P solubility in water. There was no relation between shoot P content of test plants and P solubility of recycled fertilizers in citric acid either (Figs. 1 and 2, Tab. 1). This may be different

when fertilizers are tested under acidic soil conditions (Brod et al., 2015).

Results from pot Experiment 1 indicate that clover cultivation led to a mobilization of P for its own use and the subsequently grown maize. Differences in shoot DM and P content of clover between treatments, pronounced at harvest dates 1 and 2, were leveled out and not anymore significant on the third harvest date. This indicates that clover was able to mobilize considerable amounts of P, thus, confirming hypothesis 3. However, a quantification of this effect is not possible from this dataset and the underlying mechanisms cannot be identified. Possible suggestions include chemical P mobilization via root exudation of carboxylates (Gerke et al., 2000; Hinsinger, 2001), a recycling of mobilized P via plant residues (Horst et al., 2001; Damon et al., 2014), enhanced root colonization by PGPR (De Freitas et al., 1997; Schilling et al., 1998) and a promoted dissolution of adsorbed soil mineral P through organic acids released during decomposition of plant residues (Sharpley and Smith, 1989). The identical setup of both experiments suggests that the differences can be attributed to the suggested P mobilization potential of legumes, which might have been measurable in the subsequent maize (Kamh et al., 1999; Vanlauwe et al., 2000; Horst et al., 2001; Pypers et al., 2007).

4.2 Effect of bioeffector application on growth and P content of plants

No influence on shoot DM and P content was measured in the two experiments due to application of bioeffectors (Figs. 1 and 2). Thus, hypothesis 4 has to be rejected. Test conditions promoting effects of bacterial applications seem very specific and might not have been optimized in the conducted experiments. Further investigations on bacterial root colonization would be of importance to ensure establishment and presence of different bacteria strains. Application strategy (Alavo et al., 2015), a combination of different bacteria strains (Liu et al., 2014), used substrate (Fröhlich et al., 2012), P and N fertilization rate (Zabihi et al., 2011; El Hussein et al., 2012; Afzal et al., 2014), specific N form contained in the N fertilizer (Nkebiwe et al., 2016), and plant species (Wang et al., 2012) are issues that could be varied in further experiments, while keeping in mind that experiments with living bacteria face limitations due to natural variation (Tailor and Joshi, 2014).

5 Conclusions

Some of the tested P fertilizers recycled from sewage sludge are efficient alternative P sources that are able to improve the P supply of plants and preserve fertility and productivity of soils in organic farming systems. Distinct differences in P bioavailability among products from different production processes lead to the conclusion that their P fertilizer efficacy is strongly determined by the specific production conditions [e.g., type of P precipitation in the sludge (struvites, pyrolysis coals), pH of the solution, temperature and duration of the production process and additives (thermally recovered fertilizers) which influence the kind of P bonding and thus bioavailability. New methods have to be found that better

describe plant availability of recycled P fertilizers in soil, other than their solubility in water, citric acid, or NaHCO_3 . The effectiveness of bioeffectors to increase plant P uptake seems to be too much dependent on very specific environmental conditions to be recommended for their general use in organic arable farming. Red clover might be very efficient in mobilizing P for its own use and for subsequently grown maize. Thus, recycled P fertilizers may be tested particularly under field conditions for their use in legume-based crop rotations in organic farming systems. In future, it will be crucial to quantify the extent to which legumes are able to mobilize low-reactive P forms provided by ashes for example, and to introduce them into the pool of reactive P, circulating in a farming system.

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3 Phosphorus bioavailability of sewage sludge-based recycled fertilizers in an organically managed field experiment

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Phosphorus bioavailability of sewage sludge-based recycled fertilizers in an organically managed field experiment

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Abstract

Recycled phosphorus (P) fertilizers from sewage sludge can contribute to the ongoing effort of closing the P cycle. Five recycled P fertilizers (Struvite SSL, Struvite AirPrex[®], P-RoC[®], Pyrolysis coal, and Na-SSA) were tested for their P availability in a two-year field experiment with maize. The experiment was conducted on an organic certified research station at soil pH 6.5. Other P fertilizer treatments included: phosphate rock (PR), compost, and an unfertilized control. In addition, the rhizobacteria strain *Bacillus* sp. Proradix (Proradix[®]) was applied to test its ability to increase P bioavailability. Each year, shoot DM and P offtake of maize was measured and P use efficiency of the tested fertilizers was calculated. No significant differences in shoot DM were found among fertilized treatments and the unfertilized control in both years of experiment. Fertilization with recycled fertilizers increased P offtake by between 0% (Na-SSA) and 27.5% (Struvite SSL) compared to the unfertilized control. Rhizobacteria application led to an increase in P offtake of maize from 25.9 to 38.7 kg P ha⁻¹ when combined with PR fertilization in the year of fertilizer application, while no significant effect was found for the recycled fertilizers. Some of the tested recycled fertilizers from urban waste water can be considered as effective fertilizers for their use in organic agriculture.

Key words: phosphorus fertilization / phosphorus mobilization / recycled phosphorus / rhizobacteria

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1 Introduction

Over the last decades, discussions on future phosphorus (P) scarcity have led to an increasing debate by governmental institutions, researchers, stakeholders, and farmers about closing the societal P cycles. Agriculture alone accounts for the consumption of approximately 80% of P from phosphate rock (PR) for fertilizer production (Scholz et al., 2014). Phosphate rock is a finite resource. The estimated period until full depletion of global PR reserves is subject to strong disagreement and ranges from a few hundred to thousand years (Chowdhury et al., 2017). Suggestions to increase societal P use efficiency are an increased recycling rate from urban areas back to arable land, optimized land use, improved fertilizer application techniques, P-reduced livestock diets, breeding towards plants with higher P use efficiency, and application of rhizobacteria to improve P availability and accessibility by plants (Rodríguez and Fraga, 1999; Withers et al., 2014).

Commercially available bioeffector products, which contain spores of living microorganisms, have been developed as inoculants to improve P supply of plants. Mineralization and solubilization of orthophosphate from organic and inorganic soil P is promoted by organic acids and phosphatases that are synthesized by various soil bacteria (Lavakush et al.,

2013). In addition, a stimulation of root growth leads to an expansion of the root system and thereby an increased volume of accessible P pools in soil (Jakobsen et al., 2005). It is widely agreed that *Pseudomonas* strains are among the most effective rhizobacteria to stimulate plant growth (e.g., Santoyo et al., 2012; Miransari, 2014). *Pseudomonas* sp. are capable of rapid growth and show good rhizosphere colonization due to their ability to use various substrates as nutrients (Santoyo et al., 2012), and are promising in terms of P solubilizing (Rodríguez and Fraga, 1999). Hameeda et al. (2008), Collavino et al. (2010), and Kaur and Reddy (2014) particularly describe a P solubilization ability of bacteria strains under supply of insoluble P sources or without additional P fertilization. This leads to the assumption that the P-solubilization ability of bacteria might be enhanced under low P supply or low P availability. Thus, a specific utilization of P solubilizing rhizobacteria could be a promising approach to enhance P availability of less labile soil P forms (Kaur and Reddy, 2014), especially in organically managed farming systems, where highly soluble P fertilizers are prohibited.

However, true long-term sustainability can be achieved only through a complete recycling of P from waste streams of



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urban areas back to agriculture (Möller et al., 2018). Municipal waste water is the largest non-agricultural potential source from which to recover P (van Dijk et al., 2016), but direct sludge applications are forbidden in organic agriculture in the European Union. In Germany, direct application of sewage sludge shall be replaced by a P recycling until year 2025. The broad range of different (chemical and thermal) P recycling technologies has been described by Egle et al. (2015). Fertilizer efficacy of recycled P fertilizers from urban wastewaters has been tested in many pot experiments (e.g., Cabeza et al., 2011; Wollmann et al., 2018; Möller et al., 2018), but there is a lack of data testing recycled P fertilizers for their bioavailability under field conditions.

Strong P imbalances, negative P budgets, low soil P status, and a long-term subtle depletion of soil P (Nesme et al., 2012; Cooper et al., 2018) have been reported for different organic farming systems. Phosphorus supply of crops poses a major challenge due to the limitation of permitted P fertilizers and the ineffectiveness of PR and meat and bone meal fertilization in soils with pH > 6.0 (Guppy and McLaughlin, 2009; Nesme et al., 2012; Möller et al., 2018). Hence, there is a need for plant-available P fertilizers for organic crop production. The use of recycled P fertilizers from urban areas in organic agriculture perfectly fits with basic organic principles of a closed nutrient cycle and the use of renewable resources (Seufert et al., 2017).

The aim of the present experiment was to assess the P fertilizer efficacy of recycled P fertilizers under field conditions. We hypothesized that (1) the P availability of recycled fertilizers is higher compared to PR and urban organic waste compost and (2) the application of the bioeffector product Pro-radix® leads to an increased plant P uptake compared to treatments without bioeffector applications.

2 Material and methods

2.1 Investigated recycled P fertilizers

The tested P fertilizers were obtained from chemical (Struvite SSL, Struvite AirPrex®, P-RoC®) and thermal processes of P recovery [Pyrolysis coal, sewage sludge ash (Na-SSA)]. Struvite SSL was produced from the liquid fraction of sewage sludge, after a chemical re-dissolution of P in a pilot plant of the Stuttgart Sludge Leaching Process, provided by the Department of Wastewater Management (ISWA, Stuttgart University). Struvite AirPrex® is a commercially available struvite based on the AirPrex® process (Berliner Wasserbetriebe, Berlin). In the P-RoC® process developed by Karlsruhe Institute of Technology (KIT, Karlsruhe, Germany), calcium silicate hydrate pellets are added to the process water which release hydroxide ions and increase the pH to 9–10. Phosphorus is then recovered by crystallization and struvite as well as non-water soluble calcium phosphates (including hydroxyapatite or brushite crystals) are formed (Berg et al., 2005). Pyrolysis coal was produced in an oxygen-free atmosphere at 650°C from sewage sludge by Pyreg GmbH (Dörth, Germany) and post-treated with 96% sulfuric acid (2 kg H₂SO₄ kg⁻¹ coal) in order to increase its P bioavailability. Sodium-treated sewage

sludge ash (Na-SSA) was produced, aiming to gain a fertilizer similar to Rhenania phosphates (Möller et al., 2018) in the ASH DEC® process. The tested compost (pH 8.6; C:N ratio 12; organic matter: 368 g kg⁻¹; dry matter: 75.6%; granule size: 0–12 mm) derived from organic household waste residues separated from municipal solid waste (90%) and green waste from garden and landscaping (composting plant Kirchheim unter Teck GmbH, Germany). The total P concentration in recycled fertilizers was (mg P kg⁻¹ DM): 233 (Struvite SSL), 223 (Struvite AirPrex®), 76.3 (Na-SSA), 59.5 (Pyrolysis coal), 49.8 (P-RoC®), and 2.40 (Compost).

Control treatments were unfertilized and fertilized with PR. The used PR was a commercially available fertilizer (Naturphosphat P26, Timac Agro, Austria) consisting of soft ground rock phosphate which was ground (< 0.06 mm) and then granulated (4–10 mm). The granule size of the remaining fertilizers ranged between: 0.09 mm (Na-SSA), < 0.125 mm (Struvite SSL), < 0.5 mm (P-RoC®), 0.5–1.5 mm (Pyrolysis coal), 1–2 mm (Struvite AirPrex®) and 0.1–12.0 mm (Compost). In this study, triple superphosphate was explicitly not included as reference fertilizer since it is not allowed in organic farming systems, thus not representing an alternative fertilization to be compared to.

2.2 Experimental setup

A two-year static field experiment was conducted at the organic certified research station Kleinhohenheim of the University of Hohenheim, Germany. The research station is located in Stuttgart, 435 m asl, with a mean annual precipitation of 700 mm and mean annual temperature of 8.8°C. The soil of the trial field was a silty loam soil with pH 6.5 (CaCl₂) and (mg kg⁻¹ DM) 837 total P (aqua regia extraction), 29.9 P_{CAL} (calcium-acetate-lactate extractable P; Schüller, 1969), and (% DM) 1.06 carbonates, 31.2 clay, 55.3 silt, and 13.5 sand. According to the official German fertilizer recommendations, the P status of the soil can be classified in the second lowest category “B” (15–30 mg P_{CAL} kg⁻¹ soil), indicating a need for fertilization (Wiesler et al., 2018).

The experiment was performed from May 2014 to August 2015 with two cultivation cycles of maize (*Zea mays* L. cv. Ronaldinio; KWS Saat) in a randomized block design including four replicates per treatment. Previous crop in 2013 was a one-year grass-clover ley. Plot size was 3 m × 10 m, consisting of four maize rows each. Recycled P fertilizers were applied manually on each plot prior to sowing in May 2014 at a rate of 80 kg P ha⁻¹. Fertilizers were incorporated to a depth of 10 cm using a rotary harrow, and maize was sown at 6 cm soil depth. After emergence, horn meal was applied (70 kg N ha⁻¹) and incorporated into the soil by harrowing. The amount of N applied with the two struvites (Struvite SSL: 9.58 mg N kg⁻¹ DM, Struvite AirPrex®: 9.42 mg N kg⁻¹ DM) was considered in the N fertilization rate. Compost N (2.40 mg N kg⁻¹ DM) was considered 15% plant available (Möller and Schultheiss, 2014). In October 2014, above-ground maize biomass was harvested using a row chopper. Per plot, 6 m of the two central maize rows were collected for data acquisition. In May 2015, soil tillage was conducted to a depth of 10 cm on the trial field using a rotary harrow. Maize

was sown at 6 cm soil depth. No additional P was applied in the second year of the experiment. Nitrogen was applied as horn meal (150 kg N ha⁻¹) as in the previous year. The lower N fertilization rate in 2014 compared to 2015 was a consequence of the supposed residual N supply of the previously grown grass–clover. In August 2015, maize plants were harvested as described before. After each harvest, fresh matter was determined per plot and dry matter (DM) was measured after drying a subsample of plant material at 60°C until constant weight.

2.3 Assessment of the effects of applied bioeffectors on plant P availability

The bacterial strain *Pseudomonas* SSL. “Proradix” (Proradix®, Sourcon Padena, Tuebingen, Germany), containing 6.60E+10 colony forming units (cfu) per g, was applied to four fertilizer treatments (Na-SSA, Compost, PR, and the unfertilized control) to test its ability to increase P bioavailability. A band application was conducted twice in each growth cycle of maize after germination and 2 weeks later. To reach the target concentration of 1.98E+10 cfu per m² per application, 9.2 g Proradix® bacteria powder were dissolved in 20 L of water and applied as 5 L into each of the four maize rows of one plot, using watering cans.

2.4 Phosphorus concentration in plant tissue

Dried above-ground plant material of maize was ground using a laboratory disk mill (TS 250, Siebtechnik GmbH, Mülheim an der Ruhr, Germany). Then, an amount of 0.5 g of plant material was weighed into microwave tubes and digested with 5 mL concentrated (65%) HNO₃ and 4 mL H₂O₂ at 210°C for 62 min (VDLUF, 2011). The P concentration was measured colorimetrically (Gericke and Kurmies, 1952). Phosphorus offtake was calculated from shoot DM and P concentration.

2.5 Calculations and statistical analysis

Apparent phosphorus use efficiency (PUE) was calculated for both years of the experiment according to the following equation:

$$PUE (\%) = \left(\frac{\text{Shoot P offtake (fertilizer)} - \text{shoot P offtake (unfertilized)}}{P \text{ applied with fertilizer}} \right) \times 100, \quad (1)$$

where *P applied with fertilizer* was 80 (kg P ha⁻¹) in the first year, and “80 – total fertilizer P offtake of first year” in the second year of the experiment.

The field trial was arranged in a completely randomized block design including twelve treatments with four replicates. Two-way analysis of variance (ANOVA) was performed (SAS 9.4) to study the effect of different P fertilizers and bioeffector application and their interaction on DM and P offtake of

maize. After finding significant differences, multiple t-test analysis was performed. One-way ANOVA was performed to study the PUE of different fertilizer treatments ($P \leq 5\%$).

3 Results

3.1 Effect of recycled fertilizers on biomass production and shoot P offtake

Shoot biomass production of maize (t DM ha⁻¹) ranged between 15.6 (Compost) and 18.7 (Struvite SSL) in the first year, and 9.9 (PR) and 11.1 (Compost and Struvite SSL) in the second year of the experiment (Tab. 1). Differences in shoot DM among fertilizer treatments were not significant in both years. Shoot P offtake of maize was significantly influenced by applied recycled P fertilizers in the year of fertilizer application, ranging between (kg P ha⁻¹) 25.9 (PR) and 44.5 (Struvite SSL; Tab. 1). No significant differences were found among recycled fertilizers and the unfertilized control treatment. A significant increase of shoot P offtake compared to the PR control was observed in treatments Struvite SSL, Struvite AirPrex®, Pyrolysis coal, and Compost in the first year of the experiment (Tab. 1). In the second year of the experiment, P offtake ranged between (kg P ha⁻¹) 20.2 (Struvite AirPrex®) and 22.9 (Struvite SSL), but differences were not significant (Tab. 1).

3.2 Apparent P use efficiency of recycled fertilizers

Apparent PUE of recycled fertilizers ranged between 0.36% (Na-SSA) and 11.9% (Struvite SSL) in the first year, and between –0.73% (Struvite AirPrex®) and 2.97% (Struvite SSL) in the second year of experiment (Tab. 2). Negative PUE was found for PR in both years of the experiment, and for Struvite AirPrex® in the second year of the experiment. Highest apparent PUE was found for Struvite SSL in both years of the experiment. However, differences among recycled fertilizers were not significant (Tab. 2).

3.3 Effects of applied bioeffectors on plant P availability

Shoot DM of maize was not influenced by bioeffector application in both years of experiment (data not shown). Shoot P offtake in the PR treatment was significantly increased by application of the bioeffector product Proradix® from (kg P ha⁻¹) 25.9 (no bioeffector) to 38.7 (Proradix® application) in the first year of experiment (Fig. 1). This effect could not be repeated in the second year of experiment, when no additional P was applied (data not shown).

4 Discussion

4.1 Phosphorus bioavailability of recycled P fertilizers

Results indicate that there has been some P supply of applied fertilizers to the maize plants (Tabs. 1 and 2). Among the tested recycled fertilizers, struvites showed the highest P ferti-

Table 1: Shoot DM (t ha^{-1}) and shoot P offtake (kg P ha^{-1}) of maize grown in the first and second year of the field experiment after fertilization with different recycled P fertilizers and phosphate rock (PR = Phosphate Rock; Struvite SSL = struvite obtained from Stuttgart Sludge Leaching Process; Struvite AirPrex[®] = Struvite obtained in the AirPrex[®] process; P-RoC[®] = phosphate compounds recovered in the P-RoC[®] process; Pyrolysis coal = P obtained from pyrolytic treatment of sewage sludge; Na-SSA = Na-treated sewage sludge ash; Compost = organic household waste compost). Data are means of four replicates. SEM = pooled standard error of the mean, LSD = least significant difference with $P < 5\%$. Different letters indicate significant differences between treatments, ns = not significant.

	P fertilizer	Year 1		Year 2	
Shoot DM (t ha^{-1})	Unfertilized	15.8	ns	10.1	ns
	PR	16.3		9.9	
	Struvite SSL	18.7		11.1	
	Struvite AirPrex [®]	17.6		10.3	
	P-RoC [®]	17.8		10.9	
	Pyrolysis coal	17.7		10.1	
	Na-SSA	17.5		10.8	
	Compost	15.6		11.1	
	SEM	1.07		0.55	
	LSD (5%)	–		–	
Shoot P offtake (kg P ha^{-1})	Unfertilized	34.9	ab	20.8	ns
	PR	25.9	b	20.3	
	Struvite SSL	44.5	a	22.9	
	Struvite AirPrex [®]	41.9	a	20.2	
	P-RoC [®]	35.5	ab	22.1	
	Pyrolysis coal	40.3	a	21.9	
	Na-SSA	35.2	ab	21.9	
	Compost	40.0	a	22.3	
	SEM	3.59		1.09	
	LSD (5%)	10.6		–	

Table 2: Mean apparent phosphorus use efficiency (PUE) \pm standard error (SE) for different P fertilizers tested in the field experiment in years 1 and 2. PUE was determined relative to the amount of applied P (80 kg ha^{-1}) in year 1 and to the amount of residual P in year 2 [applied P minus apparent offtake of fertilizer P in year 1 (reference value)], ns = not significant ($P \leq 5\%$).

P fertilizer	Mean apparent PUE (%) \pm SE		Mean apparent PUE (%) \pm SE		Reference value (kg P ha^{-1})
	Year 1		Year 2		
PR	-11.33 ± 4.13	ns	-0.62 ± 0.66	ns	80.0
Struvite SSL	11.92 ± 3.32		2.97 ± 0.29		70.5
Struvite AirPrex [®]	8.75 ± 6.04		-0.73 ± 0.17		73.0
P-RoC [®]	0.69 ± 3.21		1.60 ± 1.32		79.5
Pyrolysis coal	6.74 ± 2.66		1.53 ± 2.11		74.6
Na-SSA	0.36 ± 3.33		1.40 ± 1.80		79.7
Compost	6.36 ± 8.25		2.00 ± 1.14		74.9

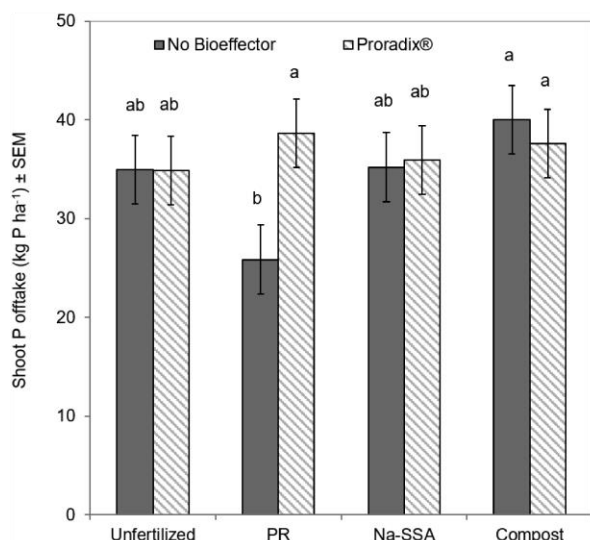


Figure 1: Shoot P uptake (kg P ha^{-1}) of maize grown in the first year of field experiment in various bioeffector treatments and treatments with recycled P fertilizers (Na-SSA = Na-treated sewage sludge ash; Compost = organic household waste compost). Data are means of four replicates, SEM = pooled standard error of the mean. Different letters indicate significant differences among treatments ($P \leq 5\%$).

lizer efficacy, which is in line with Möller et al. (2018). The slightly reduced shoot P uptake and shoot DM in the Struvite AirPrex® treatment compared to the Struvite SSL treatment might be caused by a co-precipitation of other P forms (e.g., Ca-/Al- or Fe-phosphates) besides struvite in the AirPrex® process (Wollmann et al., 2018). In the SSL process, citric acid is added in order to chelate potentially toxic elements as well as Ca-, Fe-, and Al-ions. Le Corre et al. (2005) report a preferential reaction of Ca-phosphates (Ca-P) over struvite crystals when high concentrations of Ca ions are present in the solution. Some Ca-P have low overall plant P availability (Möller et al., 2018). Thus, a possibly higher share of non-water soluble Ca-P in Struvite AirPrex® might be the reason for its lower P availability compared to Struvite SSL. Degryse et al. (2017) reported a strong effect of particle size on the dissolution rate of struvite and thus on its fertilizer efficacy. Thus, differences in physical shape between both struvites may additionally explain the higher P availability of the very fine-grained Struvite SSL, compared to the coarser Struvite AirPrex®.

Shoot P uptake in the P-RoC® treatment indicates that no additional P had been taken up by maize from this fertilizer, compared to the unfertilized control (Tab. 1). The ineffectiveness of P-RoC® precipitates under the almost neutral test soil conditions (pH 6.5) might be caused by their high portion of hydroxyapatite like compounds (Berg et al., 2005), resulting in a low P availability of P-RoC® compared to struvite. Furthermore, there are strong indications that the P fertilizer efficacy depends on soil pH (Berg et al., 2005; Cabeza et al., 2011; Möller et al., 2018). Thus, P-RoC® is a source material for fertilizer production rather than a P-fertilizer itself. Otherwise, unlike struvites, an application of P-RoC® fertilizer is

effective only when applied to soils with $\text{pH} < 5.5$ (Cabeza et al., 2011).

A few studies have been published on the P fertilizer efficacy of rhenania phosphate-like Na-SSA, highlighting its high P availability and recommending its use in both acidic and alkaline soils (e.g., Stemmann et al., 2015; Wilken et al., 2015). These results are based on laboratory experiments, measuring P solubility in neutral ammonium citrate (Stemmann et al., 2015) or in greenhouse pot experiments (Wilken et al., 2015). These findings could not be confirmed for the Na-SSA when tested under field conditions in our study (Tab. 1). It becomes clear that findings from lab or greenhouse experiments cannot unreservedly be transferred to field conditions, where different external factors might influence the efficacy of a fertilizer. There is a lack of knowledge why the Na-SSA did not show any effect under the present field conditions. Therefore, more field experiments are needed to evaluate the fertilizer efficiency of Na-SSA and its influencing factors.

Although treated with a surplus of concentrated H_2SO_4 ($2 \text{ kg H}_2\text{SO}_4 \text{ kg}^{-1} \text{ coal}$) in order to increase P availability, fertilization with Pyrolysis coal did not enhance DM or shoot P uptake compared to the unfertilized control (Tab. 1). This is possibly caused by its rather coarse particle size. Mindermann et al. (2014) reported a high P efficiency of pyrolysis coals produced at 500°C tested at soil pH 6.1. Decreased P availability of the Pyrolysis coal in our experiment might thus be caused by the higher production temperature of 650°C (Bruun et al., 2017).

Huge amounts of compost ($27 \text{ t compost fresh matter ha}^{-1}$) were applied per plot due to its relatively low P content. Unspecific response to high compost application on plant P uptake has been reported (Eichler-Löbermann et al., 2007) possibly through high amounts of sparingly soluble P species such as apatites and octacalcium phosphates contained in composts with a relatively alkaline pH (Frossard et al., 2002).

The significantly reduced shoot P uptake in the PR treatment compared to the treatments Struvite SSL, Struvite AirPrex®, Pyrolysis coal, and Compost (Tab. 1) can possibly be explained as an indirect effect of increasing P precipitation and immobilization of soil P by an increase of soil pH caused by the alkalinity of PR (Sinclair et al., 1993) or by providing Ca ions into the soil solution. Similar results have been shown in pot experiments (Wollmann et al., 2018) for the PR and an SSA treatment, raising issues about the fertilizer efficacy of PR, being one of the few permitted P fertilizers in organic farming. The hypothesis that P availability of recycled fertilizers is higher compared to PR and urban organic waste compost can partly be accepted, showing higher P availability of the recycled fertilizers Struvite SSL, Struvite AirPrex® and Pyrolysis coal compared to the PR treatment, but not when compared with the Compost treatment. Overall non-significant differences in DM and shoot P uptake between fertilized treatments and the unfertilized control in the first year might have been promoted by a residual P supply of the leguminous pre crop (Nuruzzaman et al., 2005). Even though there was only 1 year of grass-clover before the experiment started, the crop rotation on this organically managed farm includes

legumes in 3 out of 8 years and a generally high contribution of rhizosphere activity and organic matter mineralization to P supply of plants can be assumed (Mäder et al., 2002). This assumption is supported by the mean fresh matter yields of maize [52.1 t ha⁻¹ (2014) and 38.8 t ha⁻¹ (2015)] which are well in line with average silage maize yields in the federal state of Baden-Württemberg, Germany, with 48.5 t ha⁻¹ (2014) and 39.3 t ha⁻¹ (2015; *Statistisches Bundesamt* 2015; 2016). In the second year of the field experiment, data clearly indicate no residual effect of the P fertilization carried out the year before, even though only a low fraction (up to 11.9% for the Struvite SSL treatment; Tab. 2) of the applied P had been taken up by plants in addition to the soil P in the first year (Tab. 1). Struvite granules dissolve slowly in soil when they are not directly surrounded by roots (Ahmed et al., 2015) and the low water solubility might protect struvite-P from being immobilized by adsorption on soil colloids (Möller et al., 2018). However, data from our field trial clearly demonstrate that even struvite has a poor residual fertilization effect in the subsequent year, indicating that struvite P is also undergoing the well-known processes of phosphorus aging, transferring the fertilizer P into the less reactive soil P pool (Bünemann et al., 2006). The results also indicate that the negative effect of PR on plant P uptake is mainly a short-term effect, lacking any residual effect in the second year after application which might also affect the overall P fertilization strategy. We conclude that fertilizer P should be applied very targeted to responsive crops (e.g., pulse legumes).

