

# 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<sup>®</sup>, P-RoC<sup>®</sup>, Mephrec<sup>®</sup>, 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<sup>®</sup>  $\geq$  Struvite AirPrex<sup>®</sup>  $\geq$  Mephrec<sup>®</sup> > 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

Accepted October 19, 2017

## 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 (Bolland 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), 11<sup>6</sup> 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 N<sub>2</sub>-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  $\text{NaHCO}_3$ ,  $\text{NaOH}$ , and  $\text{H}_2\text{SO}_4$ . The amount of  $\text{NaHCO}_3$ -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  $\text{NaHCO}_3$ -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<sup>®</sup> is commercially available and produced by Berliner Wasserbetriebe (BWB Berlin, Germany). In the AirPrex<sup>®</sup> 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  $\text{CO}_2$ -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<sup>®</sup> 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<sup>®</sup> 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<sup>®</sup> process (INGITEC 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<sup>®</sup> process (Outotec, Finland). Sewage sludge ash is thermochemically treated with  $\text{MgCl}_2$  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<sup>®</sup> 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  $\text{CaSO}_4$ . 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  $2 \times 10^8$  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  $\text{CaSO}_4$  solution. For the ryegrass in Experiment 1, the application strategy was changed towards a less frequent, but higher concentrated application rate of  $2 \times 10^9$  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 ( $\text{CaCl}_2$ ; VDLUFA, 1991), total P: 55.4 mg P 100 g<sup>-1</sup> DM,  $P_{\text{CAL}}$ : 2.20 mg P 100 g<sup>-1</sup> 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<sup>-1</sup>) and P contained in different fractions of sequentially extracted P (NaHCO<sub>3</sub>, NaOH, H<sub>2</sub>SO<sub>4</sub>), indicated as total amounts (mg P g DM<sup>-1</sup>) and relative quantities (% of total P, bold numbers in parentheses) and solubility of P in water (H<sub>2</sub>O) 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<sup>®</sup> = struvite obtained from the AirPrex<sup>®</sup> process, P-RoC<sup>®</sup> = Ca-Phosphates recovered in the P-RoC<sup>®</sup> process, Mephrec<sup>®</sup> = 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 <sup>a</sup> (mg P g DM <sup>-1</sup> )	Sequentially extracted P			P <sub>H<sub>2</sub>O</sub> (% of total P)	P <sub>2% citric acid</sub>
		NaHCO <sub>3</sub> (mg P g DM <sup>-1</sup> ) and (% of total P)	NaOH (mg P g DM <sup>-1</sup> ) and (% of total P)	H <sub>2</sub> SO <sub>4</sub> (mg P g DM <sup>-1</sup> ) and (% of total P)		
Struvite SSL	233	3.89 (2.98)	20.7 (15.8)	106 (81.2)	6.56	56.1
Struvite AirPrex <sup>®</sup>	223	3.09 (2.63)	20.7 (17.6)	93.6 (79.7)	0.89	37.1
P-RoC <sup>®</sup>	49.8	2.83 (7.29)	0.58 (1.50)	35.5 (91.2)	0.14	67.8
Mephrec <sup>®</sup>	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

<sup>a</sup>Determined 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<sup>-1</sup> with an average light intensity of 430 μmol m<sup>-2</sup> s<sup>-1</sup>, 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 N<sub>2</sub>-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(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub>, 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<sup>-1</sup> DM. Other nutrients were applied in amounts to secure sufficient supply with (per kg soil DM) 100 mg potassium (K) as K<sub>2</sub>SO<sub>4</sub>, 50 mg Mg as MgSO<sub>4</sub> and 35 mg N as Ca(NO<sub>3</sub>)<sub>2</sub>. 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<sup>®</sup> (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<sup>-1</sup> soil DM as NH<sub>4</sub>NO<sub>3</sub>, respectively and with 200 mg K kg<sup>-1</sup> soil DM as K<sub>2</sub>SO<sub>4</sub>. 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(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub>, 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<sup>-1</sup> soil DM were applied as NH<sub>4</sub>NO<sub>3</sub>. 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 NaHCO<sub>3</sub> were added, and tubes were shaken at 180 rpm for 16 h. Suspension was centrifuged at 2500 g for 10 min and NaHCO<sub>3</sub> 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 (VDLUF, 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

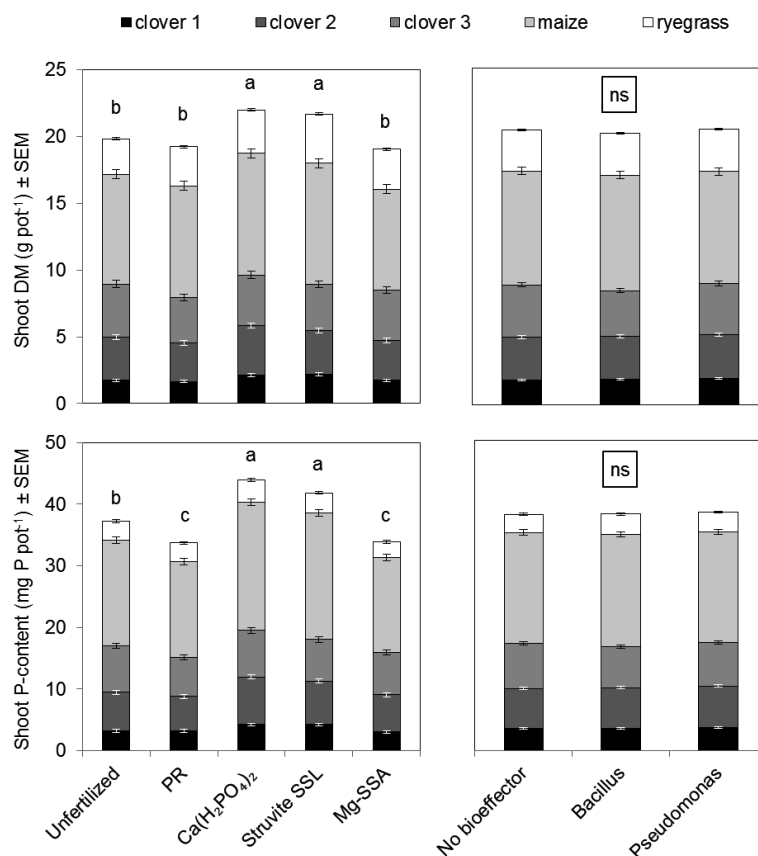
Both experiments were arranged in a completely randomized design with pot rotations. They included 15 treatments with five replicates (Experiment 1), and 18 treatments with four replicates (Experiment 2). Two-way analysis of variance (ANOVA) was performed (SigmaPlot 11.0) to study the effect of different 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 P fertilizers significantly influenced the sum of shoot DM and P content of the test plants (Fig. 1). Fertilization with  $Ca(H_2PO_4)_2$  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-