4.2 Effect of applied bioeffectors on plant P availability

The response to the Proradix® application was visible only in combination with PR fertilization (Fig. 1). Similarly, Kaur and Reddy (2014) described the P solubilizing potential of a *Pseudomonas* strain along with PR fertilization in a field experiment, suggesting PR as a proliferation substrate for applied bacteria. This assumption is supported from pot experiments (Li et al., 2017) and field experiments (Kaur and Reddy, 2015) measuring an increased population of P-solubilizing bacteria in soil when *Pseudomonas* inoculation was combined with PR or tri-calcium phosphate. The reported P solubilizing efficacy of bacteria when combined with organic residues (Thonar et al., 2015) could not be confirmed from the Compost treatment in our experiment. In general, the reproducibility of the plant growth-promoting effect of bioeffectors is limited (Thonar et al., 2017), emphasizing that underlying modes of action are not yet fully understood and being very dependent on environmental conditions, soil substrate, and a combination of various bacterial strains (Thonar et al., 2015). The results indicate that biologically active soils have a high intrinsic P mobilization potential and usually no additional effects can be expected from soil supplementation with organisms.

5 Conclusion

The results clearly indicate that the application of recycled P fertilizers show a fertilization effect only in the year of application and no measurable P residual effect in the subsequent

year. Therefore, P fertilizers should be applied to responsive crops in the crop rotation. Struvites are the most efficient recycled P source for application in organic farming systems. However, it can be assumed that also struvite P is underlying P aging processes in soil. The high P mobilization potential of biologic active soils makes an application of P mobilizing microorganisms usually rather ineffective.

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4 Red clover (*Trifolium pratense* L.) increases plant growth and P offtake from sewage sludge ashes for a subsequent maize

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Abstract

A recycling of Phosphorus (P) from the human food chain is mandatory to secure the future P supply for food production. However, many available recycled P fertilisers from sewage sludge do not have an adequate P bioavailability and, thus, are not suitable for their application in soils with a pH > 5.5 - 6.0, unless being combined with efficient mobilization measures. The aim of the study was to test the P mobilization ability of red clover (*Trifolium pratense* L.) from two thermally recycled P fertilisers for a subsequently grown maize. Two sewage sludge ashes (SSA) were investigated in a pot experiment at soil pH 7.5 with red clover differing in its nitrogen (N) supply (mineral N fertilisation or biological N₂ fixation (BNF)), followed by maize (*Zea mays* L.). Shoot dry matter of maize was almost doubled when N supply of previous grown clover was covered by BNF, instead of receiving mineral N fertilisation. Similarly, shoot P offtake of maize following clover with BNF was significantly increased for both recycled P fertilisers. It can be assumed that the mobilization is strongly related to the BNF, and a proton release of N₂ fixing clover roots led to the measured decrease in soil pH and thereby increased P availability of the tested fertilisers. Other possible P mobilization mechanisms that might have contributed to an improved P supply of the following maize plants are discussed.

Keywords: *sewage sludge ash, phosphorus mobilization, red clover, organic farming*

1 Introduction

Phosphorus (P) is an essential component of all living organisms. Thus, a sufficient P supply for plants is a key factor in agricultural production. Unlike nitrogen (N), P cannot be synthesized chemically, and P fertiliser production is based on mined phosphate rock (PR). Approximately 80% of P from mined PR is used for fertiliser production (Scholz et al. 2014). Future availability of the world's main source of phosphorus is uncertain (Cordell 2010). New technologies have been developed to recycle P from urban areas back to agriculture. An overview of different technologies for recovering P from municipal wastewater has been published by Egle et al. 2015. Various recycled

P fertilisers from urban waste water have been tested in pot and field experiments, showing huge differences in P bioavailability, depending on their production process and soil pH (Cabeza et al. 2011; Möller et al. 2018; Wollmann and Möller 2018; Wollmann et al. 2018). For example, struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6 \text{H}_2\text{O}$) seems to be an effective P fertiliser when applied to range of soil pH while the efficacy of untreated sewage sludge ashes (SSA) strongly depends on the soil pH (Nanzer et al. 2014; Möller et al. 2018). There are indications that P availability of recycled fertilisers depends on the degree of P-crystallization which is altered during thermal treatment of waste water, determining the P availability of thermally recycled fertilisers like chars, slags and SSA (Nanzer et al. 2014; Möller et al. 2018). In general, P availability of incinerated recycled fertilisers is low compared to readily bioavailable water soluble phosphate fertilisers (Cabeza et al. 2011; Möller et al. 2018; Wollmann et al. 2018).

There are different approaches to increase P bioavailability of fertilisers, such as a soil or seed inoculation of bioeffectors (e.g. bacteria, fungi, algae extracts) to increase mineralization and solubilization of orthophosphates (Nkebiwe et al. 2016; Meyer et al. 2017; Wollmann et al. 2018), strategic placement and banding of fertilisers in soil, and the use of P-efficient crops within the crop rotation (Richardson et al. 2009).

In organic farming systems, the use of water soluble, mineral P fertilisers is prohibited, and PR is allowed, but barely plant available on soils with $\text{pH} > 6.0$ (Nachimuthu et al. 2009; Nesme et al. 2012; Möller et al. 2018). Negative P budgets and a depletion of soil P stocks have been reported for many organically managed farms (Oehl et al. 2002; Nesme et al. 2012; Cooper et al. 2018). Nutrient management in organic farming systems relies on the cultivation of legumes that allow symbiotic nitrogen-fixing bacteria to convert atmospheric N_2 into reactive N forms (biological nitrogen fixation, BNF). It has been suggested that legumes are able to take up P from sparingly soluble soil P fractions (Hassan et al. 2012) and increase internal P cycling (Horst et al. 2001). Different legume species have been shown to be efficient in P mobilization through the root excretion of carboxylates (e.g. white lupin, field pea, faba bean) or acid phosphatases (Nuruzzaman et al. 2005; Sugihara et al. 2016). In soils with $\text{pH} > 7.0$, P availability might be enhanced through a rhizosphere acidification, resulting from a

proton release during BNF of legumes (Hauter and Steffens 1985; Yan et al. 1996; Hinsinger et al. 2003). Several studies have shown an increased P availability of PR when combined with a cultivation of leguminous crops. For example, Hauter and Steffens (1985) reported an increased P offtake of red clover under BNF, compared to clover supplied with mineral N fertilisation in shoots (+ 2.08%) and roots (+ 14.2%) in combination with a PR fertiliser. Vanlauwe et al. (2000) and Pypers et al. (2007) reported a P uptake from PR applied to maize that followed velvet bean twice as high compared to maize following a first maize crop in the rotation. There are indications that also P availability of fertilisers recycled from waste water can be enhanced through a cultivation of red clover in the crop rotation (Wollmann et al. 2018).

In the present study, the P mobilization potential of red clover was investigated for fertilisers recycled from sewage sludge in a greenhouse pot experiment. Two sewage sludge ashes (Mg-SSA and Na-SSA) and PR (Naturphosphat P26, Timac AGRO, Austria) were tested for their P availability on maize with and without previous cultivation of red clover. We hypothesized that (1) Red clover which covers its N supply by biological N₂ fixation (BNF) is able to mobilize soil P and P from the tested fertilisers PR and SSA, therefore (2) P availability of PR and SSA to maize can be enhanced through a previous cultivation of red clover with BNF in the crop rotation, and (3) the P mobilization potential of red clover can be attributed to a soil acidification by clover roots under BNF.

2 Material and methods

2.1 Recycled P fertilisers

Two thermally recycled P fertilisers from sewage sludge and PR were used as fertilisers in the experiment. The Mg-containing SSA (Mg-SSA) was produced at 950 °C based on the ASH DEC® process (Outotec) and has a low plant P availability (Cabeza et al. 2011; Wollmann et al. 2018). Na-SSA was produced by calcination of sewage sludge ash with sodium under reducing conditions in a modified ASH DEC® process. P-bearing minerals in Na-SSA have a higher reactivity than untreated ashes

(Herzel et al. 2016), resulting in a high P bioavailability (Möller et al. 2018). Despite its chemical analogy to Rhenania-P after calcination of the SSA, the fertiliser is called “Na-SSA”, following the term that is used for this fertiliser in recent literature (“Na-SSA”, (Wollmann and Möller 2018); “SSA-Na”, (Vogel et al. 2018)). Further information on specific production conditions of the tested fertilisers has been published by Wollmann et al. (2018) and Wollmann and Möller (2018). The total P concentration ($\text{mg P g}^{-1} \text{ DM}$) of ashes was 76.3 (Na-SSA) and 57.2 (Mg-SSA). Control treatments were monocalciumphosphate ($\text{Ca}(\text{H}_2\text{PO}_4)_2$, MCP), PR and unfertilised.

2.2 Experimental setup

A pot experiment was conducted in the greenhouse to test the P mobilization potential of red clover for subsequently grown maize. The used substrate was a 1:1 mixture of a silty sandy loam soil and silica sand. Soil chemical and physical properties included: pH: 7.5 (CaCl_2 ; VDLUFA, 1991), total P: $554 \text{ mg P kg}^{-1} \text{ DM}$, P_{CAL} (P extractable in calcium acetate lactate): $22.0 \text{ mg P kg}^{-1} \text{ DM}$, carbonates: $47 \text{ mg g}^{-1} \text{ DM}$, organic carbon: $18 \text{ mg g}^{-1} \text{ DM}$, clay: $192 \text{ mg g}^{-1} \text{ DM}$, silt: $488 \text{ mg g}^{-1} \text{ DM}$, sand: $320 \text{ mg g}^{-1} \text{ DM}$. The experiment was performed in 1.8 L pots, containing 0.95 kg (105° C) dry soil and dry sand, respectively.

Pot substrate and fertilisers were mixed in April 2015, according to a target application of $30.4 \text{ mg fertiliser P pot}^{-1}$ or $32 \text{ mg fertiliser P kg}^{-1} \text{ soil}$ (without sand), comprising 12 replicates of each P fertiliser treatment. Four pots of each fertiliser treatment were kept without clover, and used later as a control treatment during maize cultivation. Five plants of red clover (*Trifolium pratense*, cv. Astur, Delley Samen und Pflanzen AG, Switzerland) were sown per pot in the remaining eight replicates of each P-fertiliser treatment, respectively. Potassium (K) and Magnesium (Mg) were provided with (per kg soil DM) $100 \text{ mg K as K}_2\text{SO}_4$ and 100 mg as MgSO_4 . Magnesium amounts contained in Mg-SSA were compensated for in the mineral Mg fertilisation. A minor initial fertilisation of N ($35 \text{ mg N per kg soil DM as Ca}(\text{NO}_3)_2$) was applied in order to promote early growth. During clover cultivation, 4 replicates of each P fertiliser treatment received mineral N fertilisation ($\text{Ca}(\text{NO}_3)_2$) each time after harvest, according to the calculated N removal with the aboveground biomass. The N offtakes with

aboveground biomass of clover was assumed to be 40 mg N g⁻¹ DM (Bergmann and Neubert 1976). However, the actual N content of the harvested plant material was not analyzed. A sufficient N supply and, thus, prevention of BNF was assumed in these pots. The N supply of the remaining 4 replicates with clover cultivation was assumed to be covered by BNF. All pots, both vacant and cultivated, were kept at 50% WHC through regular watering with deionized water. Aboveground biomass of clover was harvested three times at flowering; nine, 14 and 21 weeks after sowing, respectively. After third harvest date of clover, pot soil of each vacant and cultivated pot was wrapped and mixed thoroughly. When dissected, clover root nodules showing a reddish inside coloration indicated a functioning BNF. From pots with clover cultivation, bulk soil and rhizosphere soil was collected separately for the measurement of pH. Rhizosphere soil was obtained by collecting soil adhering to clover roots after shaking off looser particles. Soil was returned to pots with clover roots remaining inside. Then, one maize plant (*Zea mays*, cv. Colisee, KWS Saat SE, Germany) was sown in each pot. During maize cultivation, soil was fertilised with total (per kg soil DM) 230 mg N as Ca(NO₃)₂, 50 mg Mg (MgSO₄) and 235 mg K (K₂SO₄). Maize was cultivated for 12 weeks (Oct - Dec 2015). During maize cultivation, additional light was used in the greenhouse for 10 h d⁻¹ with an average light intensity of 430 μmol m⁻² s⁻¹, measured at the height of maize shoot tips. Plants were harvested at flowering stage. Aboveground plant biomass of clover and maize was dried at 60°C until constant weight and DM content was determined.

2.3 Laboratory analysis and calculations

2.3.1 Phosphorus concentration in plant tissue

Dried aboveground plant material of clover and maize was ground using a laboratory disk mill (TS 250, Siebtechnik GmbH, Mülheim an der Ruhr, Germany) and 0.5 g of plant material was extracted in concentrated HNO₃ using the chemical digestion method according to VDLUFA (2011). The P extract was measured colorimetrically (Gericke and Kurmies 1952). Phosphorus offtake was calculated from shoot DM and P concentration.

2.3.2 Soil pH

Soil pH of bulk soil and rhizosphere soil from experiment 2 after clover cultivation was measured in a 1:2.5 mixture with CaCl_2 using a digital pH-meter (Metrohm E 532, Herisau, Switzerland). Soil pH in soil from unplanted pots was not measured.

2.3.3 Calculation of P mobilized by symbiotic clover cultivation

The amount of additional fertiliser P offtake of clover through BNF was calculated as the difference of shoot P offtake of BNF clover and clover with mineral N supply.

The amount of additional fertiliser P offtake of maize following clover with BNF, compared to maize following clover with mineral N supply, was calculated according to the following formula, using various references:

$$\text{Additional fertilizer P offtake} = \frac{(P \text{ offtake symb. N (SSA)} - P \text{ offtake min. N (SSA)})}{\text{reference}} * 100$$

Where:

P offtake refers to the shoot P content of maize following clover with different N supply (symb. N = symbiotic N supply of clover; min. N = mineral N supply of clover), and “reference” refers to

- the amount of applied fertiliser P per pot ($30.4 \text{ mg P pot}^{-1}$) (% of applied P), or
- the amount of remaining fertiliser P after clover cultivation per pot (% of remaining fertiliser P after clover).

2.4 Data analysis

The experiment was arranged in a completely randomized design including 15 treatments with four replicates, respectively. Two-way analysis of variance (ANOVA) was performed to study the effect of different P fertilisation and N supply of clover, and their interaction on DM, P concentration and P offtake of clover (SigmaPlot 11.0). Two-way ANOVA was performed to study the effect of different P fertilisation and clover pre-cultivation, and their interaction on DM, P concentration and P offtake of maize, and pH of soil solution (SigmaPlot 11.0). After finding significant differences, all pairwise multiple comparison of the means was conducted using a Tukey test at the

0.05 level of significance. One-way ANOVA was performed to study the additional fertiliser P offtake of clover and maize, induced through clover cultivation with BNF.

3 Results

3.1 Biomass production of clover and maize

Both fertilisation with different P fertilisers and the N supply of clover significantly influenced shoot DM of clover and maize plants (Table 1), however, there were no interactions between both factors. Shoot DM of clover decreased from first to third harvest independently from P fertilisation and N supply. The fertilisation with Na-SSA resulted in high shoot DM of clover and maize at a similar level like the MCP fertilisation. Shoot DM of clover and maize fertilized with Mg-SSA did not differ from the unfertilised control. Shoot DM of clover was significantly decreased without mineral N supply. However, there was a significant increase in shoot DM of maize when N supply of the previously grown clover was covered by BNF (11.8 g pot⁻¹) instead of receiving mineral N fertilisation (5.94 g pot⁻¹). Highest shoot DM of maize (18.9 g pot⁻¹) was measured without any clover pre crop (Table 1).

3.2 Shoot P offtake and P concentration of clover

Shoot P offtake of clover was influenced by different P fertilisation and mineral N supply (Table 3), however, there were no interactions. At first harvest date of clover, there were huge differences in shoot P offtake which ranged between (mg P pot⁻¹) 5.61 (Unfertilised) and 11.0 (MCP). This increase was equivalent to 17.7% of total fertiliser P applied. These differences were levelled out towards the third harvest date of clover, where differences between P fertilisers were not anymore significant (Table 3). The total P offtakes of all three clover harvests ranged between 17.4 mg pot⁻¹ (Unfertilised) and 26.0 mg P pot⁻¹ (MCP). The statistically higher P offtakes in the Na-SSA and the MCP treatments corresponded to 19.7% and 28.3% of the total applied fertiliser P, respectively. For the fertilisers MCP and Na-SSA, shoot P-offtake decreased from first to third harvest date, while for PR, Mg-SSA and the unfertilised control, an initial increase of shoot P offtake was measured towards second harvest date.

At first and second harvest, mineral N supply of clover led to increased shoot P offtake compared to clover with BNF. At third harvest, this proportion was reversed (Table 3). Considering the sum of all clover harvests, different N supply paths did not affect shoot P offtake of clover. Shoot P concentration was influenced by P fertilisation only at first clover harvest and not thereafter. From second clover harvest on, and concerning the sum of all clover harvests, symbiotic N supply significantly increased shoot P concentration compared to clover with mineral N supply (Table 2).

Table 1 Mean shoot DM (g pot⁻¹) of clover, maize and the sum of clover and maize plants grown in the pot experiment in dependency of fertilisation with different P fertilisers (SSA = sewage sludge ash, treated with Mg or Na, MCP = monocalciumphosphate) and different pre-crop treatment. SEM = pooled standard error of the mean. Lower case letters indicate differences between P fertilisation, capital letters indicate differences between pre-crop treatment (Tukey, P ≤ 5%)

P fertiliser	clover 1	clover 2	clover 3	Σ clover	maize	Σ clover + maize						
Unfertilised	4.59	b	3.54	b	2.50	10.6	b	9.90	c	17.0	b	
Phosphate Rock	4.91	b	3.39	b	2.05	10.4	b	10.1	c	17.9	b	
MCP	6.26	ab	4.26	a	3.00	ns	13.9	a	16.0	a	25.9	a
Mg-SSA	5.46	ab	3.60	b	2.33		11.4	ab	11.5	bc	19.4	b
Na-SSA	6.84	a	4.27	a	2.24		13.9	a	13.7	ab	24.3	a
SEM	0.58		0.16		0.35		0.78		0.67		0.91	
Pre-crop treatment												
without previous clover	-	-	-	-	-	-	-	-	18.9	A	-	-
mineral N supply of clover	5.79	ns	4.23	A	2.48	ns	12.9	A	5.94	C	18.8	B
symbiotic N supply of clover	5.43	ns	3.40	B	2.35		11.2	B	11.8	B	23.0	A
SEM	0.37		0.10		0.22		0.49		0.52		0.57	

Table 2 Mean shoot P concentration (mg P g DM⁻¹) of clover, maize and the sum of clover and maize plants grown in the pot experiment in dependency of fertilisation with different recycled P fertilisers (SSA = sewage sludge ash, treated with Mg or Na, MCP = monocalciumphosphate) and different pre-crop treatment. SEM = pooled standard error of the mean. Lower case letters indicate differences between P fertilisation, capital letters indicate differences between pre-crop treatment (Tukey, P ≤ 5%)

P fertiliser	clover 1	clover 2	clover 3	Σ clover	maize		
Unfertilised	1.26	b	2.11	1.83	5.19	0.63	b
Phosphate Rock	1.43	a	2.07	1.99	5.49	0.67	ab
MCP	1.68	a	2.15	ns	5.95	0.62	b
Mg-SSA	1.33	a	2.10	1.96	5.39	0.60	b
Na-SSA	1.59	a	2.01	2.00	5.60	0.74	a
SEM	0.09		0.08	0.09	0.20		0.02
Pre-crop treatment							
without previous clover	-	-	-	-	-	0.56	B
mineral N supply of clover	1.46	ns	1.96	B	1.68	5.10	A
symbiotic N supply of clover	1.46		2.21	A	2.28	5.95	B
SEM	0.06		0.05	0.06	0.13		0.02

3.3 Shoot P offtake and P concentration of maize

When clover was cultivated previously, significantly higher shoot P offtake of maize was achieved when N supply of clover was covered by BNF (overall mean: 6.67 mg P pot⁻¹) instead of receiving mineral N fertilisation (overall mean: 4.73 mg P pot⁻¹) (Table 3). Furthermore, the highest overall mean P offtakes were found with the fertiliser treatments MCP (overall mean: 7.30 mg pot⁻¹) and Na-SSA (overall mean: 7.43 mg pot⁻¹), while the mean offtakes of the treatments PR and Mg-SSA (4.71 mg pot⁻¹) did not differ from the unfertilised control (Table 3).

When statistical evaluation included maize without previous clover cultivation, shoot P offtake of maize was determined by significant interactions between applied P fertilisers and previous N supply of clover (Figure 1). Shoot P offtake of maize was significantly higher in pots without previous clover cultivation in all P fertiliser treatments (a mean of 10.4 mg P pot⁻¹) and ranged between (mg P pot⁻¹) 8.32 (Unfertilised) and 13.1 (MCP) (Figure 1). The increase in the treatments MCP and Na-SSA in comparison to the unfertilised control was equivalent to 15.7% and 14.4% of the applied fertiliser P, respectively.

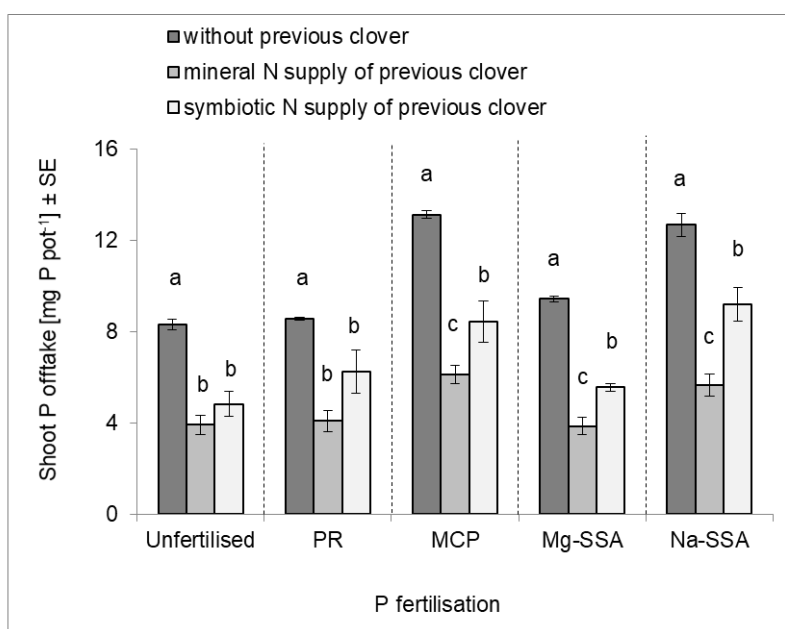


Figure 1 Shoot P offtake (mg P pot⁻¹) of maize grown in the pot experiment after different pre-crop treatments and fertilised with different recycled P fertilisers and control fertilisers (PR = Phosphate Rock, MCP = monocalciumphosphate, SSA = sewage sludge ash, treated with Mg or Na). Data are means of four replicates and error bars represent standard error of the mean. Different letters indicate significant differences between precrop treatments within the same fertiliser treatment (Tukey, $P \leq 5\%$)

Table 3 Mean shoot P uptake (mg P pot^{-1}) of clover, maize and the sum of clover and maize plants grown in the pot experiment in dependency of fertilisation with different P fertilisers (SSA = sewage sludge ash, treated with Mg or Na, MCP = monocalciumphosphate) and different pre-crop treatment. SEM = pooled standard error of the mean. Lower case letters indicate differences between P fertilisation, capital letters indicate differences between pre-crop treatment (Tukey, $P \leq 5\%$)

P fertiliser	clover 1	clover 2	clover 3	Σ clover	maize	Σ clover + maize
Unfertilised	5.61 b	7.38 bc	4.38	17.4 b	4.37 b	21.7 b
Phosphate Rock	6.83 b	6.91 c	3.98	17.7 b	4.71 b	22.9 b
MCP	11.0 a	9.04 a	5.96 ns	26.0 a	7.30 a	33.3 a
Mg-SSA	6.98 b	7.37 bc	4.44	18.8 b	4.71 b	23.5 b
Na-SSA	10.8 a	8.62 ab	4.46	23.4 a	7.43 a	30.6 a
SEM	0.57	0.32	0.54	0.86	0.42	1.00
N supply of clover						
mineral N	8.65 ns	8.29 A	3.91 B	20.8 ns	4.73 B	25.5 B
symbiotic N	7.82	7.44 B	5.38 A	20.4	6.86 A	27.3 A
SEM	0.36	0.20	0.35	0.55	0.26	

Table 4 Mean additional P uptake \pm SEM of clover and maize through symbiotic N supply of clover compared to mineral N supply of clover, and fertilisation with different P fertilisers (SSA = sewage sludge ash, treated with Mg or Na, MCP = monocalciumphosphate) and control fertilisers. ns = not significant, SEM = standard error of the mean (ANOVA, $P \leq 5\%$)

	Additional P uptake of		Additional P uptake of	
	BNF clover \pm SEM		maize \pm SEM	
	(mg P pot^{-1})		(mg P pot^{-1})	
Unfertilised	- 0.47 \pm 2.08		0.90 \pm 0.76	
Phosphate Rock	- 0.39 \pm 1.66		2.16 \pm 0.82	
MCP	- 3.22 \pm 2.02 ns		2.33 \pm 0.53 ns	
Mg-SSA	0.20 \pm 1.38		1.69 \pm 0.33	
Na-SSA	- 3.35 \pm 2.60		3.54 \pm 1.09	
mean	- 1.45 \pm 1.93		2.12 \pm 0.36	
			7.99 \pm 1.25	
			31.4 \pm 6.03	
			(% of remaining fertilizer P after clover)	
			16.9 \pm 6.45	
			43.9 \pm 10.1 ns	
			14.7 \pm 2.87	
			50.3 \pm 15.5	

With mineral N fertilisation of clover, no P fertiliser effects in the subsequent maize crop were measured for the treatments PR and Mg-SSA, compared to the unfertilised control, and only a slight increase in P offtake in the treatments MCP (7.27% of applied P) and Na-SSA (5.72% of applied P) were achieved. Shoot P offtake of maize was higher when grown after clover with BNF, compared to clover with mineral N fertilisation. These differences were significant in the treatments MCP, Mg-SSA and Na-SSA (Figure 1). The additional fertiliser P offtake of maize following clover with BNF, compared to clover with mineral N supply, ranged between 1.69 (Mg-SSA) and 3.54 mg P pot⁻¹ (Na-SSA) (Table 4), corresponding to 5.57% and 11.6% of applied P, respectively. High variation within P fertiliser treatments are indicated by high standard errors, and differences were not significant. In relation to the remaining fertiliser P after the clover cycle, the lowest additional P offtakes due to clover with BNF were achieved with fertilisers with an overall low plant P availability, Mg-SSA (14.7%) and PR (16.9%), while much higher effects were achieved with MCP (43.9%) and Na-SSA (50.3%). However, these differences were not significant.

Table 5 pH (measured in CaCl₂) of soil solution measured after clover cultivation in bulk soil and rhizosphere soil, in dependency of different N supply of clover and fertilisation with different P fertilisers (SSA = sewage sludge ash, treated with Mg or Na, MCP = monocalciumphosphate) and their interaction. Different letters indicate differences between different N supply of clover. F-values of two-way ANOVA indicate source of variation as *** (significant at $P \leq 0.001$) and ns = not significant

N supply of clover (N)	P fertilizer (P)	pH	
		bulk soil	rhizosphere soil
Mineral N application	Unfertilized	7.45	7.39
	Phosphate Rock	7.49	7.35
	MCP	7.45	7.36
	Mg-SSA	7.49	7.36
	Na-SSA	7.45	7.39
	mean	7.47	7.37
Symbiotic N fixation	Unfertilized	7.36	7.17
	Phosphate Rock	7.36	7.26
	MCP	7.30	7.16
	Mg-SSA	7.32	7.18
	Na-SSA	7.35	7.19
	mean	7.34	7.19
Source of variation (ANOVA)			
P		0.106 ns	0.478 ns
N		< 0.001 ***	< 0.001 ***
P x N		0.235 ns	0.134 ns

3.4 pH value of bulk soil and rhizosphere soil

Soil pH of bulk soil and rhizosphere soil after clover cultivation was significantly influenced by the different N supply during clover cultivation, independently from the P fertilisation (Table 5). On average, pH decreased from 7.47 (mineral N fertilisation) to 7.34 (BNF) in bulk soil, and from 7.37 (mineral N fertilisation) to 7.19 (BNF) in rhizosphere soil.

4 Discussion

4.1 Phosphorus mobilization by symbiotically grown red clover

The differences in biomass growth, P concentration and P offtakes indicate that the growth of clover and maize was limited by the plant P availability in soil (Tables 1, 2 and 3, Figure 1). Furthermore, although the P offtakes by clover accounted for at maximum 28.3% (MCP) of the applied fertiliser P, leaving at least theoretically enough P for a subsequent crop, growth of subsequent maize was strongly limited by the P availability in all treatments following a clover pre-crop. This indicates that soil and fertiliser P were not able to compensate for the P offtakes from clover aboveground biomass, even in the treatments with efficient P fertilisers (MCP, Na-SAA). The higher shoot DM and P offtake of maize without clover pre-crop (Table 1, Figure 1) presumably resulted from a higher net P supply for maize because no pre-crop had extracted P for its own use (Mat Hassan et al. 2012).

In the present experiment, the assumed P mobilization potential of symbiotically grown red clover to enhance the offtakes of soil and fertiliser P for its own use (hypothesis 1) has to be rejected, contradicting results of Hauter and Steffens (1985). Clover grown based on N provided via BNF was not able to increase its shoot P offtake compared to clover supplied with mineral N fertilisation (Table 3). However, hypothesis 2, assuming that a clover growth with BNF is able to enhance the crop growth to a subsequently grown maize, could be confirmed partially for the fertilisers MCP, Mg-SSA and Na-SSA (Figure 1). In the unfertilised control, clover with BNF was not able to enhance its own P supply (data not shown) as well as the P supply of a following

maize (Figure 1), indicating that there was no mobilization of soil P through clover cultivation with BNF, compared to mineral N fertilization.

Differences in shoot P offtake of clover due to different P fertilisers were pronounced at first harvest, and levelled out towards the third harvest of clover (Table 3). At the end of the second harvest, the fertilised treatments had taken up only a minor ratio of fertiliser P (for example, in the MCP treatment, the additional P offtake in comparison to the unfertilised control after the second harvest accounted for 23.1% of the applied fertiliser P). Nevertheless, no further P fertiliser effect was found at third clover harvest (Table 3). This indicates primarily an immobilization of fertiliser P during the clover growth cycle. The higher shoot P offtake of clover with mineral N supply compared to clover with BNF at second harvest was reversed until third harvest (Table 3), indicating an increasing P mobilization after second clover harvest by clover with BNF. This assumption seems reasonable, taken into account a certain period of time between bacterial infection of clover roots and the onset of the BNF (Marschner, 1988).

The decreased pH values in bulk soil and rhizosphere soil after clover cultivation with BNF (Table 5) support the assumption that a proton release from roots of symbiotically grown clover might have led to a dissolution of sparingly soluble P, e.g. calcium-phosphates (Hauter and Steffens 1985; Gerke 2015). Therefore, hypothesis 3 can be partially confirmed. Changes in soil pH might have influenced the plant P availability at later stages of clover and to the subsequent maize crop. Apparently, within the pH range of 7.5, a decreased pH by only 0.2 units is sufficient for this process. Other possible mechanisms that might have contributed to the P mobilization by red clover, yet were not investigated in the experiment, include a growth promotion of plant growth promoting rhizobacteria (Horst et al. 2001; Pypers et al. 2007), a release of organic acid anions and an increased phosphatase activity within the rhizosphere of BNF clover (Kamh et al. 1999, 2002; Gerke 2015).

4.2 Phosphorus mobilization by symbiotically grown red clover for subsequent maize

The higher P offtakes of maize in all P-fertilised treatments indicated that clover grown with BNF was able to enhance the availability of the applied fertiliser P to the

subsequent crop, even for the non-reactive P fertiliser Mg-SSA (Table 4). Substantial increase has been measured also in the treatment PR, however, differences were not significant due to variation. Data indicate, that the P fertiliser effects of PR and Mg-SSA were stronger to the following maize (accounting for 7.09 and 5.57% of the applied P fertiliser amounts (Table 4)) than to the clover itself (Table 3, Figure 1), at least considering the plant shoots that were investigated in the experiment. Hauter and Steffens (1985) reported a strongly increased DM, density and surface of red clover roots when grown under BNF, compared to red clover grown with mineral N fertilisation, while aboveground biomass yield was the same in both treatments. Thus, an increased exploitation of soil volume through an increased root biomass of clover with BNF might have contributed to a better utilization of available P in soil solution, which is an important factor determining P uptake by a crop (Mat Hassan et al. 2013).

Mat Hassan et al. (2012) reported a depletion of labile and less labile P pools in the rhizosphere of *Lupinus albus*. However, this P was apparently not taken up by lupin itself, but instead was transformed into labile organic P and microbial P, and led to an increased growth and P offtake of a following wheat (compared to other legume species). Similarly, huge increases in P offtake of maize have been reported when cultivated after a velvet bean (applied with PR or without P fertilisation), compared to a sole maize crop rotation (Pypers et al. 2007). Vanlauwe et al. (2000) reported that the legumes *Mucuna pruriens* and *Lablab purpureus* may have improved the availability of P from RP in excess of their own need and by this contributed to an improved P nutrition of a following maize.

A mobilization effect of P from clover to the subsequently grown maize can be concluded from significant differences in shoot P offtake of maize in the Mg-SSA treatment due to N supply of clover (Figure 1) following non-significant differences in shoot P offtake for the sum of all clover plants (Table 3). Therefore, clover grown with BNF were able to mobilize sparingly soluble fertiliser P and MCP in all fertilised treatments apart from PR. It can be assumed that the reported proton release through BNF of clover (Aguilar S. and van Diest 1981; Hauter and Steffens 1985; Liu et al. 1989) led to a mobilization of sparingly soluble P from recycled fertilisers. This

assumption is supported by the decreased soil pH after clover cultivation (Table 4), which was pronounced the most in treatments with BNF. Apparently, in the present test conditions, a minor decrease of the pH compared to the initial soil pH was sufficient to mobilize significant amounts of P even from Mg-SSA, which is characterized by poor bioavailability on soils with pH > 6.0 (Cabeza et al. 2011; Nanzer et al. 2014; Wollmann et al. 2018). This effective decrease in pH could be achieved by just 21 weeks of clover cultivation in potted soil.

Besides the mobilization of sparingly soluble P, a recycling of P from decomposed clover roots might in addition have contributed to an increased P supply of maize (Horst et al. 2001; Nuruzzaman et al. 2005; Damon et al. 2014). Hauter and Steffens (1985) found a higher biomass, density, length and surface of roots from red clover with BNF, compared to those receiving mineral N fertilisation. Thus, another factor that might have promoted P nutrition of maize after clover with BNF might be an increased amount of recycled P from a higher root biomass in these treatments. However, root biomass was not measured in our experiment, and the contribution of crop residues to P availability is likely to be significant only under conditions where large amounts of crop residues of relatively high P concentration are applied to soil (Damon et al. 2014).

Apparently, the response to the P mobilization mechanisms by clover with BNF is higher for fertiliser P compared to soil P (Figure 1). However, clover cultivation with BNF increased P offtake of the following maize by tendency, also in the treatments Unfertilised and PR. Possible suggestions are a reduced N₂ fixation caused by a deficiency of (bioavailable) P (Morton and Yarger 1990; Hellsten and Huss-Danell 2000) in the treatments PR and Unfertilised. However, 7.09% of the applied PR fertiliser could be mobilized by symbiotically grown clover (Table 4) and results are not significant due to high variation of the data. Thus, more replicates might be valuable for further experiments. Overall, the additional P removal of up to 11.6% of applied P (Na-SSA) through symbiotic clover cultivation (Table 4) is a promising result, underlining the potential of biological P mobilization which should be investigated in further experiments.

Highest shoot P offtake of maize was measured in treatments without any clover pre crop (Figure 1). This indicates that P was a growth limiting nutrient in the experiment after clover cultivation. It becomes clear, that, dependant in the P buffer capacity of the soil, red clover is not capable of providing sufficient P for optimum growth of a subsequent maize plant. These findings have been shown also for a range of other legume crops in a rotation with maize and MCP fertilisation (Kamh et al. 2002) or PR fertilisation (Pypers et al. 2007). We conclude, that red clover with BNF in the crop rotation can contribute to an enhanced P supply of a following maize plant, but external P inputs are still necessary during maize cultivation to secure optimum plant growth.

4.3 Phosphorus availability of different sewage sludge ashes

The overall high P availability of Na-SSA, compared to Mg-SSA has been described by Stemmann et al. (2015) and Möller et al. (2018). During thermochemical treatment of SSA, a sequence of chemical reactions takes place, forming new mineral phases (Adam et al. 2009), thus determining the P bioavailability of the final product. The main P-bearing component of Na-SSA is buchwaldite (CaNaPO_4) which is characterized by a high plant P bioavailability (Stemmann et al. 2015; Möller et al. 2018), as has been demonstrated also in the described experiment. On the contrary, a high fertiliser efficacy of Mg-SSA has been shown only under acidic ($\text{pH} < 6.5$) soil conditions (Nanzer et al. 2014). The dominant P-bearing phase in Mg-SSA are Ca- and Mg-phosphates, including chlorapatite, stanfieldite and farringtonite (Adam et al. 2009), whose dissolution in soil might be controlled by protons (Nanzer et al. 2014). Thus, unlike Na-SSA, Mg-SSA has been recommended to be used under acidic soil conditions only (Möller et al. 2018). It can be concluded that the mineral phases contained in SSA are crucial for their fertilizer efficacy in soil. This should be considered in the further development of the respective thermal recovery and treatment processes of sewage sludge. Similar to both tested ashes, also the fertilizer efficacy of MCP was enhanced after clover with BNF compared to clover with mineral nitrogen supply. This indicates that the well-known processes of phosphorus aging might be counteracted by the clover cultivation with BNF. The precise underlying mechanisms need to be addressed in further experiments.

5 Conclusions

The effect of red clover on the soil P turnover is dependent on the BNF activity, however clover did not take up increased amounts of mobilized P itself. The described P mobilization potential of red clover from PR for subsequently grown crops has been shown to be effective also for P fertilisers recycled from sewage sludge. Approximately 5.57% of P from the sparingly soluble Mg-SSA were mobilized by symbiotically grown clover for a subsequently grown maize. But additionally, clover grown based on BNF seems to delay the soil immobilization of readily available P fertilisers. Both effects might be dedicated to a decreased soil pH, induced by the proton release during BNF of clover. It has been shown that a marginal decrease of pH is sufficient for the mobilization of significant amounts of fertiliser P, but not soil P. Possible mechanisms to mobilize P by a decreased soil pH include a dissolution of sparingly soluble P from fertilisers and a promotion of phosphatase activity in the rhizosphere. Overall, an additional P removal by maize of up to 11.6% of applied SSA after clover cultivation with BNF emphasizes the efficacy of red clover to mobilize P also from sparingly soluble P sources including those recycled from waste water. Internal P cycling via clover roots might in addition have contributed to an improved P supply of the following maize. Thus, a P fertilisation to red clover might contribute to an increased P availability for a following maize in the rotation, however, external P inputs to maize are still necessary to secure optimum plant growth. It seems viable to introduce P fertilisers recycled from sewage sludge into organic farming systems, which often have a high share of clover in their crop rotation.

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5 Improved Phosphorus Recycling in Organic Farming: Navigating Between Constraints

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CHAPTER FOUR

Improved Phosphorus Recycling in Organic Farming: Navigating Between Constraints

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Abstract

Phosphorus (P) is an essential element for all living organisms. At the current rate of extraction, global reserves of mineable deposits will be exhausted within the next few centuries. This publication aims to summarize the current knowledge on P recycling for organic farming. The evaluation of recycled P fertilizers (RPFs) includes (i) a chemical characterization, (ii) assessment of their plant P availability and added effects in the soil, (iii) life cycle assessments, (iv) a risk assessment of their long-term impacts on soil pollution, and (v) the compilation of other environmental impacts of different treatment approaches to produce RPFs.

The highest nutrient recovery rates for P are achieved by rather simple process approaches of P recycling, while more sophisticated approaches often result in lower P recovery rates (e.g., chemical approaches for P precipitation), lower plant P availability in the final product (e.g., most thermal approaches), and losses of organic matter and nutrients like nitrogen and sulfur (e.g., thermal approaches). The plant P availability of many RPFs is higher than that of phosphate rock. Each P recycling approach has strengths and weaknesses. We conclude that any decision not to use a potential recycled P source or to introduce sophisticated treatments may have consequences compromising the ability of future generations to meet their needs. Nevertheless, we need to minimize risks for current and future generations caused by contamination linked to fertilization. Therefore, any management of nutrient recycling requires navigation between constraints. The challenge for the organic agriculture sector is to assess RPFs using a balanced approach that compromises neither the principle of ecology nor the principle of care.



1. INTRODUCTION

1.1 Phosphorus in Organic Farming

Together with nitrogen (N) and potassium (K), phosphorus (P) is one of the most limiting nutrients in organic farming ([Oberson et al., 2013](#); [Römer and Lehne, 2004](#)). Phosphorus balances for organically managed arable cropping

systems indicate that in most cases more P is removed with the products than applied as fertilizer (Berry et al., 2003; Kolbe, 2015; Watson et al., 2002) leading to reductions in plant available soil P (Kolbe, 2015; Løes and Ogaard, 1997) which threatens the long-term sustainability of organic farming systems. Moreover, P deficiencies may also feedback through the system limiting other processes which have a direct impact on yield, such as symbiotic N₂ fixation (Oberson et al., 2013; Römer and Lehne, 2004) or the release of P from crop residues (Damon et al., 2014). Currently, 80%–90% of societal recoverable P sources, referred to as recycled P fertilizers (RPFs) in this study, are not permitted in organic farming due to public or private legislation standards (Løes et al., 2016). This includes many digestates and any product made from human excreta. RPFs currently permitted include source-separated organic household waste and urban green wastes, with slaughterhouse wastes and anaerobically digested substrates allowable only under limited conditions. This strict approach hampers efficient recycling of nutrients and contributes to negative environmental impacts, such as carbon emissions from incineration of waste materials. Therefore, organic farming in its current state relies largely on phosphate rock for its P supply, either directly or by import of conventional animal manure.

1.2 Principles of Phosphorus Management and Recycling in Organic Systems

The majority of global P demand (80%–90%) is for fertilizer use in agricultural production (Smil, 2000). At the current rate of extraction, it is likely that global phosphate rock reserves will be exhausted within the next few centuries (Desmidt et al., 2015; Elser, 2012), and that the rate of production of economically available P reserves will peak between 2030 and 2040, after which demand would likely exceed supply (Schroder et al., 2011). Thus future availability of the world's main sources of P is uncertain (Cordell and White, 2011). Without the input of mined P, maintenance of food production at current global yield levels will not be possible (Cordell, 2010; Elser, 2012). The reliance on nonrenewable sources of P in food production contradicts the principle of sustainable development, i.e., “development that meets the needs of the present without compromising the ability of future generations to meet their own needs” (WCED, 1987). Furthermore, the lack of recycling of organic matter and nutrients from urban areas back to farmland contradicts some of the

basic principles of organic farming stated by the International Federation of Organic Agriculture Movements (IFOAM). The principle of ecology states that the production is to be based on ecological processes and recycling. Furthermore, nonrenewable inputs should be reduced by reuse, recycling, and efficient management of materials and energy in order to maintain and improve environmental quality and conserve resources and the climate (IFOAM, 2017). This principle is the reason for the emphasis on closed nutrient cycles in organic farming systems (Heckman, 2006; Howard, 1943; Vogt, 2007). This principle calls for a decreased dependency on phosphate rock in organic farming.

The principle of care states that organic agriculture should be managed in a precautionary and responsible manner to protect the health and well-being of current and future generations and the environment. Usually, this principle is used to argue against the utilization of organic waste and costreams in organic farming, since recycled products may contain unwanted elements and compounds such as potentially toxic elements (PTEs), persistent organic pollutants (POPs) (Brändli et al., 2007; Eriksen et al., 2009), pharmaceuticals (Eriksen et al., 2009), plastic compounds (Pinto da Costa et al., 2016), and many other compounds like personal care products (Eriksen et al., 2009). In the case of PTEs, many scientific findings on detrimental effects of sewage sludge were based on long-term experiments in England and Germany that had received substantial amounts of highly polluted biosolids from the Second World War and postwar years (López-Rayó et al., 2016). In consequence, several very relevant P recycling sources derived from human excreta are currently not permitted by the OF regulations. In addition, they may pose risks due to the presence of pathogens (Pepper et al., 2006). However, it can also be argued that the principle of care calls for recycling of societal wastes, since the current reliance on phosphate rock is clearly an unsustainable practice.

These concerns about reliance on mined P have resulted in increasing research focus and public discussion about P recycling (Schoumans et al., 2015; Schroder et al., 2011; van Dijk et al., 2016), even within the organic sector (Cornish and Oberson, 2008; Løes, 2016; Soil Association, 2010). The concerns have also supported ambitious aims, e.g., in Denmark where the organic sector has pledged to phase out conventional manure (Oelofse et al., 2013). In light of these discussions, it is an opportune time for a thorough review of the potential sources of P emerging from societal waste streams for use in organic agriculture, and for an assessment of their risks.

1.3 Aims of the Present Publication

In order to develop and evaluate sustainable strategies for increased recycling of P, a team of scientists within the European CORE Organic II-project IMPROVE-P (IMproved Phosphorus Resource efficiency in Organic agriculture Via recycling and Enhanced biological mobilization) assessed potential societal waste streams for P recycling, and the implications of current processing of these waste streams on the agronomic and environmental performance of the final RPFs. All assessments within this study were conducted on the RPFs listed in [Table 1](#) originating from the sources described below ([Section 2.1](#)). This list reflects the waste streams commonly available within Europe and includes the most promising RPFs obtained from more sophisticated approaches of P recovery from the sewer system ([Kabbe et al., 2015](#)). Water-soluble mineral P fertilizers ($\text{Ca}(\text{H}_2\text{PO}_4)_2$) and phosphate rock ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH},\text{F},\text{Cl})_2$) are included as standard P mineral sources used in conventional and organic farming, respectively. Animal manures are not defined as RPFs in this study and were only included for benchmarking purposes in some of the assessments.

The aim of the present publication was to develop criteria to evaluate the suitability of RPFs in organic agriculture. Therefore, the work comprised:

- Collection of data on P content, treatment process details, and P recovery rates for a range of relevant RPFs.
- Chemical characterization of relevant RPFs, and assessment of their plant P availability using results from pot and field experiments.
- Assessment of the environmental impacts of the production process by life cycle assessment (LCA).
- Risk assessment on the use of the RPFs by modeling the potential long-term soil accumulation of PTEs and selected POPs.
- Computation of an index of pollution potential for PTEs relative to concentrations of P, N, and K, taking into consideration the relative toxicity of the PTE in question.

The comparative evaluation of the RPFs was carried out by a multicriteria assessment (MCA), taking into account the current status of development for each RPF. This evaluation provides a general overview of the performance of the different approaches for P recycling. The objective of this paper is to provide a balanced analysis of the risks and benefits associated with increased use of RPFs in organic farming. This analysis focuses on the following specific research questions:

- (i) How does the treatment of RPFs affect their chemical composition?
Will a higher relative P fertilizer effectiveness (rP_{eff}) be attained with

Table 1 Control References and Recycled P Fertilizers Included in the Overall Assessments

Fertilizer Type	Provenance	Status Within the Project
Green waste compost	Plant residues of parks, gardens, etc.	Recycled P fertilizer
Household waste compost	Urban household waste	Recycled P fertilizer
Household waste digestate (liquid and solid)	Urban household waste	Recycled P fertilizer
Digestate from industrial organic waste processing	Catering and retailer organic wastes	Recycled P fertilizer
Meat and bone meal (dried)	Slaughter industry	Recycled P fertilizer
Meat and bone meal ashes	Slaughter industry	Recycled P fertilizer
Biosolids (dewatered)	Sewage sludge	Recycled P fertilizer
Struvite	Sewage sludge	Recycled P fertilizer
Ca-phosphates	Sewage sludge	Recycled P fertilizer
Untreated sewage sludge ashes	Sewage sludge	Recycled P fertilizer
Mg-treated sewage sludge ashes (ASH DEC [®])	Sewage sludge	Recycled P fertilizer
Rhenania ASH DEC [®] (CaNaPO ₄)	Sewage sludge	Recycled P fertilizer
Solubilized sewage sludge ashes (LeachPhos©)	Sewage sludge	Recycled P fertilizer
Slags (Mephrec)	Sewage sludge, meat and bone meal	Recycled P fertilizer
Fish sludge	Fish industry	Recycled P fertilizer
Superphosphate	Mined	Control
Phosphate rock	Mined	Control

more sophisticated treatment of feedstocks containing recoverable P? Furthermore, how is the rPeff of RPFs influenced by soil pH and by the water-soluble P fraction contained in RPFs?

- (ii) What is the risk of soil accumulation of PTEs and selected POPs from the use of RPFs? Does this risk compromise food quality, food safety, and soil fertility? Will technologies that attempt to extract P from waste streams or clean the waste streams reduce potential risks due to harmful substances?
- (iii) How does the treatment of RPFs affect other environmental impacts (e.g., greenhouse gas (GHG) emissions, energy consumption)? Will technologies that attempt to clean waste material increase the overall environmental impact?



2. SOCIETAL URBAN WASTE STREAMS FOR P RECYCLING AND P RECOVERY APPROACHES

2.1 Potential Societal P Sources

According to the available inventories on P cycles, the most important societal P sources are wastewater-based residues (sewage sludge), meat and bone meal (MBM), food wastes and food industry byproducts/wastes other than MBM and those going to animal food, and organic household wastes (Antikainen et al., 2005; Fricke and Bidlingmaier, 2003; Klinglmair et al., 2015; Oelofse et al., 2013; Ott and Rechtberger, 2012; van Dijk et al., 2016; Zoboli et al., 2016). These P sources, if sufficiently recycled, could replace approximately 50%–60% of the current P import from phosphate rock into agriculture in Europe (Klinglmair et al., 2015; Zoboli et al., 2016).

2.1.1 Wastewater-Based Residues

Sewage sludge is defined in the EU Sewage Directive 86/278/EEC (EC, 1986) as “The residual sludge from sewage plants treating domestic or urban wastewaters and from other sewage plants treating wastewaters of a composition similar to domestic and urban wastewaters.” Treated sludges are usually referred to as biosolids after hygienization and stabilization (MAFF, 2000). Phosphorus in sewage represents the main source of P that may be recycled to agriculture (Fricke and Bidlingmaier, 2003; van Dijk et al., 2016), accounting for more than 50% of the P available in off-farm P cycles (Antikainen et al., 2005; Fricke and Bidlingmaier, 2003; Ott and Rechtberger, 2012; van Dijk et al., 2016; Zoboli et al., 2016).

Around 11,000 Mg DM of biosolids are generated annually in the EU (<http://pp.eurostat.ec.europa.eu>) representing ~320,000 Mg of P. Currently, use of biosolids on agricultural land varies depending on the country. In England, Denmark, and Norway, about 70% of biosolids are used as fertilizers on conventionally managed agricultural land, and in Germany 30% of biosolids are spread on agricultural land, while in Switzerland and The Netherlands, no biosolids are applied to the land and all sewage sludge is incinerated (Dawson and Hilton, 2011).

2.1.2 Slaughterhouse Wastes

Almost 25%–45% of the live weight of slaughtered animals ends up as offal, depending on the animal species. Some of this is processed to MBM, a byproduct of the rendering industry, but a significant part is incinerated, e.g., due to lack of infrastructure for producing MBM. Total production of MBM in the EU exceeds 18 million Mg per year, including 2.5 million Mg in France, 1.5 million Mg in United Kingdom, 350,000 Mg in Austria, 500,000 Mg in Denmark, and 2.8 million Mg in Germany (Dobbelaere, 2013). The total P potential in the EU from the rendering industry offal is about 310.000 Mg P per year, whereas food wastes and food industry byproducts other than MBM have a yearly P potential of about 215.000 Mg per year (Lesschen et al., 2013; van Dijk et al., 2016).

2.1.3 Urban Organic Household Wastes

In urban areas, apart from the sewage and food industry wastes, different sources of organic material are available:

- green wastes from gardens or park areas, such as grass or flower cuttings, foliage and hedge trimmings,
- source-separated organic waste from private households,
- food waste from retail outlets, often with high proportions of plastic and often including waste defined according to Regulation (EC) No 1069/2009 (animal byproducts), and
- food waste from catering and food processing, including waste defined according to Regulation (EC) No 1069/2009.

Source-separated organic household waste accounts—based on a German assessment—for about 10%–15% of the currently recycled P in Central and North European countries (Fricke and Bidlingmaier, 2003). In many European countries, the quantity of source-separated organic household waste amounts to approximately 100 kg per person per year (Kern et al., 2010; Knappe et al., 2007; Quested et al., 2013) which is equivalent to

40% of the total municipal solid waste mass. However, currently only 30% of the organic household wastes across Europe are source separated, collected separately, and recycled as organic fertilizer.

2.1.4 Other Potential P Sources

Ashes, chars, and slags from incineration or other thermal treatment processes of a broad range of waste streams are other potential sources of RPFs. This includes products of incineration of MBM (Cascarosa et al., 2012) and sewage sludge, which is an important method of disposal in the United States, some European countries (e.g., in Germany, Switzerland, and the Netherlands), and Japan (Donatello and Cheeseman, 2013). Currently, 1.7 million Mg of monoincinerated sewage ash is produced annually worldwide (Donatello and Cheeseman, 2013), with a P content of about 66.8–93.7 g kg⁻¹ DM (Table 2). There is also considerable potential for use of ashes from incineration of household and industrial wastes, although these may have relatively low P concentrations. Finally, industrial wastes containing P are also a potential P source, although the size of this P source is much smaller than for sewage and household waste. Little is currently known about the fate of these P-containing residues (Fricke and Bidlingmaier, 2003; van Dijk et al., 2016).

2.2 P Recovery Approaches and Rates

Since P in sewage is the main potential off-farm P source, any approach to establish efficient P recycling must consider appropriate pathways for recycling of sewage P. The second most important source, slaughterhouse wastes, is currently often incinerated and landfilled due to concerns about hygienic risk (prion diseases). Any approach for improved P recycling should achieve maximum recovery rates for each of the major recoverable sources (Kabbe et al., 2015; Withers et al., 2015), combined with approaches to make more P sources accessible for recycling, e.g., incineration ashes. This should be complemented by a long-term effort to develop better sanitation and waste management systems (Magid et al., 2006).

The desire to lower the risks from the unwanted elements and compounds described above (Section 1.2) has led to the development of new approaches for decontamination, and extraction of P from waste streams (Hukari et al., 2016; Kabbe et al., 2015; Tarayre et al., 2016) (Fig. 1). For sewage sludge as an alternative to direct field application after stabilization and hygienization, P can be selectively removed by P precipitation via chemical treatment, to produce struvite or calcium phosphates,

Table 2 Mean Values (and Ranges) for Dry Matter (DM) (% Fresh Matter), Organic Matter (OM), and Macronutrient Concentrations (% DM) of Recycled Phosphorus Fertilizers (RPF) Reported in the Literature

RPF	DM	OM	N	P	K	S	Mg	Ca	Reference
Green waste compost	62.6 (52–74)	36.9 (23–51)	1.15 (0.7–1.6)	0.22 (0.14–0.32)	0.85 (0.4–1.3)	n.a.	0.44 (0.19–0.79)	n.a.	BGK (2013) cited by Möller and Schultheiss (2014)
Household waste compost	64.5 (52–78)	39.5 (26–54)	1.45 (0.9–2.0)	0.31 (0.18–0.44)	0.98 (0.6–1.4)	n.a.	0.45 (0.22–0.72)	n.a.	BGK (2013) cited by Möller and Schultheiss (2014)
Household waste compost including catering and retailer organic wastes	64.0 (52–77)	39.5 (26–52)	1.53 (0.9–2.0)	0.36 (0.18–0.49)	1.10 (0.6–1.5)	n.a.	0.51 (0.22–0.74)	n.a.	BGK (2013) cited by Möller and Schultheiss (2014)
Liquid household waste digestate	12.0 (5.0–20.5)	58.1 (42–78)	4.47 (2.3–6.9)	0.68 (0.36–1.23)	3.24 (1.48–6.57)	n.a.	0.65 (0.42–1.06)	n.a.	BGK (2013) cited by Möller and Schultheiss (2014)
Liquid digestate of catering and retailer organic wastes	3.34 (1.9–7.1)	56.5 (48–80)	16.3 (4.93–26.5)	2.21 (1.03–3.14)	4.49 (2.7–8.7)	0.86 (0.44–2.74)	0.21 (0.08–1.12)	n.a.	Möller and Schultheiss (2014)

Solid household waste digestate allowed for use in OF	45.8 (25–69)	61 (44–87)	1.84 (1.1–2.6)	0.60 (0.24–1.10)	1.20 (0.5–2.0)	n.a.	0.51 (0.35–0.75)	n.a.	BGK (2013) cited by Möller and Schultheiss (2014)
Solid household waste digestate including catering and retailer wastes	42.4 (26–84)	58.2 (38–81)	2.87 (1.81–4.86)	0.92 (0.41–1.45)	1.32 (0.35–2.32)	n.a.	0.59 (0.40–0.87)	n.a.	BGK (2013) cited by Möller and Schultheiss (2014)
Bone meal	95.3 (92.5–97.3)	40.3 (16.0–52.9)	5.25 (1.00–7.70)	11.4 (6.03–16.5)	0.34 (0.05–1.66)	0.42 (0.27–0.62)	0.48 (0.06–1.81)	23.4 (15.1–30.2)	Möller and Schultheiß (2014)
Meat meal	93.9 (92.5–95.0)	74.8 (54.4–98.9)	8.00 (5.80–15.0)	3.42 (0.30–4.74)	2.09 (0.08–6.50)	0.44 (0.33–0.50)	0.16	6.79 (0.11–9.44)	Möller and Schultheiß (2014)
Meat and bone meal	96.2 (91.2–99.3)	65.4 (47.1–91.1)	8.28 (3.00–12.0)	5.31 (2.21–9.62)	0.67 (0.14–3.95)	0.45 (0.04–1.90)	0.29 (0.14–1.02)	9.60 (5.30–19.6)	Möller and Schultheiß (2014)
Meat and bone meal ashes	97.8	n.a.	0.17	14.0 (6.07–18.9)	1.50	0.14	0.64 (0.58–0.70)	20.9 (13.5–28.2)	Pinnekamp et al. (2011), Czaja and Herrmann (2011), and Coutand et al. (2008)
Liquid biosolids	3.81 (1.71–6.51)	66.9 (42.7–78.1)	7.72 (4.70–17.2)	3.75 (2.42–5.63)	0.84 (0.30–1.68)	1.05 (0.39–2.79)	0.56 (0.28–1.14)	2.81 (1.17–5.86)	Heck (2015)
Dewatered biosolids	23.3 (17.6–40.3)	61.6 (40.4–82.8)	5.17 (3.07–10.6)	3.68 (2.21–5.55)	0.27 (0.10–1.20)	1.11 (0.17–1.80)	0.59 (0.23–1.66)	3.80 (1.22–8.19)	Heck (2015)

Continued

Table 2 Mean Values (and Ranges) for Dry Matter (DM) (% Fresh Matter), Organic Matter (OM), and Macronutrient Concentrations (% DM) of Recycled Phosphorus Fertilizers (RPF) Reported in the Literature—cont'd

RPF	DM	OM	N	P	K	S	Mg	Ca	Reference
Struvite	56.0 (55.1–56.8)	n.a.	4.75 (2.80–5.35)	11.3 (6.11–13.5)	0.16 (0.05–0.27)	0.62 (0.14–1.16)	10.6 (7.84–15.2)	1.42 (0.52–2.79)	Antakyali et al. (2006), Pinnekamp et al. (2007), Kern et al. (2008), Weideler et al. (2008), Uysal et al. (2010), Antonini et al. (2012), Hermans et al. (2012), Linderholm et al. (2012), Liu et al. (2012), Meyer and Steinmetz (2013), and IMPROVE-P (unpublished data)
Nonwater-soluble calcium phosphates (P-RoC®)	83– 21.1 (19.0–22.5)	n.a.	0.48 (0.3–0.7)	8.68 (4.1–10.9)	0.23 (0.08–1.0)	0.03–	0.69 (0.65–0.72)	19.4 (14.3–25.0)	Berg et al. (2005), Ehbrecht (2014), and Okano et al. (2016)
Untreated sewage sludge ashes	n.a.	n.a.	n.a.	7.66 (6.68–9.37)	1.10 (0.69–1.50)	1.05 (0.89–1.21)	1.33 (1.04–1.64)	11.6 (9.66–13.7)	Steinmetz et al. (2014), Appel and Friedrich (2016), and Herzel et al. (2016)

Mg- or Ca-treated SSA (ASH DEC [®])	n.a.	n.a.	n.a.	8.21 (5.72–11.2)	1.18 (0.22–2.29)	0	2.22 (0.93–5.43)	11.2 (8.94–16.2)	Nanzer et al. (2014a), Adam et al. (2009), and IMPROVE-P (unpublished data)
Rhenania ASH DEC [®] (CaNaPO ₄)	n.a.	n.a.	n.a.	7.7	1.2	2.6	1.3	9.3	Herzel et al. (2015)
Converter-Slags (Mephrec)	n.a.	n.a.	n.a.	5.01 (2.01–9.0)	0.77 (0.54–1.0)	0.12–	1.94 (1.5–2.28)	25.4 (12.0–36.1)	Scheidig (2009), Herzel et al. (2015), and IMPROVE-P (unpublished data)
Biochars	n.a.	n.a.	n.a.	5.52 (5.25–5.85)	n.a.	n.a.	n.a.	n.a.	Appel et al. (2016) and Appel and Friedrich (2016)
Triple superphosphate	n.a.	n.a.	n.a.	20–	n.a.	n.a.	5.67–	n.a.	Taylor et al. (2016)
Phosphate rock	n.a.	n.a.	n.a.	13.5 (8.6–16.7)	0.15 (0.10–0.20)	1.25 (1.20–1.30)	0.80 (0.07–6.39)	31.0 (20.1–35.2)	Chien and Hammond (1978), Mortvedt (1996), Bolland et al. (2001), Sahrawat et al. (2001), and Dissanayake and Chandrajith (2009)

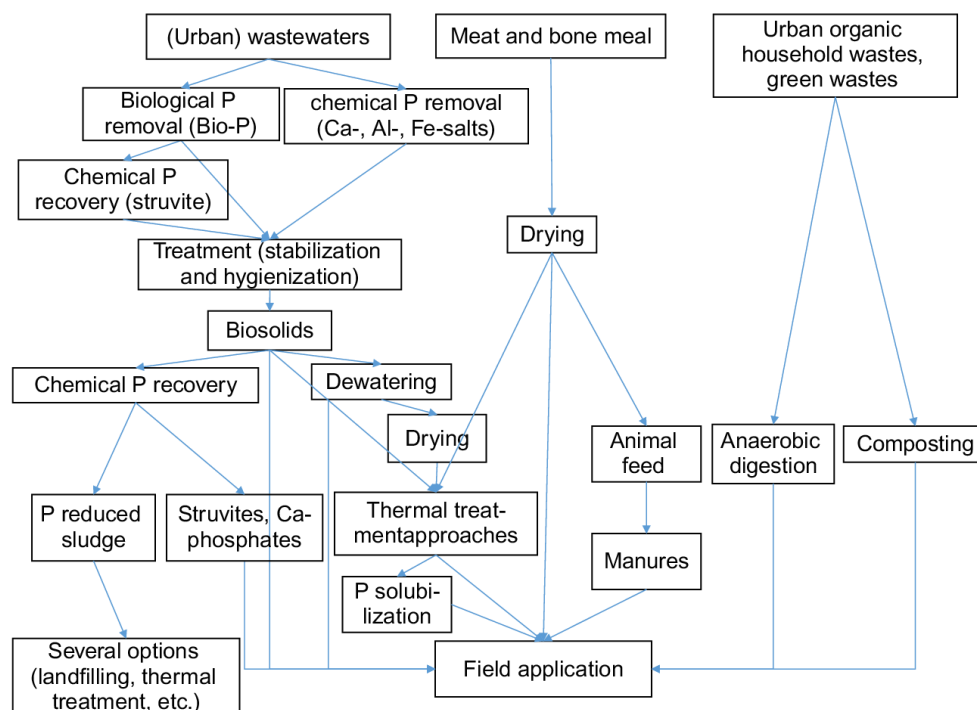


Fig. 1 Main recycled phosphorus (P) fertilizer sources and their major treatment options.

which are essentially free of contaminants (Ewert et al., 2015; Kabbe et al., 2015). The main phosphate fertilizers produced by precipitation are struvites (by struvite crystallization), water-soluble di-Ca-phosphates (e.g., Budenheim Carbonic Acid Process), or nonwater-soluble Ca-phosphates (e.g., P-RoC© process), containing a mixture of hydroxy apatite-like P (Berg et al., 2006) and other P-containing precipitates like struvite. Struvite is magnesium ammonium phosphate hexahydrate, and it consists of Mg^{2+} , NH_4^+ , and PO_4^{3-} ions in the molar ratio of 1:1:1, respectively ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$). Other crystals belonging to the struvite group are $\text{MgKPO}_4 \cdot 6\text{H}_2\text{O}$, $\text{CoNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$, $\text{CoKPO}_4 \cdot 6\text{H}_2\text{O}$, $\text{NiNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$, and $\text{NiKPO}_4 \cdot 6\text{H}_2\text{O}$ (Kern et al., 2008). Nonwater-soluble Ca-orthophosphate products treated with calcium-silicate-hydrate (CSH) particles (P-RoC© process) contain considerable amounts of silica ($280\text{--}340 \text{ g SiO}_2 \text{ kg}^{-1} \text{ DM}$).

Similarly, thermal treatment of a variety of waste materials can be used to obtain a P-rich ash free from organic pollutants (although still containing PTEs) as a base for mineral fertilizer production (Kabbe et al., 2015; Rulkens, 2007). Almost any kind of organic residue can undergo thermal treatment, e.g., via traditional combustion, or alternative methods such as hydrothermal carbonization (HTC) (Funke et al., 2013), or pyrolysis which

produces energy through gasification plus a P-containing biochar (Sohi et al., 2010). During or after incineration, the chars or ashes can be further treated to remove PTEs, and to achieve a reconfiguration of the P in the crystal structure (e.g., Herzel et al., 2016; Kabbe et al., 2015). Thermochemical treatments at temperatures $>900^{\circ}\text{C}$ will remove many PTEs (e.g., arsenic [As], cadmium [Cd], mercury [Hg], and lead [Pb]). A further thermal treatment approach is calcination, e.g., as carried out by Rhenania ASH DEC[®], where the alkali minerals (sodium or K) replace carbonate in the P crystals, leading to new P-bearing mineral phases with higher reactivity than untreated ashes (Herzel et al., 2016). Furthermore, calcination under reducing conditions leads to a significant removal of As (61% removal), Cd (80%), Hg (68%), and Pb (39%) and minor removal of zinc (Zn, 9%) (Herzel et al., 2016). Thermal treatment processes are among the most efficient methods for destroying harmful organic contaminants, such as dioxins and furans (Lindberg et al., 2015; Rulkens, 2007) and pharmaceuticals. For lower temperature pyrolysis used in the production of chars, however, there is a risk of creating polycyclic aromatic hydrocarbons (PAH) if the process is not well regulated (De la Rosa et al., 2016).

The approaches and processes for P recycling affect the fertilizer value, the recovery rates, the flows of organic matter, and other plant nutrients, with direct implications on agronomy, emissions, and the consumption of nonrenewable resources. For example, P recovery from sewage by calcination (a thermal treatment approach) demands significant inputs of energy for dewatering and drying, production and transport of chemicals, drying of precipitates and pressurization, and of chemicals such as acids and bases in order to manipulate the pH (Ewert et al., 2015; Kabbe et al., 2015).

Contrary to these advanced approaches, high P recovery rates (close to 100%) may be achieved with rather simple treatment methods like composting or anaerobic digestion of P-containing wastes, or by direct application of sanitized biosolids. Thermal approaches have also high recovery rates (e.g., about 80% for Mephrec converter slags and nearly 100% for incineration (Kabbe et al., 2015). Chemical approaches for P recovery from sewage sludge address only the water-soluble P and P attached to colloids, resulting in lower recovery rates between 5% and 75% (Braak et al., 2016; Ewert et al., 2015; Kabbe et al., 2015). Only the water-soluble P as well as the P attached to the surface of the organic matter and other colloids may be addressed by chemical approaches of P recovery. The lower the pH during treatment, the higher the dissolution rate of adsorbed P, but increases consumption

of chemicals (Antakyali et al., 2013; Ewert et al., 2015; Yilmazel and Demirer, 2013).



3. COMPOSITION OF RPFs

3.1 Chemical Composition of RPFs

The composition of RPFs varies considerably for all parameters, depending on the feedstocks used and the treatment method (Table 2). Digestates and biosolids that are not dewatered are characterized by low DM contents (2%–10%), whereas composts have intermediate DM contents (50%–75%) and ashes and dried MBM have high DM contents (>90%). The organic matter content varies as well, with the highest values found for biosolids (60%–70%), digestates (45%–85%), and MBM (50%–90%), and intermediate values for composts with approximately 40% organic matter (25%–55%). Thermal treatment of organic wastes leads to almost complete removal of organic matter and N, and increases the P concentration on a DM base.

The lowest P concentrations in the DM were reported for composts (1.5–4.5 g kg⁻¹ DM), biosolids (2.5–14.5 g kg⁻¹ DM), and liquid digestates (4.0–31.0 g kg⁻¹ DM). The mean P concentration in biosolids on a dry basis is approximately 15 times higher than in urban organic waste composts. Struvite (60–130 g kg⁻¹ DM), MBMs (60–165 g kg⁻¹ DM), nonwater-soluble Ca-phosphates obtained via P-RoC© (40–110 g kg⁻¹ DM), and ashes (40.0–189 g kg⁻¹ DM) are characterized by relatively high P concentrations. The P concentration of ash-based fertilizers depends largely on the source materials used (Adam et al., 2008; Nanzer et al., 2014a) and the combustion temperature (Christel et al., 2014).

RPFs can also contain significant quantities of other plant nutrients. High concentrations of N were reported for urban organic waste digestates (25–265 g kg⁻¹ DM), MBM (30–120 g kg⁻¹ DM), meat meal (60–150 g kg⁻¹ DM), biosolids (45–70 g kg⁻¹ DM), and struvite (28–54 g kg⁻¹ DM), while composts are characterized by low N concentrations (7–20 g kg⁻¹ DM) due to N losses during treatment and storage. The K and sulfur (S) concentrations of most RPFs are rather low. The calcium (Ca) concentration of ashes and MBM is relatively high. The magnesium (Mg) concentration in most RPFs with the exception of struvite (50–100 g Mg kg⁻¹ DM) is relatively low (Table 2). In ashes, the main elements in addition to P are silicon (Si), aluminum (Al), iron (Fe), Mg, Ca, and K.

Table 3 Mean Concentrations (and Ranges) of Potentially Toxic Element Contents (mg kg⁻¹ DM) in Recycled Phosphorus Fertilizers (RPF) Reported in the Literature

	Zn	Cu	Pb	Cd	Ni	Hg	Cr	Reference
Green waste compost	34.4 (22.3–50.0)	154 (106–213)	32.4 (17.0–50.7)	0.40 (0.19–0.70)	22.8 (12.0–35.9)	13.8 (5.70–23.5)	0.11 (0.05–0.16)	BGK (2013) cited by Möller and Schultheiss (2014)
Household waste compost	40.5 (26.8–56.6)	150 (114–184)	27.9 (18.2–38.0)	0.35 (0.20–0.51)	21.5 (13.4–30.5)	12.2 (6.29–18.9)	0.10 (0.05–0.16)	BGK (2013) cited by Möller and Schultheiss (2014)
Household waste compost including catering and retailer organic wastes	54.2 (26.8–80.9)	213 (114–280)	44.8 (18.2–67.0)	0.46 (0.20–0.70)	28.1 (13.4–42.2)	17.5 (6.29–27.8)	0.11 (0.05–0.17)	BGK (2013) cited by Möller and Schultheiss (2014)
Liquid household waste digestate	49.2 (27.6–76.8)	200 (139–312)	20.1 (3.0–33.2)	0.41 (0.20–0.62)	18.9 (5.20–29.6)	12.6 (6.94–18.0)	0.10 (0.05–0.14)	BGK (2013) cited by Möller and Schultheiss (2014)
Liquid digestate of catering and retailer organic wastes	40.0 (10.1–176)	177 (115–338)	3.85 (1.03–24.4)	0.33 (0.05–1.25)	7.56 (4.13–22.7)	9.45 (3.69–23.6)	0.07 (0.02–0.33)	Möller and Schultheiß (2014)
Solid household waste digestate	24.1 (7.50–38.3)	102 (46.0–177)	14.7 (1.88–34.0)	0.23 (0.03–0.38)	19.0 (3.40–40.2)	8.81 (2.50–16.8)	0.07 (0.01–0.18)	BGK (2013) cited by Möller and Schultheiss (2014)

Continued

Table 3 Mean Concentrations (and Ranges) of Potentially Toxic Element Contents (mg kg^{-1} DM) in Recycled Phosphorus Fertilizers (RPF) Reported in the Literature—cont'd

	Zn	Cu	Pb	Cd	Ni	Hg	Cr	Reference
Solid household waste digestate including catering and retailer wastes	42.5 (19.2–65.0)	193 (89.2–287)	25.7 (3.00–48.8)	0.44 (0.15–0.88)	21.1 (8.28–40.4)	16.4 (4.30–31.0)	0.09 (0.01–0.14)	BGK (2013) cited by Möller and Schultheiss (2014)
Bone meal	113 (89–133)	8.48 (0.50–50.0)	3.10 (0.40–10.0)	0.24 (0.21–0.30)	8.85 (1.00–24.0)	0.04 (0.02–0.05)	13.3 (0.30–37.5)	Möller and Schultheiß (2014)
Meat meal	95.6 (47.0–140)	21.8 (0.05–29.4)	4.25	0.43	3.00 (0.77–3.10)	0.18	7.11 (5.90–8.31)	
Meat and bone meal	107 (28.0–174)	11.2 (0.19–26.5)	2.97 (0.01–36.2)	0.21 (0.003–1.74)	3.31 (0.01–37.9)	0.02 (0.01–0.03)	4.15 (1.00–9.70)	
Meat and bone meal ashes	171.6 (16.3–373.1)	21.7 (3.61–46.6)	13.1 (8.66–17.6)	0.82 (0.30–1.34)	27.9 (2.11–78.4)	<0.001	11.2 (5.28–20.8)	Pinnekamp et al. (2011), Czaja and Hermann (2011), and Coutand et al. (2008)
Liquid biosolids	848 (310–1700)	308 (84.6–679)	27.4 (8.03–99.0)	0.78 (0.14–1.32)	28.5 (15.0–72.0)	0.44 (0.12–0.91)	29.9 (15.0–64.0)	Heck (2015)
Dewatered biosolids	993 (346–1400)	360 (69–797)	38.2 (10.1–89.5)	0.84 (0.11–1.48)	30.5 (13.0–68.0)	0.54 (0.10–0.93)	37.3 (16.1–257.0)	Heck (2015)

Struvite	123 (1.0–403)	41.6 (0.18–160)	12.8 (0.01–44.0)	0.45 (0–1.76)	7.3 (0–28.6)	0.53 (0–4.23)	9.54 (0–42.0)	Antakyali et al. (2006), Pinnekamp et al. (2007), Kern et al. (2008), Weideler et al. (2008), Uysal et al. (2010), Liu et al. (2012), Antonini et al. (2012), Meyer and Steinmetz (2013), Linderholm et al. (2012), Hermanussen et al. (2012), and IMPROVE-P (unpublished data)
Nonwater-soluble calcium phosphate (P-RoC®)	73.8 (5.6–142)	8.7 (2.4–15.0)	1.16 (1.1–4.0)	Detection limit	2.14 (0–7.3)	Detection limit	5.70 (0–23.0)	Ehbrecht (2014), Okano et al. (2016), and IMPROVE-P (unpublished data)
Untreated sewage sludge ashes	1483 (31.7–2479)	1017 (875–1240)	102 (58.0–141)	1.73 (0.32–3.47)	66.0 (64.0–68.0)	0.21	133 (88.2–233)	Steinmetz et al. (2014) and Appel and Friedrich (2016)
Mg- or Ca-treated sewage sludge ashes (ASH DEC®)	136 (41.1–275)	98.0 (10.5–249)	1.84 (0.10–6.70)	0.11 (0.10–0.20)	48.7 (32.1–72.4)	<0.1	91.2 (58.1–120)	Adam et al. (2009), Nanzer et al. (2014a), and Wollmann (unpublished data)
Rhenania ASH DEC® (CaNaPO ₄)	1710	601	60.1	0.3	56.4	0.3	127	Herzel et al. (2015)

Continued

Table 3 Mean Concentrations (and Ranges) of Potentially Toxic Element Contents (mg kg⁻¹ DM) in Recycled Phosphorus Fertilizers (RPF) Reported in the Literature—cont'd

	Zn	Cu	Pb	Cd	Ni	Hg	Cr	Reference
Converter slags (Mephrec)	79.8 (65.0–85.0)	85.8 (74.0–110)	16.2 (1.1–20.0)	0.058 (<0.1–0.15)	15.8 (14.1–20.0)	<0.01—	30.0 (1.0–83.0)	Scheidig et al. (2013), Herzel et al. (2015), and IMPROVE-P (unpublished data)
Biochars	1917 (530–2533)	732 (420–952)	68.5 (30.0–100)	1.07 (0.20–1.85)	64.9 (46.0–77.0)	0.23 (0.15–0.30)	96.8 (40.0–143)	Appel and Friedrich (2016), Appel et al. (2016), and Stemann et al. (unpublished)
Triple superphosphate	401 (348–489)	28.8 (17.0–42.0)	10.2 (1.6–17.0)	22.1 (14.0–26.8)	31.3 (26.6–36.3)	0.83–	211 (146–288)	Kerschberger et al. (2001), Steinmetz et al. (2014), and Taylor et al. (2016)
Phosphate rock	163 (0.01–325)	16.3 (0.02–30.5)	9.32 (1.20–35.0)	21.3 (0.1–60.0)	25.4 (2.0–41.0)	0.46 (0.01–3.41)	138 (1.0–328)	Chien and Hammond (1978), Mortvedt (1996), Bolland et al. (2001), Kerschberger et al. (2001), Sahrawat et al. (2001), and Dissanayake and Chandrajith (2009)
Threshold values for fertilizers derived from urban organic house- hold wastes (Reg. (EC) 889/2008)	70	200	45	0.7	70	25	0.4	(Reg. (EC) 889/2008)

3.2 Potentially Toxic Elements

The concentration of PTEs in the assessed RPFs varies in a broad range when expressed on a DM base (Table 3). Slaughterhouse wastes (e.g., MBM) are characterized by low PTE concentrations, followed by composts and digestates. Most of the certified (German) household waste digestates do not comply with the copper (Cu) thresholds of 200 mg kg^{-1} DM set by the EU legislation for organic farming (EC, 2008). In addition, the digestate Zn concentration is often higher than the EU threshold values of 70 mg kg^{-1} DM. Relatively high PTE concentrations have been found in biosolids, with a large spatial variation (e.g., between rural and urban areas) (Kupper, 2008; Tavazzi et al., 2012). Varying PTE loads were reported depending on the different approaches for production of sewage ash (Egle et al., 2013).

Struvites have been found to have low PTE concentrations on a DM base, except for Zn and chrome (Cr). High PTE depletion rates are reported for wet chemical approaches (95%–100%), partial depletion of PTEs with ASH DEC[®] and LeachPhos[®] (20%–100%), and no decontamination with RecoPhos[®] (Egle et al., 2013, 2014). In biochar and hydrochar production, PTEs are concentrated with the exception of low-temperature sublimating elements such as Hg and Cd (Bridle and Pritchard, 2004).

3.3 Organic Contaminants

There are no inventories available for the contents of organic pollutants and pharmaceuticals, allowing balanced comparisons across the different types of P recycling sources. The available data indicate the lowest concentrations of organic pollutants in MBM (Eljarrat et al., 2002; Möller and Schultheiß, 2014, 2015), followed by animal manures and food waste digestates (Berset and Holzer, 1995; Brändli et al., 2007; Govasmark et al., 2011; LfU, 2007; Mairitsch et al., 2011). However, MBM may contain residues of pharmaceuticals such as tetracycline (Kühne et al., 2000).

Higher POP concentrations than for MBM were found in urban organic waste composts (Brändli, 2006; Brändli et al., 2005; Déportes et al., 1995; Kuch et al., 2007). However, concentrations of POPs of composts may be as low as soil background levels (Erhart and Hartl, 2010). The POP database for digestates is weak, however, the few available studies indicate low levels of these compounds (BLfU, 2007; Kuch et al., 2007; Pfundner, 2004 cited by Mairitsch et al., 2011). The highest concentrations of POP are usually found in biosolids (Berset and Holzer, 1995; Clarke and Smith, 2011; Petersen et al., 2003; Rideout and Teschke, 2004; Stasinakis, 2012;

Stevens et al., 2001, 2003; Stevens and Jones, 2003). A Swiss inventory from more than 20 locations in a monitoring network found PAHs, PCBs, organochlorine pesticides, phthalates, UV filters, biocides, synthetic fragrances, polybrominated flame retardants, and surfactants in biosolids, with concentrations ranging between a few $\mu\text{g kg}^{-1}$ to several g kg^{-1} DM (Kupper, 2008). The author concluded that the calculated loads of contaminants which end up in soils via sludge could be substantial compared with the other input sources (Kupper, 2008). The concentrations of several important pollutants like polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/F) and PCBs in biosolids have been decreasing over time due to changes in legislation (Brändli, 2006; Zennegg et al., 2013).

We found only a few studies that evaluate the presence of POPs such as PCBs and PAHs in struvite precipitated from sewage treatment plants. These show low levels of POPs in struvite (Kern et al., 2008; Steinmetz and Meyer, 2014; Uysal et al., 2010). It is likely that these micropollutants were concentrated and incinerated with the remaining filter cake, the solid residue after waste water treatment. The study of Steinmetz and Meyer (2014) found only small amounts of pharmaceuticals attached to the struvite crystals, and these could be removed by rinsing the crystals after filtration. It was found that more than 98% of hormones as well as pharmaceuticals remained in solution during selective P precipitation (Ronteltap et al., 2007). However, uncertainties remain regarding pathogens such as *Ascaris* eggs or viruses in struvite (Decrey et al., 2011).

Organic pollutants and pharmaceuticals are destroyed nearly completely by incineration (Egle et al., 2013; Lindberg et al., 2015; Rulkens, 2007) as shown for POPs after treatment in a cupola furnace (Mephrec metallurgical sludge or ash treatment) (Lindberg et al., 2015; Scheidig, 2009). The production of chars from sludges including HTC chars degrades some of the organic pollutants, however in many cases significant amounts of chlorinated aromatic compounds are formed as recalcitrant transformation products (Oleszczuk et al., 2014; Weiner et al., 2013).



4. AGRONOMIC ASPECTS

4.1 P Forms and P Solubility

4.1.1 P Forms in RPFs

In organic amendments, the majority of the P is actually present in inorganic forms, like water-soluble inorganic phosphate as well as precipitated Ca-phosphates, struvites, etc. For example, in composts as well as in digestates, more than 55% of the P is present as inorganic P (Frossard

et al., 2002; Kuo et al., 1999; Preston et al., 1986; Traoré et al., 1999). In biosolids, organic P forms account for only about 5%–15% of total P (Annaheim et al., 2015; Frossard et al., 1997; Krogstad et al., 2005). Orthophosphate can react with Mg^{2+} and cations like NH_4^+ or K^+ to form struvite, and with Ca^{2+} , Fe^{3+} , Fe^{2+} , and Al^{3+} to form a series of minerals (Chang et al., 1983; Hjorth et al., 2010; Jokinen, 1990; Maguire et al., 2001; Nelson et al., 2003). In MBM, the primary P mineral is a Ca-deficient hydroxyapatite $[\text{Ca}_{10-x}(\text{HPO}_4)_x(\text{PO}_4)_{6-x}(\text{OH})_{2-x} \ (0 < x < 1)]$ (Jeng et al., 2006; Wopenka and Pasteris, 2005) which is somewhat more reactive than apatitic phosphate rock due to a higher content of carbonates formed by isomorphic substitution of P (Bekele and Höfner, 1993; Kahiluoto and Vestberg, 1998).

In composts, digestates, and biosolids, the majority of organic P consists of various orthophosphate monoesters such as inositol phosphates and sugar phosphates, while diesters such as phospholipids and nucleic acids often make up less than 5% of total P (Annaheim et al., 2015; Chang et al., 1983; Haynes et al., 2009; Peng et al., 2010; Preston et al., 1998; Toor et al., 2006). Both composting and anaerobic digestion lead to the transformation of organic P into inorganic P (Field et al., 1984) and reduce the ratio of easily soluble P to recalcitrant P compounds (Möller and Müller, 2012; Wei et al., 2015). This occurs also in anaerobic digestion when the pH of the digestate increases (Möller and Müller, 2012) shifting the chemical equilibrium toward the formation of phosphate ($\text{HPO}_4^{2-} \rightarrow \text{PO}_4^{3-}$) and subsequent precipitation as calcium- or magnesium-phosphate (e.g., $\text{Ca}_3(\text{PO}_4)_2$) as well as struvite (Hjorth et al., 2010; Nelson et al., 2003).

The P form in struvite is PO_4^{3-} incorporated into a crystal with Mg^{2+} and another cation (NH_4^+ , K^+ , among others), however, precipitates may contain other P compounds like Ca-phosphates, Al-phosphates, or Fe-phosphates as coprecipitates (Darwish et al., 2016; Le Corre et al., 2005; Lee et al., 2013, 2016; Marti et al., 2010; Muster et al., 2013; Römer, 2013a, b, c; Yan and Shih, 2016).

The thermal treatment of sewage and other organic wastes leads to significant transformations of the P. For example, incineration transforms organic and rather simple inorganic P molecules to crystalline molecules like Whitlockite ($\text{Ca}_3(\text{PO}_4)_2$) as well as Whitlockite-like compounds $\text{Ca}_9\text{Fe}(\text{PO}_4)_7$ or $\text{Ca}_9\text{MgK}(\text{PO}_4)_7$, Al-phosphates (AlPO_4) (Brod et al., 2015; Komiyama et al., 2013; Nanzer et al., 2014a; Peplinski et al., 2009), and at temperatures $>700^\circ\text{C}$ the formation of hydroxyapatites ($\text{Ca}_5(\text{PO}_4)_3(\text{F,OH,Cl})$) has been observed (Cascarosa et al., 2012; Coutand et al., 2008; Deydier et al., 2005; Thygesen et al., 2011). In chars obtained from hydrothermal carbonization (HTC-chars) and biochars

obtained from pyrolysis, P may be incorporated in or occluded by the aromatic structures formed during treatment (Cantrell et al., 2012), although this depends on the treatment temperature. In biochars, P will likely be associated with Ca and Mg due to biochar's elevated pH (Ippolito et al., 2015). HTC leading to hydrochars may also reduce the solubility and reactivity of P in the solid phase by increasing the share of apatitic P (Dai et al., 2015).

4.1.2 Chemical P Extractability

The water solubility of P in fertilizers is often referred to as a quality indicator for plant P availability (Brod et al., 2015; Kern et al., 2008; Kratz et al., 2010; Römer, 2013a, b, c). Other indicators are the solubility in 2% citric acid (e.g., for assessment of Thomas phosphates), neutral ammonium citrate (e.g., for assessment of water-soluble fertilizers like superphosphate and triple superphosphate), 2% formic acid extractable P (e.g., for assessment of phosphate rock), or NaHCO₃-soluble P (Bekele and Höfner, 1993; Brod et al., 2015; Kratz et al., 2010; VDLUFA, 1995). All RPFs considered in this chapter have a low water solubility for P. For example, in struvite, 1%–2% of the P is water soluble while about 50% is soluble in citric acid (Cabeza et al., 2011; Johnston and Richards, 2003; Kern et al., 2008; Kratz et al., 2010; Römer, 2013a, b, c). Only a small proportion of P in organic fertilizers like biosolids, composts, and digestates as well as animal manures is water soluble (<20%) (Krogstad et al., 2005; Maguire et al., 2001; Toor et al., 2006). The water solubility of P in ashes and MBM is also low (Kratz et al., 2010).

4.2 Plant P Availability and the Relative P Fertilizer Effectiveness

4.2.1 General Aspects and Methodological Approach

To conduct the present assessment of the plant P effectiveness of the RPFs, data were compiled from pot experiments conducted within the project, unpublished data provided by associated colleagues, and previously published field and pot experiments. We defined the P fertilizer value in field and pot experiments relative to a water-soluble mineral P fertilizer, corrected for the P offtake from the untreated control. The measure “relative P fertilizer effectiveness” (rPeff) was calculated by the following formula:

$$\text{rPeff}(\%) = 100 \times (P_{\text{offtake RPF}} - P_{\text{offtake 0}}) / (P_{\text{offtake WSP}} - P_{\text{offtake 0}}) \quad (1)$$

where $P_{\text{offtake RPF}}$ is the mean of the plant P offtake obtained with the RPF, $P_{\text{offtake 0}}$ is the mean of the plant P offtake of the unfertilized control treatment, and $P_{\text{offtake WSP}}$ is the mean of the plant P offtake measured with the water-soluble mineral fertilizer control treatment (e.g., triple superphosphate).

4.2.2 Relative P Fertilizer Effectiveness of Organic RPFs

The high rPeff of composts and biologically precipitated biosolids (Fig. 2) is probably a mixed result, composed by direct effects due to application of mineral and organic P (Toor et al., 2006), but partly also indirect effects on plant P availability, described in Section 4.2.5. The lower rPeff of chemically precipitated biosolids can be explained by the high Al and/or Fe doses resulting in biosolids having low available P concentrations, with Fe and Al phosphates as dominant P forms (Shober and Sims, 2007; Strawn et al., 2015). Precipitants added in excess may also interact with soil P decreasing even the availability of soil P. The short-term effects on plant P uptake followed the order Ca treated > Fe treated > Al treated (Soon and Bates, 1982; Torri et al., 2017), probably as a consequence of differences in the binding energy (Delgado and Torrent, 1997; Elliott et al., 2002). Conversely, biosolids obtained by biological P removal exhibit higher rPeff as a consequence of both elevated total P and water-extractable P (Ebeling et al., 2003; Penn and Sim, 2002; Stratful et al., 1999). However, at slightly acidic, neutral, or alkaline soil pH, Al-, and Fe-bound P will be desorbed by transformation of the Al- and Fe-phosphates into water-soluble P and precipitation of Al- and Fe-oxides (Brod et al., 2015; Huang et al., 2008; Lindsay, 1979), releasing the orthophosphate to the soil solution and then becoming plant available.

Among the organic P fertilizers, MBM has the lowest rPeff, which is related to the apatitic structure of bone P (Jeng et al., 2006; Ylivainio et al., 2008). As the dissolution of apatitic P requires H^+ ions, soil pH is a driving factor influencing the P release from MBM (Jeng et al., 2006; Ylivainio et al., 2008). Therefore, plant P availability is higher in soils with a pH < 6 than in neutral and alkaline soils (Albert, 2008; Bekele and Höfner, 1993; Jeng et al., 2006). The highest overall P efficiency can probably be achieved when the bone fraction of MBMs is used as animal feeds and then applied as fertilizer, since the acid environment in parts of the digestive tract is highly efficient in destroying the apatitic structures and mobilizing the contained P (Shastak et al., 2012; Van der Klis and Versteegh, 1996). However, for ethical and hygienical reasons this is a

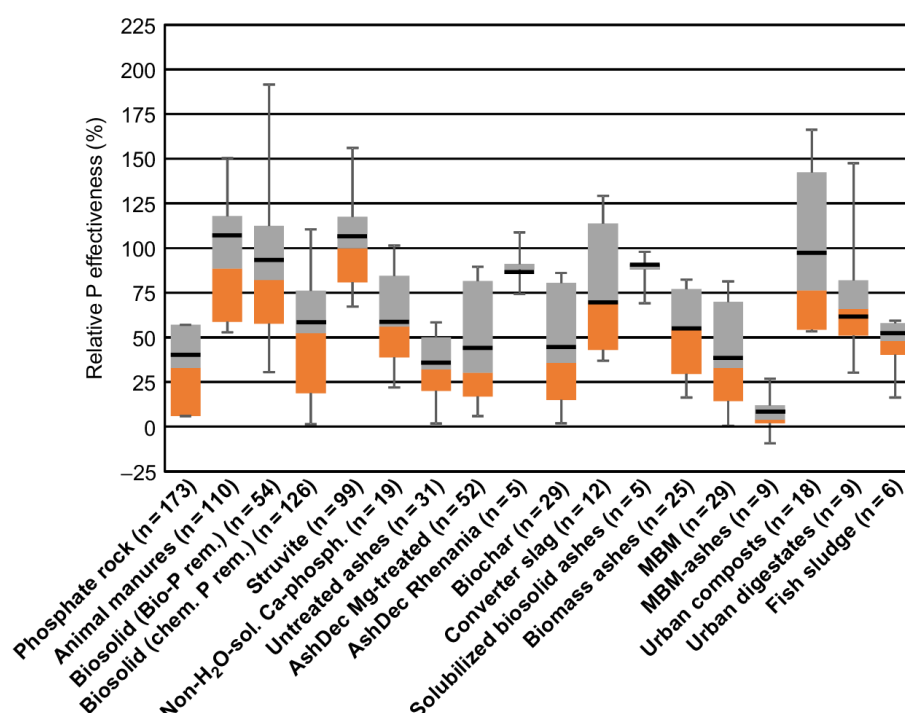


Fig. 2 Boxplot of the relative phosphorus (P) effectiveness of recycled P fertilizers (% of water-soluble P fertilizer). *Plot* indicates 50% of the values, whiskers 90%, the *thick bar* indicates the mean (References: see Achat et al., 2014a; Ackerman et al., 2013; Albert, 2005; Antonini et al., 2012; Appel and Friedrich, 2016; Árendás and Csathó, 2002; Bach, n.d.; Barbazan et al., 2009; Barker, 2012; Bekele and Höfner, 1993; Bezolla et al., 1994; Blake et al., 2000; Bolland et al., 2001; Brod et al., 2015, 2016; Cabeza et al., 2011; DeHaan, 1980; Delin, 2015; Delin et al., 2011; Ebeling et al., 2003; Ebertseder and Gutser, 2001; Eghball and Power, 1999; Eichler-Löbermann et al., 2007; Ellis et al., 1955; Fardeau et al., 1988; Foereid, n.d.; Frossard et al., 1996; Gell et al., 2011; Ghosh et al., 1996; González-Ponce et al., 2009; Goss and Stewart, 1979; Grigatti et al., 2015; Hao et al., 2015; Iglesias Jimenez et al., 1993; Johnston and Richards, 2003; Kahiluoto et al., 2015; Kaiser et al., 2010; Katanda et al., 2016; Kratz et al., 2010; Kristaponyte, 2005; Krogstad et al., 2005; Kuligowski et al., 2012; Kurz et al., 2014; Lekfeldt et al., 2016; Leon et al., 1986; Leytem and Westermann, 2005; Leytem et al., 2011; Liu et al., 2016; Mallarino and Haq, 2013; Massey et al., 2009; McCoy et al., 1986; McLaughlin and Champion, 1987; Meena et al., 2007; Meyer and Oberson, n.d.; Miller and O'Connor, 2009; Mohanty et al., 2006; Montgomery et al., 2005; Morel and Fardeau, 1990; Motavalli et al., 1989; Nachimuthu et al., 2009; Nanzer et al., 2014a, b; O'Connor et al., 2004; Øgaard and Brod, 2016; Oladeji et al., 2008; Onnen, 2001; Plaza et al., 2007; Richards and Johnston, 2001; Römer, 2006, 2013; Römer and Samie, 2002; Schick, 2010; Schiemenz and Eichler-Löbermann, 2010; Schuh et al., 2013; Severin et al., 2013, 2014; Siebers et al., 2012; Siegenthaler, 1990; Sikora and Enkiri, 2003; Simons, 2008; Simons and Clemens, 2006; Sinaj et al., 2002; Slaton et al., 2013; Starostka et al., 1955; Sutton et al., 1986; Symanczik, n.d.; Szogi et al., 2010; Talboys et al., 2016; Terman and Taylor, 1965; Thompson, 2013; Van der Paauw, 1965; Vogel et al., 2015; von Tucher et al., 2012; Waida and Weinfurter, 2011; Weber, 2010; Weinfurter et al., 2005; Wilken et al., 2015; Wollmann et al., 2016; Ylivainio et al., 2008; Zapata and Zaharah, 2002; Zvomuya et al., 2006).

controversial approach. Currently, we do not know of any other alternative biological approaches for P solubilization from bone P in order to transform the P from apatitic structures into more reactive P forms.

4.2.3 Relative P Fertilizer Effectiveness of P Precipitates

Struvites and Ca-phosphates are the main fertilizer products obtained by chemical approaches to P recovery from liquid waste streams. The available data indicate a wide range of chemical forms present in fertilizers obtained from precipitation processes, strongly influencing the respective rPeff. The data indicate a considerably higher rPeff for struvites than for the P-RoC© product composed by nonwater-soluble Ca-phosphates with a high rate of hydroxyapatites (Fig. 2). Furthermore, the rPeff of nonsoluble Ca-phosphates like the P-RoC© product strongly depends on the pH of the test soil (Fig. 3C) as also found by Cabeza et al. (2011), while the rPeff of struvites is independent of the test soil pH (Fig. 3D). Therefore, from an agronomical point of view, struvite is a desirable recovered P product due to the higher rPeff and the independency of the rPeff from soil pH.

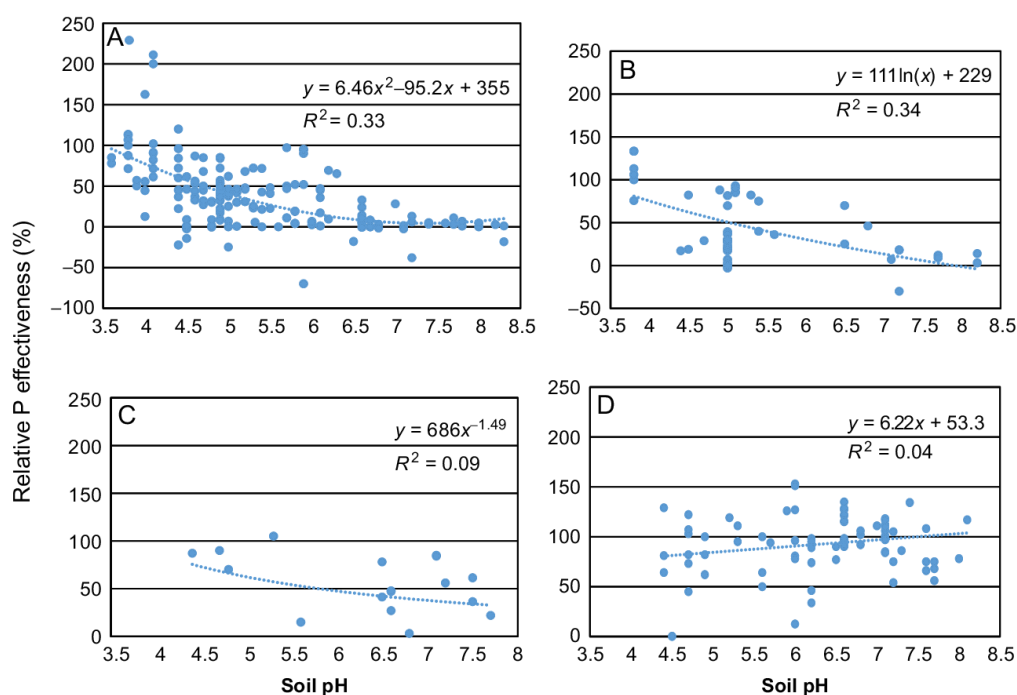


Fig. 3 Influence of the soil pH on the relative phosphorus (P) fertilizer effectiveness (% of water-soluble P fertilizers) of (A) phosphate rock (no. of data pairs: 173), (B) Mg-treated biosolid ashes (no. of data pairs: 47), (C) non-H₂O soluble Ca-phosphates (no. of data pairs: 19), and (D) struvite (no. of data pairs: 80) (for references: see the caption of Fig. 2).

However, while struvite is a good product per se, it should be noted that the residual waste will have a higher concentration of contaminants relative to P content. Considering that struvite extraction efficiency is less than 50% there is a risk that the residual P will be more difficult to recycle.

Struvites are often described as slow release P fertilizers due to their low water solubility (Bashan and Bashan, 2004; Diwani et al., 2007; Rahman et al., 2014; Talboys et al., 2016); however, the present compilation indicates that struvite fertilizers have a high rP_{eff} in both acidic and calcareous soils, comparable to or even higher than for water-soluble mineral P fertilizers (Fig. 2). Therefore, struvite-P shows a low solubility but a higher reactivity. This result is confirmed by several studies (e.g., Achat et al., 2014b; Bonvin et al., 2015). Interestingly, struvite fertilizer tends to have a slower dissolution rate when root growth is absent (Ahmed et al., 2016). Probably, the removal of nutrients included in the struvites from soil solution by plant roots leads to a continuous decomposition of struvite crystals by increasing the concentration gradient of the involved elements (Achat et al., 2014a, b). The low water solubility will protect struvite-P from being adsorbed on soil colloids.

The available data indicate a large variation of the rP_{eff} for struvites, even when tested under identical growing conditions in one experiment (Antonini et al., 2012; Johnston and Richards, 2003; Römer, 2013a; Wilken et al., 2015; Wollmann et al., 2016). Some of the different observations might be due to different crystal sizes of tested struvites. For example, Bonvin et al. (2015) used a fine powder, whereas Talboys et al. (2016) applied crystals with about 2.4 mm diameter. Furthermore, struvites are sometimes also described to be better suited for soils of moderate or low pH, with a limited effectiveness in soils with high pH (Ackerman et al., 2013; Bowden et al., 1980; Kataki et al., 2016). However, in the present survey, we could not identify any significant effect of soil pH on the rP_{eff} of struvites (Fig. 3D). Therefore, other factors related to differences in the struvite quality might be responsible for variations in P fertilizer performance. According to the molecular formula of struvite ($MgNH_4PO_4 \cdot 6H_2O$), the theoretical P concentration is 12.6%, however, the measured P concentrations in struvites are often lower (Table 2). This may be due to the presence of Ca and ferric ions in the precipitation tank/reactor which significantly inhibit the formation of struvite crystals (Marti et al., 2010; Yan and Shih, 2016), especially under higher pH conditions ($pH > 8.5$) (Darwish et al., 2016) and can result in impurities in the final product. The Mg–Ca– NH_4 – PO_4 system may provide at least seven different crystalline phases as precipitates, differing in soil reactivity and plant P availability. They include struvite,

hydroxylapatite ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$), newberyite ($\text{MgHPO}_4 \cdot 3\text{H}_2\text{O}$), brushite ($\text{Ca}[\text{PO}_3(\text{OH})] \cdot 2\text{H}_2\text{O}$), or their dehydrated form monetite ($\text{Ca}[\text{PO}_3(\text{OH})]$), whitlockite ($\text{Ca}_3(\text{PO}_4)_2$), or their anhydrous, sodic form merrillite ($\text{Ca}_9\text{NaMg}(\text{PO}_4)_7$), and octocalcium phosphate ($\text{Ca}_8\text{H}_2(\text{PO}_4)_6 \cdot 5\text{H}_2\text{O}$) (Muster et al., 2013), depending on the chemical environment during the precipitation process. For example, the Ca/Mg molar ratio in the initial solution is negatively related to the struvite proportion in the precipitates (Marti et al., 2010; Muster et al., 2013; Yan and Shih, 2016). Therefore, struvites may contain several other elements related to impurities and coprecipitation of Fe-phosphates (Römer, 2013a, b, c) or Ca- and Mg-phosphates (Le Corre et al., 2005; Lee et al., 2013, 2016), and carbonates. In experiments of Marti et al. (2010) with different wastewaters, the percentage of precipitated P as struvite measured by X-ray diffraction varied between 37% and 99%. Furthermore, the N/P molar ratios also influence the ratio between struvite and newberyite in the solid phases of the recovered products (Lu et al., 2016). Both Ca- and Fe-phosphates have a considerably lower P fertilizer effectiveness than struvite (Bauer et al., 2007; Johnston and Richards, 2003; Römer, 2013a; Simons, 2008; Wollmann et al., 2016), potentially explaining the high variation in struvite rPeff.

Also the specific composition of Ca precipitates may influence the P fertilizer value of the obtained products. For example, monocalcium and dicalcium phosphates have a high P fertilizer value, while precipitates and nonwater-soluble Ca-phosphates with a higher Ca/P ratio have a moderate to low rPeff (e.g., from P-RoC©) (Römer, 2013a, b). Precipitates may also have indirect effects on soil P availability, for example, silicate contained in the product may compete with phosphate sorption sites in the soil, namely by replacing orthophosphate on Fe and Al (hydr)oxides and releasing orthophosphate into the solution (Koski-Vähälä et al., 2001). Nonwater-soluble amorphous compounds were more labile and resulted in greater P uptake by plants than crystalline compounds contained in RPF (Grigatti et al., 2015). However, also the plant availability of amorphous compounds varies widely, depending on Ca:P ratio, aggregate size, and inclusion of cations such as Mg (Grigatti et al., 2015).

We can conclude that in future experiments assessing the plant P availability of RPFs obtained from chemical approaches of P recovery, a better description of the precipitate production process (e.g., pH in the solution during precipitation; crystal size), and the precipitate composition is needed to learn more about how the specific production factors influence the struvite and Ca-precipitate composition.

4.2.4 Relative P Fertilizer Effectiveness of P in Incineration Ashes

The P fertilizer effectiveness of untreated ashes after incineration of any kind of organic matter is very low (Fig. 2) and strongly dependent on the soil pH (data not shown). The P fertilizer value will depend on the feedstocks used (Nanzer et al., 2014a), as well as on the conditions during combustion which induce a range of structural modifications, such as crystallization of calcium phosphate or substitution reactions, and may induce a decrease of the particles' specific surface area because particles will stick to each other (Deydier et al., 2005). For instance, sewage sludge incineration ashes contain different amounts of sparingly available P-forms like brushite, stanfieldite, and hydroxyapatite-like compounds (Nanzer et al., 2014a) significantly reducing their plant P availability. Still, the specific surface area of the ashes studied in Nanzer et al. (2014a) varied from 4 to 57 m² g⁻¹. Furthermore, the higher the incineration temperature, the stronger the decrease of the P fertilizer value (e.g., Christel et al., 2014; Thygesen et al., 2011). The increased variation of the measured rPeff with decreasing soil pH suggests an increasing influence of the specific ash fertilizer characteristics on the P fertilizer value with decreasing soil pH. Thus, ashes from incineration may be used in acidic soils for improvement of plant P supply and for providing carbonates for buffering the soil pH, but they are not suited as P fertilizers for soils with a pH > 6.0. From an agronomical point of view, any incineration approach represents a downgrading of the treated material in terms of specific P fertilizer value and the flows of N, S, and organic matter. Consequently, ashes should be treated appropriately—e.g., by acidic solubilization—before application to slightly acidic, neutral, or alkaline soils in order to get a plant available P fertilizer. To the best of our knowledge, a biological approach for P solubilization of ashes—e.g., by supplementation to animal feeds—has not been tested. However, several treatment approaches are available for chemical P solubilization of ashes, either by wet chemical treatment with acids (e.g., LeachPhos®, PASCH) aiming to provide monocalcium phosphate or struvites (Egle et al., 2014, 2016). Or by thermochemical treatment with Na and K additives, which under reducing conditions react with the Ca-phosphate and transform it to buchwaldite (also called rhenanite—CaNaPO₄) or calcium potassium phosphate (CaKPO₄) when calcined in the presence of Na or K (ASC DEC® Rhenania) (Egle et al., 2016; Herzel et al., 2016; Stemmann et al., 2015). These calcined P fertilizers have a high plant P availability similar to water-soluble mineral P fertilizers and struvite (Fig. 2).

An alternative to incineration of sewage sludge and other organic wastes is char production (Ippolito et al., 2015; Yuan et al., 2015). The dataset on P availability of chars is rather weak and the available data indicate a low rPeff (Fig. 2), except for pyrochars produced from enhanced biological P removal processes (Symanczik et al., unpublished data). The presence of multivalent metal cations, such as Al, Ca, Mg, and Fe, is responsible for the formation of insoluble P in the chars produced from chemically precipitated sewage sludge (Dai et al., 2015, 2016; Huang and Tang, 2016; Qian and Jiang, 2014; Uchimiya et al., 2015; Uchimiya and Hiradate, 2014; Wu et al., 2012a), reducing the reactivity and plant availability of P. Furthermore, P availability in chars depends on feedstock material and is inversely related to treatment temperature (Chan et al., 2007; Christel et al., 2014; Gaskin et al., 2008; Zheng et al., 2013). There are some indications, that the P availability of ash produced at 300°C can exceed that of char produced at the same temperature (Christel et al., 2014), suggesting that chars are even worse than ashes regarding the P fertilizer value. Therefore, the production of chars seems not to be a best practice approach in order to obtain an efficient P fertilizer. An interesting alternative may be to apply alkaline pyrolysis directly to sewage sludge rather than to treat sewage sludge ashes (Stemann et al., 2015; Herzel et al., 2016), or to add Na before the pyrolysis process (Appel et al., 2016).

4.2.5 Added Fertilizer P Interactions

The compiled data indicate that the rPeff of RPFs is also driven by other factors than the soil pH (Fig. 3B and D), e.g., the crop, the experimental setup (field vs pot experiments), and specific quality characteristics of the RPFs, leading to added fertilizer P interactions (Oberson et al., 2010), which often were not taken into consideration when analyzing the rPeff of fertilizers based on the difference method (Eq. 1). For example, several studies have shown that soil P sorption strength was decreased by the application of organic amendments (Agbenin and Igbokwe, 2006; Azeez and Van Averbeke, 2010; Gao et al., 2014; Jiao et al., 2007; Guppy et al., 2005; Øgaard, 1996; Sato and Comerford, 2005; Siddique and Robinson, 2003; vanden Nest et al., 2016), changing the soil P sorption isotherms (Sato and Comerford, 2005) and enhancing plant P availability. It is stated that organic acids released during decomposition of organic matter will decrease the soil P sorption strength on mineral soil particles by (i) competition between organic acids and orthophosphate on Al- and Fe-oxide binding

sites, or (ii) reducing the numbers of binding sites by chelation and solubilization of soil Al- and Fe-oxides with organic acids (Grimal et al., 2001; Nziguheba et al., 1998; vanden Nest et al., 2016). Organic matter inputs also stimulate microbial growth, resulting in microbial P immobilization (Oberson et al., 2010). These effects will no longer be available when wastes are incinerated, leading to ashes low in organic matter, or when P is chemically recovered by precipitation.

Furthermore, all RPFs are multicomponent fertilizers containing a broad range of elements (Table 2), which may interact with P (Shaheen and Rinklebe, 2015). For instance, P may interact with anions and cations such as Na^+ , K^+ , ammonium (NH_4^+), Mg^{2+} , Ca^{2+} , Fe^{2+} and Fe^{3+} , Al^{3+} , bicarbonate (HCO_3^-), silicon (e.g., Si^{4+}) and sulfate (SO_4^{2-}) (Ahmad et al., 2008; Curtin et al., 1987; Shaheen and Rinklebe, 2015), and P sorption and desorption have been shown to vary with the soil ionic strength and cation species in the soil solution (Ahmad et al., 2008; Nair et al., 1984). Supply of Ca^{2+} and Mg^{2+} ions may promote the P-sorption through electrostatic forces (Shaheen and Rinklebe, 2015; Stachowicz et al., 2008; Vandecasteele et al., 2014; vanden Nest et al., 2016). Therefore, the supplementation of some metals like Ca, Fe, and Al (e.g., for P precipitation in wastewater treatment plants, for reducing N emissions, and for desulfurization in anaerobic digesters) may negatively influence plant P availability. These elements may form insoluble precipitates with P, and sometimes result in a negative P fertilizer value due to immobilization of soluble soil P (Bøen et al., 2013; Guppy et al., 2005; Huang et al., 2008; Krogstad et al., 2005; Maguire et al., 2001; Siddique and Robinson, 2003; Toor et al., 2006). This is probably the reason for the lower rPeff of biosolids based on chemical P precipitation, and may also influence the P fertilizer effectiveness of composts and digestates. Otherwise, Na^+ may induce a higher P release of P associated with oxide surfaces or due to dissolution of Ca-phosphates (Ahmad et al., 2008; Curtin et al., 1987). Further factors potentially increasing the soil P plant availability could be a high ionic strength resulting from the surpluses of elements and provided by anions such as Si^{4-} , SO_4^{2-} , and OH^- that compete with phosphate ions for surface sorption sites, reducing the amount of P adsorbed on soil colloids (Ahmad et al., 2008; Nair et al., 1984). Therefore, in order to improve our understanding regarding the fertilizer value of RPFs, a better description of the feedstock materials used (e.g., chemically or biologically treated biosolids), a thorough description of the production conditions (pH range, temperature, etc.), and of the element composition of the finally produced fertilizer might be approaches for a better understanding of RPF effects on plant P availability.

Another added fertilizer P interaction may arise from the effect of RPF application on soil pH. Most RPFs increase the soil alkalinity (Eghball, 1999; Hargreaves et al., 2008; Möller and Schultheiß, 2015; Pockney and Sumner, 1997; Whalen et al., 2000). In organic RPFs, biomass usually has a surplus of cations (e.g., K^+ , Ca^{2+} , Mg^{2+} , Na^+ , and NH_4^+) over anions (e.g., $H_2PO_4^-$, SO_4^{2-} , and Cl^-), which are compensated for by bicarbonate (HCO_3^-), carbonate (CO_3^{2-}), or anions of organic acids (Hargreaves et al., 2008; Husted et al., 1991; Tang et al., 1999). The decomposition of organic acids as well as of carbonates to CO_2 in the soil are proton consuming processes (e.g., Yan et al., 1996) leading to a liming effect. For example, in composts on average 8% of the carbon is carbonate-C (He et al., 1995), and composts have a liming effect which corresponds to 5%–10% of calcium carbonate (Gutser and Ebertseder, 2002). Also most inorganic RPFs provide alkalinity to soil. For example, struvite has a liming effect, as struvite dissolution in the soil is a proton consuming process ($MgNH_4PO_4 + 2H^+ \rightarrow Mg^{2+} + NH_4^+ + H_2PO_4^-$) (Bauer et al., 2007; Rahman et al., 2011). In ashes, the acid neutralization capacity is equivalent to 15%–50% of $CaCO_3$ (Ohno, 1992; Zhang et al., 2002). Combustion removes elements like S and Cl, increasing the liming properties of the treated organic feedstock. The higher the combustion temperature, the higher the liming properties of the residual product (Hass et al., 2012; Ippolito et al., 2015; Wang et al., 2012). The liming effect of biochars depends on the treatment procedure (Hass et al., 2012).

4.2.6 Implications of the Results Regarding the Relative P Fertilizer Efficiency

The results indicate that the rPeff of many RPFs is higher than phosphate rock, and therefore, if a farmer needs external P inputs, RPF should be preferred over phosphate rock. Especially in neutral and alkaline soils, phosphate rock, untreated ashes, and MBM are not recommended as the P release will be slow. In this case, compost, digestates, converter slags, and calcined P fertilizers based on ASH DEC[®] Rhenania and struvite are more suitable (Fig. 3A–D). The same is true for biologically (Bio-P) precipitated biosolids.

One of the major challenges for assessing the suitability of different approaches of RPFs for an efficient P recycling is the assessment of the short-term and long-term P fertilizer effectiveness. It is well known that the short-term P fertilizer effectiveness varies widely among the assessed RPFs (Fig. 2), also depending on the pH of the tested soils (Brod et al., 2015; Cabeza et al., 2011; Römer, 2013a, b, c; Wollmann et al., 2016). However, the long-term fate of RPFs in soils is not well understood. For

phosphate rock, the available data indicated a low P release even over a period of 10 years or more (Bolland et al., 1989). However, similar to soil P, it can be assumed that most of the P from phosphate rock or RPFs will become plant available over a very long-term perspective, even for the less reactive P species. Furthermore, the field application of high amounts of P fertilizers with a negligible short-term plant P availability in order to secure plant P supply over a longer period implies higher soil total P concentrations and therefore an increased risk of P losses. This ineffective P would also be a burden in nutrient budgets.

The results regarding the P fertilizer value of RPFs have also several methodological consequences. For example, water P solubility is one of the most important parameters to characterize the quality of P-containing fertilizers, as it is often used as a tool for estimating plant availability and nutrient uptake effectiveness (Kratz et al., 2016). However, the results regarding the fertilizer value of struvites, animal manures, biologically precipitated biosolids, or Rhenania ASH DEC[®] show that the water solubility of P fertilizers and their plant availability in the soil are not necessarily correlated. Therefore, water solubility, or citric acid solubility alone, are not appropriate quality parameters for most RPFs, as a low solubility are not necessarily linked to a low reactivity of fertilizer P. Sequential extraction methods are laborious. Several studies indicate that the readily extractable P fractions (extracted using anion exchange resin plus NaHCO₃) may provide an assessment of the P availability of the RPF (Brod et al., 2016; Nanzer et al., 2014b). However, other authors did not find any correlation between the readily extractable P fractions and the plant P uptake (Wollmann et al., 2016).

Data on rPeff of phosphate rock and ashes show no or a small P fertilizer effect at neutral soil pH, and an increasing variation of the rPeff with decreasing soil pH (Fig. 3A and B). Therefore, quality assessments of a specific RPF with a high sensitivity to soil pH (e.g., ashes, MBM, and Ca-phosphates obtained by P-RoC© precipitation) and differentiation of the P fertilizer value among different provenances should be carried out in a pH range where the highest fertilizer value is expected in order to get a better differentiation among the proveniences. This approach must be complemented by plant growth experiments over a broad range of soil pH values in order to assess the basic suitability of RPFs in soils with differing pH.

4.3 Influence of Plant Growth Promoting Rhizobacteria on Plant P Availability of RPFs

Plant growth promoting rhizobacteria (PGPR) belongs to diverse genera of rhizosphere bacteria, e.g., *Pseudomonas*, *Bacillus*, and *Rhizobium* (Halpern

et al., 2015). It is stated that PGPRs may positively influence plant P availability by enhancing P mobilization (Gyaneshwar et al., 2002; Hinsinger, 2001). This mobilization might be achieved by direct mechanisms such as: (i) the secretion of gluconic acids leading to a direct anion exchange of PO_4^{2-} or by chelating Fe or Al associated with phosphates (Gyaneshwar et al., 2002), (ii) by soil acidification as a result of gluconic acid release for dissolution of Ca precipitates (Hinsinger, 2001), or (iii) by release of phosphatases which mineralize organically bound P (Richardson and Simpson, 2011). It may also be an indirect effect of root growth stimulation (Gómez-Muñoz et al., 2016; Sánchez-Esteva et al., 2016) or stimulation of arbuscular mycorrhizal fungi (Bonfante and Anca, 2009; Miransari, 2011). Various experiments with PGPR were conducted within the IMPROVE-P project yielding inconsistent results. Under controlled conditions, promising effects were mainly obtained with PGPRs in combination with organic RPFs such as composted animal manures, fresh digestate of organic wastes, and biosolids leading to improved maize growth (Thonar et al., 2017). However, when PGPRs were combined with mineral RPFs, beneficial effects were only rarely observed. In only one out of six experiments, the relative nutrient effectiveness of mineral RPFs was enhanced by PGPR application. Meyer et al. (2017) showed that *Pseudomonas protegens* CHA0 increased P availability from MgCl_2 -treated sewage ash in liquid media, but had no effect in soils. Also Wollmann et al. (2016) found no effect of PGPRs on plant P uptake by legumes, ryegrass, or maize grown in pot and field trials and fertilized with different RPFs. Therefore, the available data on the effects of PGPRs on rPeff of RPFs indicate that in most cases no relevant effects can be achieved in field. Data on field and pot experiments in soils with very low P levels indicate that the endogenous soil P mobilization potential is high in biologically active soils, thus PGPR application remained widely ineffective with only minor exception (e.g., Meyer et al., 2017; Wollmann et al., 2016; Symanczik et al., unpublished data). In general, it can be stated that positive effects are hardly obtained under field conditions, and that more detailed studies on the mode of action of PGPRs are needed to shed more light on how and under which conditions they stimulate plant growth and improve P uptake, to possibly develop efficient strategies for using PGPRs for crop production.

4.4 Fertilizer Effectiveness of Other Nutrients

Most RPFs also supply macro- and micronutrients other than P. For example, biosolids, urban organic waste digestates, and MBM provide relatively high amounts of N per unit P. However, in relation to plant needs even

these fertilizers are P accentuated. The relative N fertilizer effectiveness of biosolids in the year of application—compared to a mineral N fertilizer reference—ranged between 5% and 70% of the applied total N (Gilmour et al., 2003), and it was estimated to be approximately 35%–40% for anaerobically digested biosolids (Al-Dhumri et al., 2013; Cogger et al., 2004; Gilmour and Skinner, 1999), but only 10% for composted biosolids (Al-Dhumri et al., 2013). Biosolids also provide residual N for release in the following years (Cogger et al., 2013). It can be assumed that the sum of the short- and long-term N fertilizer effectiveness of biosolid digestates will be equivalent to animal slurries and digestates (70%–80%) and for biosolid composts equivalent to solid farmyard manures (50%–70%).

The relative N fertilizer effectiveness of urban organic waste composts in the year of application is rather low: –5% to +10% of the applied total N (Möller and Schultheiß, 2014). For urban organic waste digestates, it was assumed that the fertilizer value in the year of application corresponds to the ammonium to total N ratio (40%–75%) (Möller and Müller, 2012), while for MBM the relative N fertilizer value is approximately 70%–90% (Bøen and Haraldsen, 2013; Chen et al., 2011; Jeng et al., 2006). The sum of the short- and long-term N effectiveness is rather low for composts (20%–40%) (Amlinger et al., 2003; Diez and Krauss, 1997; Gutser and Claassen, 1994), whereas for digestates and for MBM it can be close to a liquid animal manure or mineral N fertilizer (70%–80% of the applied total N for digestates and 80%–85% for MBM) (Gutser and Claassen, 1994; Schröder et al., 2005). The RPFs may also contain significant amounts of K, which is generally readily plant available (Möller and Müller, 2012; Ohno, 1992).

Organic RPFs are also able to restore soil organic matter. There are several references that the organic matter of biosolids restored soil organic carbon on an equivalent or even higher level than animal manures (Gerzabek et al., 2001; Parat et al., 2005). The strong microbial decomposition of the organic matter during the sewage sludge treatment (either by anaerobic digestion or composting, or the combination of both) leaves the biosolid organic matter as a relatively recalcitrant residue (Gilmour et al., 2003). The C-conversion factor (transfer coefficient of the amendment-C to stabilized soil-C) based on long-term field experiments was found to range between 18% and 43% for urban organic waste compost (De Haan and Lubbers, 1984; Gutser and Ebertseder, 2002), and for biosolids between 17% and 27% (Cogger et al., 2013; De Haan and Lubbers, 1984; Gutser et al., 2010). Organic matter in digestates is relatively recalcitrant and was reported to have a C conversion of 40% (Gutser et al., 2010).



5. ENVIRONMENTAL PERFORMANCE

5.1 General Aspects

Losses of nutrients like P in the global food cycle as well as increased GHG emissions compromise the ability of future generations to meet their own needs, a key indicator for assessment of the sustainability of current practices. Therefore, the recycling procedures should have high nutrient recovery rates, minimize the utilization of nonrenewable resources, and have a low negative environmental impact (principle of ecology), whereas the final fertilizer product should not pose a risk to soil quality or human or animal health (principle of health and principle of care). Simultaneously, the chosen approaches for treatment and recycling should address and balance the concerns of the present generation (prevention of hygienic risks and pollution with organic and inorganic pollutants) with those of later generations (e.g., availability of P sources and GHG emissions). Prevention of pollution by PTEs and organic contaminants is a concern for present as well as future generations. There is an agreement that pollution with organic contaminants (e.g., pharmaceuticals) is a relevant concern for present generations, while there is less clarity about long-term implications. Many organic contaminants are degraded during fertilizer processing or storage or in the soil within biologically relevant timescales, while some are slowly degraded (e.g., organotins, brominated flame retardants, and high molecular mass PAHs) and only a few are highly persistent taking years (PCDD/F), decades (e.g., PCBs), or even centuries (PCDD/F, perfluorooctanesulfonic acid (PFOS), perfluorooctanoic acid (PFOA)) for decomposition (Eriksen et al., 2009; Haynes et al., 2009; Hemkemeyer et al., 2015; Ockenden et al., 2003; Sinkkonen and Paasivirta, 2000; Zareitalabad et al., 2013). In soil, degradation of many organic contaminants is retarded compared to other media (Sinkkonen and Paasivirta, 2000). Most POPs occur not only in organic wastes, but are also distributed in the environment by other routes, e.g., gaseous (e.g., dioxins and organochlorines).

5.2 Risk Assessment

5.2.1 Methodological Approach

The overall aim of the risk assessment was to forecast the potential risk of accumulation of PTE and selected POPs by modeling the probable input and output flows in order to calculate predicted soil concentrations (PSCs). To assess the PTE accumulation in soil due to fertilizer application,

Table 4 Soil Background Concentration in Agricultural Topsoil, Atmospheric Deposition, Lime Input and Proposed Threshold Values Used for Modeling Soil Accumulation Risk From PTEs

		Cd	Cr	Cu	Ni	Pb	Zn
Soil background concentration ^a	kg ha ⁻¹	0.92	308	56.2	121	106	221
Atmospheric deposition ^{b,c}	g ha ⁻¹ y ⁻¹	0.36	9.30	34.0	10.0	11.9	227
Input with lime ^{d,e}	g ha ⁻¹ y ⁻¹	0.14	0.92	1.38	0.78	0.94	2.00
Proposed threshold values in soil ^a	kg ha ⁻¹	3.25	244	163	163	163	488

^aFOREGS, 2005, Original values (mg kg⁻¹) were converted to kg ha⁻¹ assuming a soil mass in the plow horizon (25 cm) of 3250 t ha⁻¹.

^bNicholson et al. (2003).

^cMSC-E (2016).

^dSmolders (2013).

^eEriksen et al. (2009).

a mass balance model based on Smolders (2013) was used. The model included fertilizer, atmospheric deposition, and liming inputs (Table 4), and outputs via leaching and crop offtake. Projections were made over a 200-year period, with leaching and crop uptake dependent on the actual trace element concentrations in the soil (for more details: Weissengruber et al., n.d.). An annual fertilizer input of 11 kg P ha⁻¹ y⁻¹ was assumed in order to replace the typical net P exports for stockless arable organic farming systems (Weissengruber et al., n.d.).

The results presented in Table 5 indicate the relative increase of soil PTE concentration caused by the application of RPFs over the current background concentrations in soils, expressed as percentage of the difference between the soil background value and threshold values set by current legislation. Therefore, the PSC after 200 years of regular application, based on current soil background values and relative to the theoretically possible increase up to the threshold values, was calculated according to the formula:

$$\text{Relative PTE increase} = 100 \times (\text{PSC}_{\text{RPF}} - \text{soil base value}) / (\text{soil threshold value} - \text{soil base value}) \quad (2)$$

where PSC_{RPF} is the predicted soil concentration of PTE by repeated application of the assessed RPF; Soil base value is the current soil background PTE concentration; and Soil threshold value is the maximum tolerable soil PTE concentration.

Table 5 Assessment of the Predicted Change in Soil Concentrations of Potentially Toxic Elements (%) at Soil pH 7.0 Where Annual Precipitation Is in Excess of 100 mm, Relative to Soil Background and Soil Threshold Values When Different Recycled Phosphorus Fertilizers (RPFs) Are Applied

RPF	Cd	Cr	Cu	Ni	Pb	Zn	Cd/P	HMP	HMN
Green waste compost	16.6	17.2	21.8	18.9	22.2	60.4	184	7.26	0.72
Organic household waste compost	10.4	11.8	18.4	10.9	13.9	44.7	121	5.26	0.89
Organic household waste digestate	5.6	5.2	10.9	3.7	5.1	30.9	25.8	1.33	0.12
Catering waste digestate	1.6	1.3	3.8	−1.2	1.1	15.3	15.3	0.58	0.06
Meat and bone meal	0.6	1.2	1.7	−2.4	0.9	11.0	4.1	0.24	0.06
Biosolids	2.6	2.4	13.3	0.2	2.3	28.6	26.9	1.64	0.66
Struvite SSL	0.4	0.9	1.6	−2.5	0.9	10.6	1.17	0.05	0.03
Struvite AirPrex [®]	0.5	1.0	2.1	−2.1	1.8	10.1	1.19	0.08	0.05
Untreated SSA	3.8	2.9	17.5	−0.8	3.4	34.9	40.5	1.71	1.65
Rhenania ASH DEC [®]	0.6	3.4	8.9	−0.1	1.9	25.6	3.9	1.38	1.19
LeachPhos [©]	2.0	1.2	9.9	−2.3	1.1	17.2	19.8	0.90	0.88
Mephrec-slugs	0.7	3.5	4.1	−1.8	1.2	10.4	1.2	0.36	0.31
Phosphate rock	14.2	2.7	1.6	−1.9	0.9	10.7	173	2.46	2.46
Triple superphosphate	28.3	2.6	1.6	−2.3	0.9	10.8	110	1.56	1.56

Abbreviations: SSL, Stuttgart sewage sludge leaching; SSA, sewage sludge ash. Annual application rate equivalent to 11 kg P ha^{−1} for 200 years (based on data of [Weissengruber et al., n.d.](#)), heavy metal-phosphorus index (HMP) and heavy metal-nutrient index (HMN).

5.2.2 Potentially Toxic Elements

The PSC of PTEs over a period of 200 years indicates for all assessed RPFs a low to moderate risk, even in the worst case scenario shown in [Table 5](#). For most PTEs with the exception of Pb and Cu, the leaching influenced by the soil pH and the climatic water balance (=precipitation excess) are the key factors influencing the soil accumulation. The calculations for assessment of the relative change of soil PTE concentration after long-term application of RPF in relation to current soil background values and soil threshold values indicate the strongest increase and therefore the highest risk of PTE

accumulation in soil for currently permitted green waste (relative increase of 17%–60%; mean 26%) and urban organic waste composts (10%–45%; mean relative increase of 18%) (Table 5). These are both RPFs with low to moderate PTE concentrations on a DM base (Table 3). Model calculations for urban organic waste digestates indicate a low PTE accumulation risk (mean relative increase of 10%), in spite of higher PTE concentrations on a DM base than in composts. Modeling results indicate that application of biosolids will moderately influence the soil Cu and Zn concentration, while the other PTE concentrations are only slightly affected, resulting in a mean relative increase of PTE concentration by 8%. Sewage-based RPFs obtained by thermal treatments and subsequent depollution are efficient in reducing the soil PTE accumulation risk (e.g., LeachPhos[®], Rhenania ASH DEC[®], and converter slags). The lowest risk among the sewage-based RPFs was achieved with struvite. Long-term application of MBM will have only minor effects on soil PTE concentrations (mean: 2%).

The contamination with PTEs is not the main constraint limiting recycling of most RPFs, as shown by the results of the risk assessments (Table 5) and also indicated by former studies (e.g., Eriksen et al., 2009). Furthermore, according to the model calculations, the highest risks of soil accumulation are predicted for the elements Cu and Zn (Table 3), indicating a shift of the elements having a major accumulation risk. For mined P fertilizer sources (TSP and phosphate rock), the model calculations identified Cd as the element with the highest soil accumulation risk. The elements Cu and Zn have a dual character as essential plant micronutrients, changing to toxic effects when exceeding optimal soil concentrations, dependent on soil pH. These elements are only of minor concern from a food safety perspective as they are often phytotoxic at concentrations that pose little risk to human and animal health (López-Rayó et al., 2016). In contrast, Cd poses a health risk at concentrations below those causing phytotoxicity, as low levels of Cd can cause accumulation in the human kidney and liver (McLaughlin et al., 1999). Therefore, Cd belongs to the most important elements to consider in terms of food chain contamination (McLaughlin et al., 1999). The modeled results contradict several older studies that have shown accumulation of PTEs in soils after long-term application of high amounts of biosolids, increasing the risk of phytotoxicity or the accumulation of undesirable residues in plant biomass (Dai et al., 2006; McGrath, 1984; McGrath et al., 1995; Sekhar et al., 2002; Udom et al., 2004). However, the PTE contents of biosolids applied in these studies were much higher than is relevant for the current situation. In the last 3 decades,

there has been a dramatic reduction in the inputs of PTEs to the sewage systems due to implementation of environmental regulations, including a stricter separation of sewage from industry and households, and changing waste management technologies (Amlinger et al., 2004; Eriksen et al., 2009; Herter and Külling, 2003; Magid et al., 2006; Scherer, 2002; Schroder et al., 2008). The quality of biosolids produced has improved, with a 3–5-fold decline in the content of PTEs such as Cd, Hg, Pb, and Zn in biosolids since the late 1970s (Amlinger et al., 2004; Eriksen et al., 2009; Herter and Külling, 2003).

A general finding from our study is that the predicted changes in soil concentrations of PTEs after 200 years of application do not correlate with the PTE concentrations in RPFs (Tables 3 and 5). For example, composts have low to intermediate concentrations of Zn, Cu, or Cd in comparison to digestates or biosolids, but show a relatively high accumulation risk for these elements after long-term application. The reason is that RPF application rates are governed by the P concentration (and sometimes by the N concentration for RPFs with a N/P ratio > 5.0) so that materials with both low PTE and nutrient concentrations such as compost can result in a higher risk of PTE accumulation in the soil. In contrast, PTE accumulation risks from biosolids are lower than from compost although their PTE concentration is approximately 4–5 times higher, because at the same time the P concentration of biosolids is roughly 15 times higher than of composts (Table 2). Consequently, PTE flows are mainly a function of the P concentration in RPFs, and the ratio between the concentrations of PTE and P is the main driving factor influencing PTE flows applied to the soil.

The paradox is that many RPFs that are currently prohibited for use in organic farming (e.g., struvite and digestates containing catering wastes) have less harmful effects and environmental impacts than some already permitted inputs. The highest accumulation risk is predicted for green waste and urban organic waste composts, which are currently allowed in organic farming, while many less preferred (e.g., urban organic waste digestates, MBM) or even prohibited RPFs (e.g., biosolids and all sewage-derived RPFs) show a considerably lower soil PTE accumulation risk (Table 5). Therefore, the use of biosolids and sewage-derived RPFs will imply a considerable reduction of the risk of soil Cd pollution in comparison to already allowed composts and phosphate rock. The same applies to all other PTEs in comparison to composts.

The results also indicate that more sophisticated techniques to recover P from sewage like struvite precipitation or some of the thermal

approaches like the production of Rhenania ASH DEC[®], converter slags, or LeachPhos[©] are efficient in decreasing the soil PTE accumulation risk in comparison to the direct application of biosolids. For example, several studies have shown that struvite crystallization is an efficient process to increase P concentration and decrease the concentration of PTEs and other pollutants like hormones and pharmaceuticals (Ronteltap et al., 2007; Simons, 2008). During struvite crystallization 40%–80% of the PTEs (Simons, 2008) remained in solution. But also incineration combined with post-treatment of ashes is effective in reducing the specific PTE load per unit P.

Nevertheless, sustainable RPF application requires a risk-based management approach, as a high contamination risk for single batches still persists, requiring regular PTE monitoring even in the future.

5.2.3 Organic Contaminants

Many investigations on biosolids as potential source of POPs, including studies on compounds and their concentrations in biosolids have been carried out. Smith (2009a) notes that according to the European Commission there are no recorded cases of human, animal, or crop contamination due to the use of sludge on agricultural soils following the provisions of Directive 86/278/EEC. In Germany, in the early 1990s, a biosolid compensation fund (*Klärschlamm-schädigungsfonds*) was introduced to provide compensation for possible contamination and damage resulting from field application of biosolids. The only compensations paid since then have been for germination of tomato seeds, with no case of compensation for damages resulting from contamination of soil or crops with PTEs or POPs during this period. In contrast, composts as a potential source of POPs have received less attention, in spite of a few cases of serious soil contamination with POPs from urban organic waste compost application which were recorded in Europe in recent years. These were limited to perfluorinated surfactants and per- and polyfluorinated chemicals (e.g., Kehres, 2015). One explanation for the lack of detected cases of serious contamination with POPs from biosolids application may be related to the design of sewage treatment systems. Water-soluble contaminants having the highest risk for immediate effects on crops and the environment are either degraded during the treatment process (Heidler and Halden, 2008), and/or they are removed mainly by the cleaned water phase in the sewage treatment process (potentially affecting the surrounding water bodies), leaving a solid phase with low water-soluble concentrations of these contaminants, which may have the highest short-term risk due to the mobility in the environment. In contrast, for compost and

digestates, there is no process to remove organic contaminants prior to field application. Only nutrients and contaminants including POPs with a high adsorption strength to surfaces of the organic matter (e.g., P, several micro-nutrients, and several organic contaminants) are accumulated in the solid phase of the sewage treatment system. The high adsorption strength of many POPs and pharmaceuticals reduces the risk of transfer to other compartments (e.g., by leaching) and plant tissue after field application, however it also reduces their overall degradability.

A concentration effect as previously indicated for PTEs may also play a role. Specific inputs of some POPs like PAHs per unit P are higher with composts than with biosolids due to the low P concentration of composts (Kupper, 2008). The results of the IMPROVE-P risk assessment clearly indicated that the soil accumulation risk for the common organic pollutants PCB, PAH, and PCDD/F is negligible with the available RPFs (Weissengruber et al., n.d.).

Concerning contamination with pharmaceuticals, the risk may be higher from animal manures than from human wastes since the livestock sector uses much more pharmaceuticals than consumed by the human population. For example, in Germany in 2011 and 2012, the annual consumption of antibiotic pharmaceuticals accounted for approximately 2350–2400 Mg, out of which 65%–70% was used in animal husbandry (Vidaurre et al., 2017).

A comprehensive Norwegian study assessed the risk of exposure to soil organisms, the aquatic environment, grazing animals, animals eating feed based on plants from biosolid-fertilized soil, children eating soil, and humans consuming drinking water, crop plants, and/or meat for a broad range of xenobiotic organic contaminants and pharmaceuticals from using biosolids over a period of 100 years as a fertilizer and soil conditioner (Eriksen et al., 2009). All the assessed organic contaminants were found to constitute a low risk to the soil environment. Of the more than 1400 drug substances sold in Norway, only 14 were predicted to exceed cut-off values of 100 or $10 \mu\text{g kg}^{-1}$ soil after biosolid application, resulting in a low risk for soil organisms. The potential transfer to the aquatic environment of metals, organic contaminants, and drug substances from biosolids applied within the boundaries set by regulatory statutes was assessed to be of no significance (Eriksen et al., 2009). This is in agreement with conclusions of some other reports (e.g., Haynes et al., 2009; Smith, 2009a; Tavazzi et al., 2012). According to Haynes et al. (2009) and Smith (2009a), POPs in biosolids are affected by a number of processes that inhibit or prevent transfer to crops and the food chain in general, including: (i) rapid evaporation and loss to the

atmosphere for some substances, (ii) the rapid biodegradation and minimal or no persistence for others, (iii) strong adsorption or persistent connections to soil colloids, and (iv) minimal or no uptake by plants and grazing animals. Therefore, the risk to human health via dietary intake of POPs from crops grown on biosolid-treated soils was found to be minimal (Eriksen et al., 2009; Haynes et al., 2009; Smith, 2009a).

While substantial evidence exists on the risks associated with POP's, there is a need for continued vigilance in assessing the significance and implications of “emerging” POPs (i.e., chemicals that have come into production in more recent years) in biosolids and urban organic household wastes. It is necessary to support and ensure the long-term sustainability of these waste management options, as previously also stated for PTEs. Clarke and Smith (2011) reviewed available knowledge and identified research and monitoring priorities for emerging POPs based on the following five criteria: (1) environmental persistence in the soil environment (>6 months); (2) potential for human health impacts resulting from the land application of biosolids; (3) evidence or likelihood of bioaccumulation in humans or the environment; (4) evidence of ecotoxicity, and (5) the quality of empirical data and trends on the contaminant in biosolids internationally.

Another consideration is that for several POPs in biosolids and other waste streams, atmospheric deposition and environmental cycling are the main sources (e.g., PCBs), and consequently the concentrations of these historically used chemicals in biosolids generally correspond to background environmental levels (Kupper, 2008; Smith, 2009a).

Regarding pharmaceutical products, there has been particular concern about the development of antibiotic resistance in populations of bacteria. However, Riber et al. (2014) found that biosolids had only a transient impact on the antibiotic resistance profile of the soil community and that abundance of resistant isolates decreased rapidly with time after fertilizer application, while bacterial community structure at phylum level remained mostly unaffected.

This discussion has shown the challenges associated with assessing the POP risk from potential RPFs. Use of any recycled product will be accompanied by specific risks, sometimes relating to illegal deposition of contaminated material. Risk assessments should consider the specific risks of different provenances of materials since large differences in contamination levels and contamination patterns can occur, e.g., among rural and urban biosolids (Kupper, 2008; Tavazzi et al., 2012). The most practical way to reduce the risk is to exclude potential point sources from the sewage or other treatment systems (Haynes et al., 2009).

5.2.4 Indices to Assess the Loads of Contaminants

Usually the risk of soil accumulation of PTEs is related to their concentration in the fertilizer material on a DM base. The relationship between the PTE concentration in a RPF and the soil PTE accumulation risk are weak, and the P concentration is the main factor influencing the soil PTE accumulation risk (Section 5.2.2). The challenge is to develop more appropriate indices indicating the suitability of a specific material for field application, as our model calculations have shown that the current system for evaluating RPFs is not appropriate for quality assessments. The heavy metal-phosphorus index (HMP index) and the heavy metal-nutrient index (HMN index) developed in Switzerland by Herter and Külling (2001, 2003) are alternative tools for assessment of PTE loads. Both indices relate PTE load (weighted according to their respective potential environmental impact) to the fertilizer nutrient concentration. The HMP index uses only P whereas the HMN index uses N, P, and K of the assessed RPFs as a reference. The higher the values, the higher the PTE load relative to the potential fertilizer benefit of the RPF (Herter and Külling, 2001, 2003). In the present publication, the HMP index was calculated according to Herter and Külling (2001), but using elemental P concentration rather than P_2O_5 , and the HMN index was calculated according to modifications introduced by Möller and Schultheiß (2014) also including N and K concentrations in the formula:

$$\text{HMP-index} = ((wF \times \text{PTE}_1^2 + wF \times \text{PTE}_2^2 + \dots + wF \times \text{PTE}_7^2) / 7)^{-2} / P \quad (3)$$

$$\text{HMN-index} = ((wF \times \text{PTE}_1^2 + wF \times \text{PTE}_2^2 + \dots + wF \times \text{PTE}_7^2) / 7)^{-2} / (P + N + K) \quad (4)$$

where PTE_x is the concentration of the assessed PTEs ($\text{g Mg}^{-1} \text{ DM}$); $x = 1, 2, \dots, 7$ (seven included PTEs); wF is the weighting coefficient based on the relative toxicity of each element as follows: Cd: 37.09, Cr: 0.37, Cu: 0.31, Hg: 37.09, Ni: 2.32, Pb: 0.37, and Zn: 0.09; and P, N, and K are the concentration of the nutrients P, N, and K, respectively ($\text{kg Mg}^{-1} \text{ DM}$).

Large differences were found for the HMP and HMN indices among the assessed RPFs. The highest value for the HMP index was found for composts (Table 5). Moderate indices were found for sewage and biosolids, while for MBM and struvites, the index value was low. A strong correlation was found between the HMP index and the predicted mean relative increase in soil PTE concentration (Fig. 4), but no correlation between the HMN index and increase in soil PTE concentration (data not shown).

These results suggest that the HMP index provides a more balanced approach for assessment of the PTE loads in RPFs (Fig. 4). However, the HMP index is not suited for assessment of recycled fertilizers low in

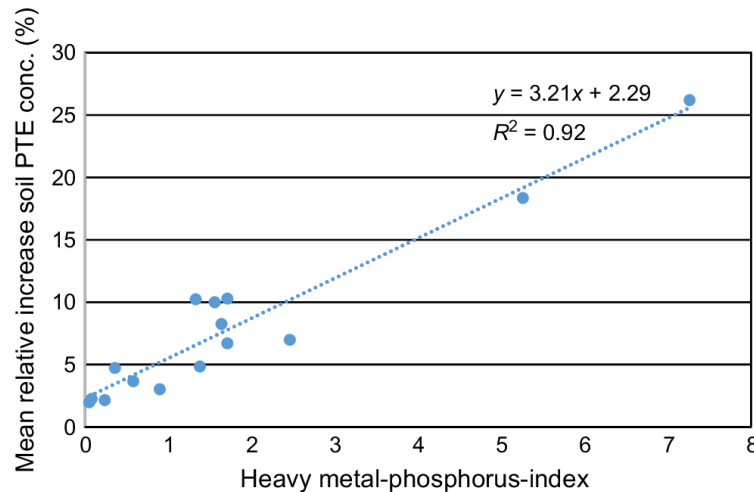


Fig. 4 Correlation between the heavy metal-phosphorus index and the mean relative increase in soil concentration of potentially toxic elements (PTE) at soil pH of 7.0 and yearly leaching of 100 kg of water m⁻² by 200 year application of recycled and control P fertilizers as modeled by [Weissengruber et al. \(n.d.\)](#).

P (e.g., keratins like horn meal and vinasse), and therefore not suited for assessment of N fertilizers.

The HMP or HMN indices are a significant improvement of former assessments based only on elemental concentrations. Even more advanced approaches for studying threshold values for PTE concentrations in RPFs may include regional factors, since the mobility and leaching of PTEs are mainly driven by soil pH and climatic water balance, which varies among regions.

5.3 Life Cycle Assessment

The data presented here are based on a change-based LCA approach. Therefore, the system boundaries start where a change of the state of the art seems to be feasible. E.g., collection of source-separated urban organic wastes has to be carried out independently from the later treatment approach. The analysis included acquisition of materials, e.g., chemicals for treatment, energy used for treatment and transport. Infrastructural processes, i.e., capital goods such as buildings and roads for transports were excluded. Five LCA impact categories were calculated for each of the P fertilizers and referred to the functional unit of 1 kg P. These were: abiotic resources depletion potential (ADP; calculated in kg antimony equivalents [Sb-eq]), primary energy demand (FED; calculated in mega joule [MJ]), global warming potential (GWP, calculated as CO₂-equivalents [CO₂-eq] with a time horizon of

100 years), acidification potential (AP; calculated as sulfur dioxide equivalents [$\text{SO}_2\text{-eq}$]), and eutrophication potential (EP; calculated as phosphate equivalents [PO_4^{3-}]). Credits were calculated for the N and K fertilizer value taking into account the long-term fertilizer value, for the long-term C sequestration/storage of the organic matter, and for the energy obtained during treatment, where applicable (for more details: [Hörtenhuber et al., 2017](#)).

The LCA indicates strong differences among feedstocks and treatment approaches. Whereas MBM results in low environmental impacts with small differences for the assessed treatment options ([Table 6](#)), strong differences were found for the treatment approaches for urban organic waste and wastewaters (composting, digestion, incineration, etc.).

For urban organic wastes, anaerobic digestion provided a more favorable environmental impact than composting in all assessed impact categories, even when composting is carried out under a controlled environment in order to reduce emissions ([Table 6](#)). Key factors influencing the differences are lower emissions by anaerobic digestion compared to composting, the additional energy gain saving fossil energies due to the produced biogas, and the higher N concentration and fertilizer value of digestates ([Hörtenhuber et al., 2017](#)). However, the potential soil C sequestration per unit P is higher for composts than for digestates. These results are in line with several other assessments of the best available technologies for management of urban organic household wastes from source-separated collection ([Funda et al., 2009](#); [Lampert et al., 2011](#); [Schott, 2012](#)). In addition to the poor N performance, composting causes direct emissions of GHG and ammonia (NH_3) ([Beck-Friis et al., 2000, 2001](#); [Brown et al., 2008](#); [Lampert et al., 2011](#)) as well as nutrient losses (N, S, and K) ([Eklind and Kirchmann, 2000](#)), along with some indirect effects on GHG emissions.

Among all options for recycling of P based on (dewatered) biosolids, the direct application of biosolids as organic amendment shows the best performance for all impact categories ([Table 6](#)). Key elements influencing the results of direct application of biosolids are related to savings of N and K fertilizer application, as well as from the carbon sequestration which can be achieved. Any treatment approach for selective recovery of P from biosolids either by chemical or by thermal approaches results in an increase in each of the impact categories addressed by the present LCA. These treatments increase the environmental impact by increasing the emissions due to higher energy consumption during treatment and by increasing the ADP due to the need for energy and chemicals, and by

Table 6 Net Life Cycle Assessment (LCA) Results for Different Approaches of P Recovery per kg Phosphorus (P)

	ADP (10^{-8} kg Sb-eq kg^{-1} P)	FED (10^1 MJ kg^{-1} P)	GWP (kg CO_2 - eq kg^{-1} P)	AP (10^{-2} kg SO_2 -eq kg^{-1} P)	EP (10^{-2} kg PO_4 -eq kg^{-1} P)
Organic household waste compost	−0.78	−8.10	76.5	46.5	13.8
Organic household waste compost— low emission	−1.07	−8.49	14.0	1.60	2.80
Organic household waste digestate	−136	−112	−102	−27.3	−36.1
biomass ash— untreated	−5.09	−7.48	−5.10	−10.5	−0.40
biomass ash— chemically solubilized	50.1	27.1	32.8	69.9	3.65
Meat and bone meal	−1.99	−4.54	−8.57	−1.61	−0.50
Digested	−6.16	−9.72	−7.34	−3.02	−1.28
Animal Feed	−19.2	−4.77	−9.40	−3.05	−1.07
Ash	−4.82	−37.6	−22.8	−7.01	−4.14
Ash—Solubilized	2.40	−27.8	−14.3	5.98	−2.21
Biosolids	−4.96	−6.63	−11.1	−3.20	−1.20
Ash	−2.06	−3.48	1.53	0.10	−0.30
Rhenania ASH DEC [®]	3.46	−0.14	3.80	1.80	1.15
Ash LeachPhos	6.52	0.26	7.85	11.2	1.41
MEPHREC converter slag	17.5	21.9	11.4	3.91	4.27
Struvite AirPrex	19.8	12.3	10.0	2.60	5.00
Struvite Stuttgart Sludge Leaching	54.6	29.4	15.7	24.6	4.20
PR	596	0.67	0.50	0.30	0.20
TSP	607	3.96	3.23	7.20	10.1

Abbreviations: ADP, abiotic depletion potential; GWP, global warming potential; FED, fossil energy demand; AP, acidification potential; EP, eutrophication potential.

reducing the fertilizer value and the organic matter content of the final fertilizer product thereby reducing the potential for soil C sequestration. Drying and incineration of dewatered biosolids are relatively neutral in terms of the direct effects on the five impact categories, while reducing the benefits from biosolids application like N fertilization and soil C sequestration. However, any approach to solubilize P from ashes in order to obtain a RPF with a higher plant P availability is related to a strong increase of the environmental impact across all categories. The highest impact results from the Stuttgart Sewage leaching process to precipitate P as struvite, while the environmental impact of the AirPrex[®] process is rather low, as no acids are used for P solubilization, but the process leads to low P recovery rates (Kabbe et al., 2015).

The results of the LCA assessments show that rather simple approaches to treat recoverable P sources provides a more favorable environmental impact in all five impact categories than more sophisticated ones, e.g., for sewage decontamination. Acids are used both to solubilize P from ashes and from sewage prior to struvite crystallization to increase P recovery (Antakyali et al., 2013; Çelen et al., 2007; Yilmazel and Demirer, 2013). This strongly affects the inputs, the costs, the fertilizer value, the effects on soil fertility (e.g., salinity effect and soil acidification), and the environmental performance of the different technologies and products (Table 6) as also found by Nakakubo et al. (2012) and Bradford-Hartke et al. (2015). Also Linderholm et al. (2012) as well as Kalmykova et al. (2015) stated that regarding energy consumption and GHG emissions, direct application of biosolids on farmland is a better option than P recovery from biosolids ash. The selective P removal and the decontamination of sewage P considerably increases the environmental impact, as several of the involved processes consume large amounts of energy and other abiotic resources (Kabbe et al., 2015). The results also indicate that solubilization of any kind of biomass ashes results in stronger environmental impacts than the solubilization of phosphate rock in order to obtain a water-soluble P fertilizer, as shown by the LCA. The P concentration in phosphate rock is much higher, and therefore, the consumption of abiotic resources (e.g., inputs of chemicals) is lower per unit P recovered.

As already stated, MBMs do not have any P fertilizer value in soils with $\text{pH} > 6.0$. The LCA results indicate a more favorable environmental performance for incineration of (already dried) MBM with subsequent chemical P solubilization in order to produce a plant available P fertilizer rather than the direct application of dried MBM. Incineration reduces the credits for the

N fertilizer value, however MBM has a high energy content. The use as animal feed is also an environmental friendly recycling option (Table 6).

The LCA assessment of urban organic wastes as well as MBM shows that the production of byproducts providing additional revenue can have a major influence on the overall performance of a treatment attempt, demonstrating that credits for coproducts may have a considerable influence on the environmental performance of the process. Important benefits from coproducts include credits for the production of energy, for soil C sequestration, for the N fertilizer value, or for use as animal feed (for more details: Hörtenhuber et al., 2017). Therefore, the challenge is to develop treatment approaches that result in win–win relationships by reducing the inputs of nonrenewable resources during treatment, and addressing not only P recovery, but also the recovery of other elements in the waste stream (e.g., N and K), e.g., by redesigning the sanitation system to allow for a more complete recovery of nutrients and energy as suggested by Magid et al. (2006). Nitrogen compounds are a key source for GHG emissions from organic amendment management; therefore, a key factor for implementation of approaches with lower environmental impacts might be the selective removal of N as NH_4^+ by stripping NH_4^+ salts for fertilizers. By developing a biorefinery approach, valuable products from the organic matter can be obtained gaining higher credits, e.g., by production of high value raw materials from the organic matter. This can reduce the environmental impact of the process when the benefits from the N fertilizer byproduct are included in the assessment.



6. COMPARATIVE MCA

Obtaining and classifying data of key characteristics such as P recovery rates, rPeff, loads of contaminants, and LCA performance on the most promising technologies for treatment of recycled P sources is crucial. Experience shows that the environmental impact and the potential effects on soil fertility are the most important criteria for decision makers. A MCA was used in this study to provide a comprehensive overview of the implications of different treatment options for recycling of P. The MCA was performed by scoring the RPFs based on a range of criteria related to fertilizer value (nutrient recovery rates, P fertilizer value, and organic matter value), impacts of processing (valuable coproducts, energy demand, GHG emissions, abiotic resource depletion, eutrophication, and acidification), and environmental and health risks from PTEs and POPs. All criteria were scored between

-- and ++ with low scores for negative and high scores for positive impacts. The assessment was conducted by the authors of the paper based on the information in this paper.

The present compilation shows that most approaches to P recycling have benefits and risks, resulting in contradictory rankings of recycling approaches and sources for each impact category (Table 7). For example, biologically precipitated biosolids have a high P fertilizer effectiveness (Fig. 2), add organic matter to the soil and contain other nutrients like N, K, and S (Table 7), and have a low risk of PTE accumulation in the soil. However, biosolids may also contain organic pollutants, which may affect soil fertility and food security. Ashes obtained from biosolid incineration are almost free of organic pollutants, but incineration downgrades the material from a soil fertility perspective by reducing the plant P availability (Fig. 2), the recovery rates of other nutrients (N and S), and the organic matter inputs to the soil. Chemical treatment of ashes for production of a plant available P fertilizer is related to a strong increase in the use of abiotic resources and higher GHG emissions (Table 6).

The present compilation of indicators for an evaluation of methods to improve P recycling shows that any feedstock or approach for treatment has both strengths and drawbacks, resulting in contradictory rankings of recycling approaches and sources for each impact category (Table 7). The high scores calculated for the direct application of biosolids shows the relevant advantages of their use in agriculture, and the strong shortcomings related to more sophisticated approaches of P recovery.

The decision not to use biosolids or a substantial part of MBM in organic farming reflects perceptions of risk biased toward the current generation, with less emphasis on the likely impacts for future generations of not utilizing these RPFs such as increased GHG emissions and depletion of abiotic resources. A balanced implementation of the precautionary principle to protect both the current and future generations means that any management approach should aim for almost complete recycling of the nutrients in society combined with the lowest GHG emissions and abiotic resource use. The challenge is to find a balance by applying the “precautionary principle” to minimize risks to humankind in the short term (e.g., organic pollutants, pharmaceutical residues, and pathogens) and those affecting humanity in the long term (e.g., PTE accumulation, abiotic resource scarcity, and climate change). In practice, we will need to find short-term smart solutions for any potential P source for an almost complete recycling of the recoverable resources (including other elements), while reducing potential GHG

Table 7 Comparative Evaluation of the Assessed Options for Improved Phosphorus (P) Recycling Based on a Multicriteria Assessment													
P Recovery Rates	P Fertilizer Value	Recovery Rates for Other Nutrients (e.g., Nitrogen, Potassium)			Potential for Recycling of Stable Organic Matter			Production of Valuable by- and coproducts			Risk of Negative Impacts by Organic Contaminants		
		P	Fertilizer Value	Nutrients (e.g., Nitrogen, Potassium)	Recycling of Stable Organic Matter	Energy Demand	Greenhouse Gas Emissions	Abiotic Resource Depletion Potential	Eutrophication and Acidification Potential	Accumulation of Potentially Toxic Elements	Risk of Negative Impacts by Organic Contaminants		
Biomass ashes: low overall P recycling potential													
Biomass ashes	++	--	--	--	--	--	--	--	++	--	++	++	
Solubilized biomass ashes	++	+	--	--	++	++	++	++	--	++	++	++	
Urban organic wastes: intermediate overall P recycling potential													
Green waste compost	++	++	--	++	++	--	-- to +	--	--	--	--	--	
Household waste compost	++	++	--	++	++	--	-- to +	--	--	--	--	--	
Household waste digestate	++	+	++	++	++	--	++	++	++	+	--	--	
Slaughterhouse wastes: intermediate overall P recycling potential													
Meat and bone meal (MBM)	++	--	++	--	++	++	++	+	++	++	++	++	
MBM digested	++	--	++	--	++	++	++	+	++	++	++	++	
MBM ash	++	--	--	--	++	++	++	+	++	++	++	++	

Solubilized MBM ash	+	+	--	--	--	--	--	--	--	--	--	++	++	++
Sewage sludge-based recycled P fertilizers: high overall P recycling potential														
(dewatered) biosolids (SS)	++	++	++	++	++	++	++	++	++	++	++	++	--	--
Untreated SS ashes (SSA)	++	--	--	--	--	--	++	++	++	++	++	--	++	++
Solubilized SSA (LeachPhos®)	+	+	--	--	--	--	--	--	--	--	--	++	++	++
Mg-treated SSA ASH DEC®	++	--	--	--	--	--	+	+	+	+	+	+	++	++
Rhenania ASH DEC®	++	+	--	--	--	--	+	+	+	+	+	+	++	++
Mcphrec-slugs	+	+	--	--	--	--	--	--	--	--	--	+	++	++
Struvite	--	++	--	--	--	--	--	--	--	--	--	to	++	++
Ca-phosphates	--	--	--	--	--	--	--	--	--	--	--	++	++	++

For: --, very low performance or very high negative environmental impact; -, low performance or high to neutral negative environmental impact; +, high performance or neutral to low positive environmental impact; ++, very high performance or positive environmental impact (e.g., for credits).

emissions and other negative impacts on the environment. This is in line with the fundamental principles of organic farming.



7. RESEARCH NEEDS

Many papers have addressed the potential consequences of organic pollutants on soil contamination. However, there is a lack of long-term assessments of organic contaminants on key soil parameters including soil biodiversity, key soil functions, and possible transfer of pollutants to the harvested products and associated impacts on human health. The development of target and nontarget screening methods and strategies for organic contaminants is probably the major methodological challenge in order to get tools for an early assessment of potential risks related to nutrient recycling. Furthermore, we need new methods to assess the risk from PTEs in order to obtain an index or value which represents the real risk by integrating relative toxicity of elements, likely application rates based on nutrient contents, and local environmental conditions impacting on elemental retention and reactivity in the soil. One major challenge is the promotion of the use of RPFs in organic farming without compromising the image of organic farming or consumer trust. This requires the inclusion of social scientists in research projects, with skills and approaches to assess attitudes and factors that determine behavior.

Research into more efficient and environmentally friendly technologies for P recovery from urban organic wastes, and how this recovery can be achieved with an added value, is also a major challenge. For the organic sector, biological approaches for P recovery are more accepted than thermal or chemical approaches. Furthermore, we need cost-effective methods for the assessment of the expected relative P fertilizer effectiveness.



8. CONCLUSIONS

This review has shown that the highest nutrient recovery rates from societal waste streams including P are achieved by rather simple approaches to P recycling, whereas more sophisticated approaches are often related to lower P recovery rates and losses of organic matter and plant nutrients. Plant P availability (indicated as rP_{eff}) of many RPFs is higher than that of phosphate rock. Thus, if organic farmers need external P inputs there are alternatives to phosphate rock. For neutral to alkaline soils, untreated ashes and MBMs are not recommended; in this case, compost, digestates, biosolids, calcined ashes treated with Na or K, as well as converter slags and struvite

are more suitable. Acid soils are less challenging for the choice of the appropriate RPF, as the acid environment solubilizes nonreactive Ca-phosphates and transform P into reactive forms (e.g., hydroxyapatites in MBM and ashes).

The results have also indicated that present generations are faced with the dilemma of balancing the needs of present and future generations, which requires an acceptance of some additional risks to our generation for the benefit of future generations. A balanced assessment of RPFs should consider a broad range of aspects in order to balance short- and long-term impacts from production and use of the RPF. Basic preconditions of any P recycling are high recovery rates. Therefore, a reliable and holistic concept for improved P recycling should offer treatment and recycling options for each of the main potential P societal sources. The organic sector should carefully consider whether it is justified to select only a few resources (e.g., green waste compost and source-separate household waste), leaving the responsibility to recycle other P sources to the conventional sector. Furthermore, recycling approaches should allow the production of RPFs with a high P fertilizer value without harming long-term soil fertility and food safety. The overall goal should be to enable organic farmers to achieve balanced P budgets by using regionally available recycled P sources, which may require significant changes in organic regulations. Regarding the flows of PTEs, the consensus view is that there appears to be no scientific rationale for including numerical limits for contaminant concentrations in quality assurance systems for agricultural use of any kind of RPF based on DM or ash content. PTEs in soil should remain limited, but in view of the huge differences in the nutrient concentrations of RPFs, and the overall low risk of accumulation with the present materials, threshold values for contaminants should be defined based on the nutrient concentration instead of DM. A complete risk assessment for organic pollutants cannot be made, however a considerable body of evidence indicates that recycling of sewage on farmland is not constrained by concentrations of PTEs or known organic contaminants found in contemporary biosolids. There is a need to continue examining the impact of emerging contaminants, but overall, the input of contaminants into waste streams should be reduced at the source, i.e., during the production phase, or by source separation.

The present review shows that any approach to improved P recycling has strengths as well as drawbacks, resulting in contradictory rankings for different impact categories. Therefore, any strategy for nutrient recycling is a navigation between constraints. A stronger consideration of RPFs within these constraints will increase the resilience and the credibility of organic farming.

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6 General Discussion

6.1 Agronomic efficacy of P fertilizers recycled from sewage sludge

In organic farming systems, a sufficient P supply of plants becomes increasingly challenging, since the variety of allowed P fertilizers is limited and fertilizer efficacy of PR or bone meal is low on many soils. The research presented here shows that most of the investigated recycled fertilizers from sewage sludge have a higher P bioavailability than PR (Möller et al., 2018; Wollmann et al., 2018). Among the tested fertilizers, struvite had the highest P efficacy, increasing the P offtake of maize by 27.5% in the field experiment, and more than sixfold in the pot experiment, compared to the unfertilized control (Wollmann and Möller, 2018; Wollmann et al., 2018). These results confirm findings in literature (Cabeza et al., 2011; Robles-Aguilar et al., 2018) and the analysis of compiled data (Möller et al., 2018). However, the range of different P fertilizers recycled from sewage sludge can be characterized by large differences regarding the reactivity of their P forms, as a consequence of e.g. the quality characteristics of their original feedstock and their specific production conditions, which mainly determine the chemical composition of the final product and, thus, the fertilizer efficacy under given conditions. For example, the composition and purity of precipitates seems to be much dependent from production conditions like pH and the presence of Ca, Al and ferric ions in the precipitation tank (Acelas et al., 2015; Darwish et al., 2016) as well as the Ca/Mg molar ratio in the initial solution (Yan and Shih, 2016). Therefore, any approach for the production of effective P fertilizers from sewage sludge starts at the sewer plant and the used techniques for P recovery. Different treatment techniques are available for the removal of dissolved P from waste water. A chemical, precipitative

removal of P is usually achieved through the addition of di- or trivalent metal salts of Fe, Al or Ca, which are coagulated to form metal phosphates that can be removed after flocculation and settlement (Melia et al., 2017). This treatment generates sludge that is often unsuitable for direct reuse due to not only the formation of insoluble precipitates with P, resulting in a reduced bioavailability (Möller et al., 2018), but also through a possible incorporation of contaminants in the precipitate, such as arsenic, pathogens and viruses (Mehta et al., 2015; Melia et al., 2017). In contrast to this, the widespread technology of enhanced biological P removal (EBPR, or “bio-P”), based on polyphosphate accumulating organisms, seems to be more suitable. Little or no chemical additions are required (Melia et al., 2017), the P bioavailability of the sludge itself is higher compared to that from chemical P removal (Möller et al., 2018) and a process stream of suitable concentration for a direct and efficient P recovery as struvite is provided (Pastor et al., 2008; Melia et al., 2017). In addition, the fertilizer efficacy of P fertilizers recycled from sewage sludge or sewage sludge ash might be influenced by the different types of P removal during sewage treatment. Thus, further research is needed in this field.

Fertilizer efficacy of thermally recycled P fertilizers is usually rather low. The bioavailability of P in chars, pyrolysis coals and slags is also dependent on their particular production conditions and the used feedstock (Nanzer et al., 2014a). Phosphorus solubility decreases with increasing production temperature (Christel et al., 2014; Bruun et al., 2017) due to the formation of water-insoluble compounds such as hydroxyapatite or stable aromatic structures (Thygesen et al., 2011).

Apparently, the fertilizer efficacy of P recycled from sewage sludge is mostly determined by two main factors; the chemical form of P contained in the fertilizer, and the soil pH. Based on this assumption, a rough classification of recycled fertilizers can be made into two groups; first, fertilizers with high P bioavailability independently from soil pH, due to reactive P forms (e.g. Struvite or treated (calcined) ashes, such as Na-SSA) (Möller et al., 2018; Robles-Aguilar et al., 2018) and, second, fertilizers comprising highly crystalline P molecules of low bioavailability at soils with pH > 6.0, such as untreated incineration ashes (Möller et al., 2018). In dependency of their composition, there are various recycled fertilizers somewhere in between those two groups. For example, some precipitates like struvite AirPrex® or P-RoC® can contain different amounts of less soluble Ca-, Al- or Fe-phosphates as co-precipitates which reduce their plant availability in soil. The Mg-SSA showed a similar P efficacy like PR at soil pH 7.2 in pot experiments (Wollmann et al., 2018) and pH 6.5 in the field experiment (Wollmann and Möller, 2018) and generally a decreasing fertilizer efficacy with increasing soil pH (Möller et al., 2018). Both Mg-SSA and PR even decreased shoot P offtake in some experiments (Wollmann and Möller, 2018; Wollmann et al., 2018).

In light of these findings, the use of pure struvite and calcined Na-SSA can be recommended for their application in agriculture to improve P supply of plants, independently from the soil pH. Moderate to high fertilizer efficacy has been shown also for Mephrec slags, even at soil pH 6.6 (Cabeza et al., 2011), due to a certain proportion of reactive silico-phosphates (Scheidig et al., 2011). Concerning Mg-SSA, P-RoC® and pyrolysis coal, P efficacy increases with decreasing soil pH (Cabeza et

al., 2011; Nanzer et al., 2014a). However, common in European agricultural regions are neutral and alkaline soils (Nesme et al., 2014) on which the use of SSA, P-RoC® or pyrolysis coal is rather ineffective. Appropriate treatment is thus needed for those fertilizers before they might be applied to increase plant P supply on neutral or alkaline soils. Otherwise, they can be used as source material for fertilizer production.

In the field experiment, the response to fertilizer application was poor (Wollmann and Möller, 2018). This is a common outcome from many field experiments (e.g. Gallet et al., 2003), caused by high soil P levels and the complex P turnover processes in soil. On the specific trial, a high amount of organic soil P can be assumed due to a high share of legumes in the crop rotation. Thus, a certain amount of mineralized organic soil P might have contributed to the P supply of maize plants, which supports findings of other authors (Mäder et al., 2002; Dodd and Sharpley, 2015). A weak point of the field experiment possibly occurred by not including MCP as reference fertilizer, which is essential in further experiments, in order to evaluate the overall response of crops to fertilizer applications. The general fertilizer efficacy of recycled P fertilizers under field conditions cannot reliably be evaluated from the presented data. This leaves a gap for future research that should be closed by investigating recycled P fertilizers in field experiments on highly P deficient soils that stronger respond to P fertilization in general.

For the first time, the agronomic efficacy of recycled fertilizers was evaluated for several fields of a crop rotation, under controlled conditions including different crops, and on a field scale with maize (Wollmann and Möller, 2018; Wollmann et al., 2018).

An immobilization of fertilizer P over time could be shown in all experiments. Thus, in the field, effects of P fertilization were measured only in the year of fertilizer application and not in the year thereafter (Wollmann and Möller, 2018). In further experiments, the sorption capacity of the test soils should be determined, in order to assess their P immobilization potential. In both pot experiments, differences among tested fertilizers were vanished during clover cultivation (from first to third clover harvest) and appeared again for the following maize (Wollmann et al., 2018). This indicates either a mobilization of fertilizer P by clover roots in excess of their own need for the subsequent maize, or an improved P supply of maize plants through internal P cycling via decomposed clover roots (Nuruzzaman et al., 2005b). To clarify the underlying processes, the root biomass of clover should be determined in following experiments, as well as forms of residual P in soil after fertilizer applications. Beyond that, it can be concluded that also P from recycled fertilizers is immobilized in soil, and normally little effects through residual fertilizer P can be expected for subsequent crops in the rotation (Wollmann and Möller, 2018). Thus, recycled fertilizers should be applied to responsive crops in the crop rotation.

6.2 Biological means of P mobilization to improve plant P acquisition from recycled fertilizers

There are different biological approaches to optimize P efficacy of fertilizers. A soil application of rhizobacteria could play a key role in enhancing P availability of less labile P forms, such as those obtained in different P fertilizers recycled from sewage sludge. In some soils, P availability might also be enhanced through a rhizosphere

acidification, resulting from a proton release during biological N₂-fixation of legumes. In the research presented here, it could be shown that the bioavailability of P fertilizers recycled from sewage sludge can be enhanced by the cultivation of red clover in a crop rotation with maize. This effect has been shown to be dedicated to the biological nitrogen fixation (BNF) ability of red clover and the related decrease in soil pH (Wollmann and Möller, submitted). Of the applied ashes, 5.57% (Mg-SSA) and 11.6% (Na-SSA) of P was additionally taken up by maize when grown after clover with BNF, compared to clover receiving mineral N fertilization (Wollmann and Möller, submitted). This corresponds to 15.7% (Mg-SSA) and 50.3% (Na-SSA) of the remaining fertilizer P after clover cultivation. One direct effect of a decreased soil pH to P mobilization is assumed through solubilization of sparingly soluble P forms contained in recycled fertilizers, such as Mg-SSA (Wollmann and Möller, submitted). Furthermore, it is possible that indirect effects following the decrease in pH contributed to an improved P supply of maize after clover cultivation. For example, an increasing activity of acid phosphatase in the rhizosphere of maize has been observed with decreasing soil pH, leading to an improved P uptake of maize from phytin mineralization (Ding et al., 2011). It is also conceivable that an internal P cycling through decomposed clover roots led to an improved P supply of maize. This residual P after clover cultivation might have also been visible in the field experiment (Wollmann and Möller, 2018), explaining the overall low response to fertilizer applications. For a clear identification of the P mobilization mechanisms by clover with BNF, more experimental data is required from future research on red clover with and without BNF, including the investigation on the implications of these different growing systems on rhizosphere processes. In addition, the Ca-demand of a N-fixing clover plant compared to a clover plant with mineral N

supply should be determined, which might influence the P mobilization in soil. Apart from that, it appears that P fertilizers recycled from sewage sludge might be better suitable for agricultural systems including legumes in the crop rotation than systems without legumes.

It has to be noted, that a mobilization of P from soil through clover with BNF did not take place to the same extent as the mobilization of fertilizer P. In fact, the mobilization decreased from Na-SSA > MCP > PR > Mg-SSA > Unfertilized (soil P), more or less in the same order that also describes plant P availability of the respective fertilizers. Thus, it might be, that the P mobilization potential of red clover is very dependent on the specific P source. This assumption has to be clarified in further research. In this light, also the conclusion that fertilizer P should be applied to responsive crops in the rotation (e.g. red clover) can be confirmed, but has to be expanded: as not the entire P demand of the subsequent maize could be covered by the P mobilized by red clover, a fertilizer P application to maize is still necessary to secure an optimum plant growth. Hence, further research should address the question whether or not the overall amount of fertilizer P in a crop rotation can be reduced by splitting the application between red clover and maize, without diminishing crop yields. It also remains to be clarified whether a (splitted) P supply with recycled fertilizers in a crop rotation is sufficient to maintain crop yields, or if a combination of recycled P fertilizers with highly bioavailable P fertilizers (like struvite or water soluble P fertilizers) is necessary. This approach might be most interesting for recycled fertilizers of low bioavailability, such as Mg-SSA, which could be combined with MCP or even Struvite or Na-SSA. When P supply is

covered by recycled fertilizers with high bioavailability (such as Na-SSA or Struvite), most likely, additional applications of reactive P fertilizers are not necessary.

Some authors have described a P mobilization ability of *Pseudomonas* and *Bacillus* strains. In growth media, a P mobilization effect of different bacteria strains has been reported (Nkebiwe, 2016; Meyer et al., 2017). In the present research, it could be shown that a transfer of this beneficial effect to soil under controlled conditions or field conditions is very limited (Wollmann and Möller, 2018; Wollmann et al., 2018). In the pot experiments, the overall increase in shoot P uptake ranged between < 1% (considering the sum of clover, maize and ryegrass plants with both *Pseudomonas* or *Bacillus* applications) and 10.1% (with maize and *Bacillus* application) (Wollmann et al., 2018). The results on the P mobilization potential of bioeffectors are ambiguous, which supports findings of other authors (Lekfeldt et al., 2016; Thonar et al., 2017). The only significant P mobilization effect was shown for the *Pseudomonas* strain Proradix® in the field experiment when combined with PR fertilization, resulting in an increased shoot P offtake of maize by 49.4% compared to PR without *Pseudomonas* application. This corresponds to an increase of 10.9% compared to the unfertilized treatment, with or without *Pseudomonas* application (Wollmann and Möller, 2018). A similar effect has been reported by Kaur and Reddy (2014). However, the underlying mechanisms of this stimulatory effect remain unclear. It has been suggested that PR constitutes a proliferation substrate for the applied inoculants (Kaur and Reddy, 2014). In this light, one might assume also promoting effects of the *Pseudomonas* strain when combined with ashes, which contain apatite-like compounds (Nanzer, 2012), comparable to PR. This could not be confirmed in the present experiments (Wollmann

and Möller, 2018) and other studies from literature (Lekfeldt et al., 2016; Meyer et al., 2017). It is also not clear why there were no results obtained in the pot experiments using the exact same inoculant, PR fertilizer and test plant, especially since plant growth in the pot experiments has clearly been limited by P supply (Wollmann et al., 2018). It could be shown in both pot experiments and in the two-year field experiment, that there were no medium- or long-term effects of bioeffector application on plant growth and P uptake of different test plants either (Wollmann and Möller, 2018; Wollmann et al., 2018). An increased P mobilization has recently been reported for maize, when inoculated with either *Pseudomonas* or *Bacillus* strains, and combined with different organic P fertilizers in various pot experiments (Thonar et al., 2017; Li et al., 2018). The presented results contradict those findings on a field scale for the *Pseudomonas* strain Proradix® in combination with compost fertilization (Wollmann and Möller, 2018). It is suggested for further experiments and interpretation of results, to prove a rhizosphere colonization of applied bacteria in soil, before their mode of action might be investigated in a second step.

The low reproducibility of plant growth promoting effects due to bacterial soil inoculation demonstrates their strong dependency on biotic and abiotic conditions (Thonar et al., 2017) and their overall weak and unreliable effect on growth promotion. To overcome the consequent fragility of promotional effects following bacteria inoculation, an application of consortia of different bacteria has been suggested by Nkebiwe (2016). Natural selection might take place, where only bacteria survive and colonize the rhizosphere that are best adapted to the environmental conditions (Nkebiwe, 2016). Yet, considering the presented results (Wollmann and Möller, 2018;

Wollmann et al., 2018) and those from other studies (Richardson, 2001; Halpern et al., 2015; Lekfeldt et al., 2016; Meyer et al., 2017), it can be concluded that, to date, the ability of inoculated bacteria strains to increase P mobilization in soil is rather poor. Clearly more research is needed in this field to identify conditions for an effective promotion of P supply by bacterial inoculants in agricultural crop production. It should also be considered that the microbial community in soil is adapted to the given environmental conditions and able to outcompete applied bacteria. Thus, an application of bioeffectors cannot be recommended, especially under organic crop production, given the fact that those farming systems usually are characterized by a high intrinsic microbial activity (Mäder et al., 2002). Reduced microbial biomass and microbial activity has been reported for cropping systems that rely on intensive soil tillage (Wang et al., 2011). Future research on applied bioeffectors should thus focus on soils with somehow disturbed or decreased microbial activity, such as e.g. those of intensive agriculture systems rather than those following typical organic farming practices.

6.3 Recycled P fertilizers in organic crop production

From an agronomic point of view, the presented data clearly demonstrate a certain benefit of using P fertilizers recycled from sewage sludge in organic agriculture, since the fertilizer efficacy of most recycled fertilizers is higher than that of PR (Möller et al., 2018; Wollmann and Möller, 2018; Wollmann et al., 2018). Our data not only show poor solubility of PR, but even an adverse effect of PR application on P bioavailability in both pot and field experiments (Wollmann and Möller, 2018; Wollmann et al., 2018), which might be caused by an increase of soil pH after PR application (Sinclair et al.,

1993). This once more emphasizes the need of bioavailable P fertilizers for organic crop production. The use of the non-renewable PR is often not effective, and certainly not sustainable, a characteristic claimed for by the principles of organic farming. Concerns have been raised that a restriction of nutrient recycling from society to the organic farming sector might impose substantial limitations on yields in many contemporary organic farming systems (Løes et al., 2017). The growing yield differences between organic and conventional systems (Seufert et al., 2012; Niggli et al., 2015), and the incapacity of supplying the growing demand of organically produced products has been identified as one of the major problems of organic systems (IFOAM, 2016). This might be attributed to the slow innovation in the organic sector (Niggli et al., 2015).

The concept of nutrient recovery and recycling is basically in line with fundamental organic ideas, such as the principle of ecology “Organic Agriculture should be based on living ecological systems and cycles, work with them, emulate them and help sustain them” (IFOAM, 2017). The aim of the precautionary principle of care in organic agriculture is a production of high quality products with a minimum level of contaminants such as pharmaceuticals, pathogens, and potentially toxic elements (Løes et al., 2017). The concentration of PTE in recycled P fertilizers from chemical approaches of P recovery (e.g. chemical precipitation) is rather low (Ronteltap et al., 2007; Antonini et al., 2012) while thermal approaches for P recovery from wastewaters (e.g. incineration) are less efficient in PTE removal (Ronteltap et al., 2007; Antonini et al., 2012; Egle et al., 2016). Thus, a reduction of PTE already during waste water treatment might be an important approach in future to overall reduce PTE in recycled

fertilizers such as chars and ashes. This might possibly be influenced by the type of P removal and added chemicals during wastewater treatment, an assumption that has to be clarified in further research. Thermally recycled P fertilizers, such as ashes and chars, are generally free from most organic pollutants and pharmaceuticals that are eliminated during the thermal production process (Egle et al., 2016). By thermochemical treatment of SSA, PTE are removed by evaporation of metal chlorines, fulfilling the quality parameters for a P fertilizer in Germany (Adam et al., 2009; Nanzer et al., 2014b; Stemmann et al., 2015; Herzel et al., 2016). All recycled P fertilizers investigated in the present study contained by far lower PTE compared to the commercially available, and in organic farming permitted fertilizers PR and urban organic household waste compost (Möller et al., 2018). For compost application, there is an even higher PTE pollution risk compared to struvites, ashes and sewage sludge, due to its high heavy metal-phosphorus ratio (Weissengruber et al., 2018). Based on these findings, a serious soil contamination with PTE through the use of these recycled P fertilizers in agriculture is not likely (Magid, 2012; López-Rayó et al., 2016; Weissengruber et al., 2018). Similar results have been reported for organic micropollutants contained in precipitated products, such as struvite. More than 98% of organic pollutants and pharmaceuticals remain in the precipitation solution and are not transferred to the struvite, and up to 20% of remaining pollutants can subsequently be removed through washing (Ronteltap et al., 2007; Antakyal et al., 2011). In this light, in general, a prohibition of recycled P fertilizers from sewage sludge in organic farming, while permitting e.g. the use of compost as fertilizer, seems to be not justified. Furthermore, the acceptance of recycled P fertilizers among stakeholders of the organic sector seems generally high, as long as means are taken to ensure their

sufficient purity, safety and environmental efficiency (Løes, 2016; Seidl et al., 2016). Obviously, a specific evaluation of potential risks has to be conducted for an application of each recycled fertilizer product, and constant quality standards of the fertilizers need to be assured before they could be permitted in organic farming regulations.

According to EU regulations, mineral fertilizers must be of low solubility (EC 889/2008), which matches the presented results on the solubility characteristics of P fertilizers recycled from sewage sludge (Wollmann et al., 2018). Generally, the efficacy of P fertilizers in Germany is described by their amount of water soluble P, assuming an increased fertilizer efficacy with increasing amounts of water soluble P. This assumption might apply for water soluble fertilizers such as MCP when used on calcareous soils, but not universally for recycled fertilizers from sewage sludge, as has been shown in the data presented here (Wollmann et al., 2018) and in literature (Cabeza et al., 2011; Brod et al., 2015; Meyer et al., 2018). Of the total P contained in struvite, for example, only 1-2% is soluble in water (Kern and Heinzmann, 2008; Wollmann et al., 2018), while bioavailability of struvites is very high (Johnston and Richards, 2004; Cabeza et al., 2011; Gell et al., 2011; Vogel et al., 2015; Løes et al., 2017; Wollmann et al., 2018). Thus, for an implementation of novel P fertilizers into agricultural production, in general, it might be necessary to turn away from traditional description methods of P fertilizers, such as their solubility in water. Instead, an implementation of other, more differentiated description methods should be launched to describe and examine their solubility and bioavailability. A new, more mechanistic soil P test has even been suggested for organic systems to identify the crop response to supplied P fertilizers, assuming enhanced microbial and other biological processes

in organic farming systems (Nesme et al., 2014). Hence, for an implementation of recycled fertilizers into organic farming regulation bodies, there are still major obstacles to overcome. Nevertheless, P fertilizers recycled from sewage sludge have been shown to be a suitable alternative for organic crop production in many ways. By an implementation of those fertilizers, organic agriculture would, once more, be pioneering the way towards a farming system of the future, which inevitably will comprise a closed P cycle between urban and rural areas.

7 Conclusion

The different P fertilizers recycled from sewage sludge hugely differ in their production processes. This results in different forms of chemical P bonding which determines their P availability to plants. Nevertheless, all tested recycled fertilizers but Mg-SSA had a higher P bioavailability than PR and, thus, can be considered as alternative fertilizers for organic crop production. Concerning the agronomic efficacy, only few of the recycled fertilizers (struvite and Na-SSA) can be recommended independently from soil pH. Other fertilizers, such as Mg-SSA, P-RoC® and pyrolysis coal are more efficient with decreasing soil pH. Those fertilizers can be applied to soils with pH < 6.0, or might be used as source material for fertilizer production. During recycling of P from sewage sludge, special attention should be paid to specific production conditions, in order to obtain fertilizer P that might directly be used in crop production to improve plant P supply. There is still a need of investigating recycled P fertilizers in field experiments on appropriate sites that generally respond to fertilizer P applications, including a responsive P fertilizer. Within several crops of a crop rotation, an immobilization of fertilizer P could be seen. Thus, recycled fertilizers should be applied to responsive crops in the crop rotation.

One promising approach to increase overall bioavailability of recycled P fertilizers to maize is a previous cultivation of red clover in the crop rotation. By a combination of different effects, red clover might contribute to a mobilization of sparingly soluble fertilizer P species and delay the soil immobilization of readily available P through BNF. However, external P inputs to maize might still be necessary to secure optimum plant growth. One consequence of this approach might though be an overall reduction of

fertilizer P input to the crop rotation through an overall increased P use efficiency. This has to be investigated in future experiments. In contrast, beneficial effects of applying bioeffectors to soil are very limited. An application of bioeffectors might be unprofitable, especially in organic farming, due to their high intrinsic abundance of rhizosphere microbes that effectively mobilize P in the rhizosphere.

Once more, the poor solubility of PR at $\text{pH} > 6.0$ was shown in our experiments, highlighting the importance of finding suitable alternative P fertilizers for organic crop production, to maintain current yields. Most recycled fertilizers have a higher fertilizer efficacy than PR, and contain by far lower PTE compared to some fertilizers that are allowed in organic farming (e.g. compost, PR). Furthermore, the concept of nutrient recovery and recycling is basically in line with fundamental organic ideas, and the acceptance of recycled P fertilizers among stakeholders of the organic sector seems generally high. These are good prerequisites on the way towards a permission of recycled P fertilizers in organic farming regulations. With the implementation of novel P fertilizers into agricultural production, in general, it is necessary to turn away from traditional description methods of P fertilizers, such as their solubility in water. Instead, an implementation of novel, more differentiated description methods has to be considered to describe and examine the solubility and bioavailability of recycled fertilizers. Phosphors fertilizers recycled from sewage sludge have been shown to be a suitable alternative for organic crop production in many ways. By an implementation of those fertilizers, organic agriculture would, once more, be pioneering the way towards a farming system of the future, which inevitably will comprise a closed P cycle between urban and rural areas.

Summary

Phosphorus (P) nutrition of plants is a key production factor in agriculture. In an approach to recycle P from urban areas back to agriculture, technologies have been developed to produce mineral P fertilizers out of municipal sewage sludge. In this study, different P fertilizers recycled from sewage sludge have been investigated in pot and field experiments for their bioavailability to maize and several plant species of a crop rotation. It was also investigated, if bioavailability of recycled P fertilizers can be enhanced either by a soil inoculation with different bacteria strains that are efficient in P solubilizing, or by a cultivation of red clover in the crop rotation. As there is a lack of bioavailable P fertilizers in organic cropping systems, P fertilizers recycled from sewage sludge were evaluated for their suitability to be used in organic crop production.

It has been shown that most of the investigated fertilizers recycled from sewage sludge have a higher P bioavailability than Phosphate Rock (PR). Fertilizer efficacy seems very dependent from specific production conditions which are decisive for the final product. Among the tested fertilizers, struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6 \text{H}_2\text{O}$) was most efficient in increasing plant P offtake of maize (+ 27.5% in the field, and more than sixfold in a pot experiment, compared to the unfertilized control). Struvite and calcined sewage sludge ash (SSA) are efficient fertilizers at both acidic and neutral soil pH. Other fertilizers (e.g. untreated incineration ashes) have low solubility at soil with pH > 6, and thus, might be used on acidic soil only, or as raw material for fertilizer production. In the field experiment, the overall response to P fertilizer input was low, which probably

can be attributed to a sufficient inherent P supply on the used site. An immobilization of fertilizer P over time could be shown in all experiments. Thus, recycled P fertilizers should be applied to responsive crops in the rotation. An improved P supply of maize could be shown when grown after red clover in the crop rotation. This might be attributed to a combination of different factors, such as a solubilization of sparingly soluble P forms in recycled fertilizers, following a drop in soil pH due to biological N₂ fixation of clover. A recycling of P to maize via decomposed clover roots might in addition have contributed to an increased P supply of the subsequent maize. Despite this promising effect, P mobilization by clover cultivation was not sufficient to cover the entire P demand of maize. Thus, additional fertilizer P inputs to maize might still be necessary to ensure optimal plant growth on P deficient soils. With one exception, an application of different bacteria strains generally did not affect P supply of the plants. Applied bacteria seem very dependent on the environmental conditions. It is conceivable, that especially in organic systems, a soil application with external bacteria does not enhance the beneficial effects of a high microbial abundance and activity which often is already present in organic cropping systems.

From an agronomic point of view, P fertilizers recycled from sewage sludge are better alternatives for organic crop production than PR. A recycling of nutrients generally fits well with basic organic principles. By introducing those fertilizers, the organic system could make a decisive contribution to the ongoing effort of closing the P cycle, and, once more, develop towards a farming system of the future.

Zusammenfassung

So wie alle anderen Lebewesen, sind auch Pflanzen auf das Element Phosphor (P) angewiesen. Es ist umstritten, wie lange die natürlichen Rohphosphat (PR) Reserven noch ausreichen, um den P-Bedarf der Landwirtschaft zu decken. Vor diesem Hintergrund haben sich verschiedene Technologien entwickelt um P-Düngemittel aus kommunalem Klärschlamm zu gewinnen. Angesichts des Mangels an pflanzenverfügbaren P-Düngemitteln im Ökologischen Landbau wurde evaluiert, ob diese Recycling-P Düngemittel aus agronomischer Sicht für die Ökologische Pflanzenproduktion infrage kommen. Dafür wurden in dieser Studie verschiedene Recycling-P Düngemittel in Gefäß- und Feldversuchen mit mehreren Pflanzen einer Fruchtfolge auf ihre Pflanzenverfügbarkeit hin untersucht. Darüber hinaus wurde untersucht, ob durch eine Beimpfung des Bodens mit verschiedenen Bakterienstämmen, oder den Anbau von Rotklee in der Fruchtfolge, die Pflanzenverfügbarkeit der Recycling-P Düngemittel erhöht werden kann.

Es konnte gezeigt werden, dass die meisten Recycling-P Düngemittel eine höhere Pflanzenverfügbarkeit aufweisen als PR. Offenbar wird die P-Düngewirkung dabei insbesondere von den jeweiligen Produktionsbedingungen bestimmt. Von allen untersuchten Recycling-P Düngemitteln hatte Struvit ($\text{MgNH}_4\text{PO}_4 \cdot 6 \text{H}_2\text{O}$) die höchste P-Düngewirkung. Im Vergleich zur ungedüngten Kontrolle erhöhte eine Düngung mit Struvit die P Aufnahme von Mais um 27.5% im Feldversuch und um mehr als das sechsfache im Gefäßversuch. Struvit und kalzinierte Klärschlammasche (KSA) sind effektive P Düngemittel, unabhängig von den jeweiligen Bodenbedingungen. Andere

Düngemittel, wie etwa unbehandelte KSA, weisen bei einem pH der Bodenlösung von > 6.0 hingegen eine niedrige Pflanzenverfügbarkeit auf. Diese kommen daher nur für eine Nutzung auf sauren Böden, oder aber als Rohmaterial für die Düngemittelproduktion infrage. Die allgemein geringe Wirkung der P-Düngung im Feldversuch ist wahrscheinlich auf eine ausreichende P-Versorgung des Versuchsstandorts zurückzuführen. In allen Versuchen konnte mit der Zeit eine Immobilisierung von Dünger-P im Boden gezeigt werden. Recycling-P Düngemittel sollten daher innerhalb einer Fruchtfolge zu Kulturen mit hohem P-Bedarf gedüngt werden. Eine gesteigerte P-Versorgung von Mais konnte durch einen vorherigen Anbau von Rotklee in der Fruchtfolge erzielt werden. Die Absenkung des pH in der Bodenlösung infolge biologischer Stickstofffixierung hat vermutlich zu einer erhöhten Pflanzenverfügbarkeit schwerlöslicher P-Verbindungen aus Recycling-P Düngemitteln geführt. Darüber hinaus hat möglicherweise auch eine Freisetzung von P aus zersetzten Kleewurzeln zu einer verbesserten P-Versorgung des darauffolgenden Mais beigetragen. Die durch den Anbau von Klee mobilisierte P-Menge war jedoch nicht hoch genug um den gesamten P-Bedarf von Mais zu decken. Zu Mais sind daher vermutlich zusätzliche P-Düngegaben erforderlich. Eine P-Mobilisierung durch die Applikation verschiedener Bakterienstämme konnte in nur einem Fall erzielt werden. Möglicherweise bleibt der gewünschte Effekt einer P-Mobilisierung durch zusätzlich applizierte Bakterien gerade im Ökologischen Landbau aus, für den eine hohe mikrobielle Aktivität im Boden charakteristisch ist. Aus agronomischer Sicht sind Recycling-P Düngemittel aus Klärschlamm geeignetere Alternativen als PR für die ökologische Pflanzenproduktion. Ein Nährstoffrecycling wird von ökologischen Leitlinien grundsätzlich befürwortet. Mit der Aufnahme dieser neuartigen Düngemittel

kann der Ökologische Landbau einen entscheidenden Beitrag zur Schließung des P Kreislaufes leisten und sich - einmal mehr - zu einem Bewirtschaftungssystem der Zukunft weiterentwickeln.

References for the General Introduction und Discussion

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Curriculum Vitae

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Common Purpose Frontrunner, Stuttgart (04.2013)

P-REX workshop, Berge (08.2014)

P-REX summerschool, Basel, CH (09.2014)

P-Symposium ETH Zürich, CH (01.2015)

Internship

Practical work experience in agriculture, goat farm

Myrdal Gard, Lundegrend, Norway (06.2005 – 06.2006)

Special skills

Computer/IT

MS Office word, excel, power point

Statistical software SAS, SigmaStat, R

languages

German, English, Norwegian, French

Articles (peer-reviewed)

Wollmann, I., Möller, K. (2019): Red clover (*Trifolium pratense* L.) increases plant growth and P offtake from sewage sludge ashes for a subsequent maize. (submitted).

Hartmann, T. E., **Wollmann, I., You, Y., Müller, T. (2019):** Sensitivity of Three Phosphate Extraction Methods to the Application of Phosphate Species Differing in Immediate Plant Availability. *Agronomy* 9(1), 29.

Wollmann, I., Möller, K. (2018): Phosphorus bioavailability of sewage sludge-based recycled fertilizers in an organically managed field experiment *J. Plant Nutr. Soil Sci.* 181, 760 – 767.

Möller, K., Oberson, A., Bünemann, E., Cooper, J., Friedel, J., Glaesner, N., Hörtenhuber, S., Løes, A.-K., Mäder, P., Meyer, G., Müller, T., Symanczik, S., Weissengruber, L., **Wollmann, I., Magid, J. (2018):** Improved Phosphorus Recycling in Organic Farming: Navigating Between Constraints. *Adv. Agron.* 147, 159 – 237.

Wollmann, I., Gauro, A., Müller, T., Möller, K (2018): Phosphorus bioavailability of sewage sludge-based recycled fertilizers. *J. Plant Nutr. Soil Sci.* 181, 158 – 166.

Benke, A. P., Rieps, A.-M., **Wollmann, I., Petrova, I., Zikeli, S., Möller, K. (2017):** Fertilizer value and nitrogen transfer efficiencies with clover-grass ley biomass based fertilizers. *Nutr. Cycl. Agroecosys.* 107, 395 – 411.

Contributions to scientific conferences

Wollmann, I., Möller, K. (2017): Phosphorus availability of fertilizers recycled from urban waste water in combination with bioeffectors – pot and field experiments. International plant nutrition colloquium (IPNC), Copenhagen, DK (**oral presentation**).

Wollmann, I., Möller, K. (2016): Phosphorus availability of recycled fertilizers from sewage sludge. Jahrestagung Deutsche Gesellschaft für Pflanzenernährung (DGP), Stuttgart (**oral presentation**).

Wollmann, I., Möller, K. (2015): Phosphorverfügbarkeit mineralischer Recyclingprodukte aus der Klärschlammaufbereitung in Kombination mit Bioeffektoren, Ergebnisse aus Gefäß- und Feldversuchen. 127. VDLUFA-Kongress, Göttingen (**oral presentation**).

Wollmann, I., Möller, K. (2015): Recycled phosphorus fertilizers from urban residues tested in agricultural crop production. 16th International Conference Rural-Urban Symbiosis (RAMIRAN), Hamburg (**oral presentation**).

Wollmann, I., Möller, K. (2015): Recycling Phosphor Düngemittel für den Ökologischen Landbau. 13. Wissenschaftstagung Ökologischer Landbau, Eberswalde (**oral presentation**).

Wollmann, I., Möller, K. (2014): Phosphorverfügbarkeit von mineralischen Recycling-Phosphor-Düngemitteln und deren Interaktion mit Bioeffektoren. 126. VDLUFA-Kongress, Stuttgart (**poster presentation**).

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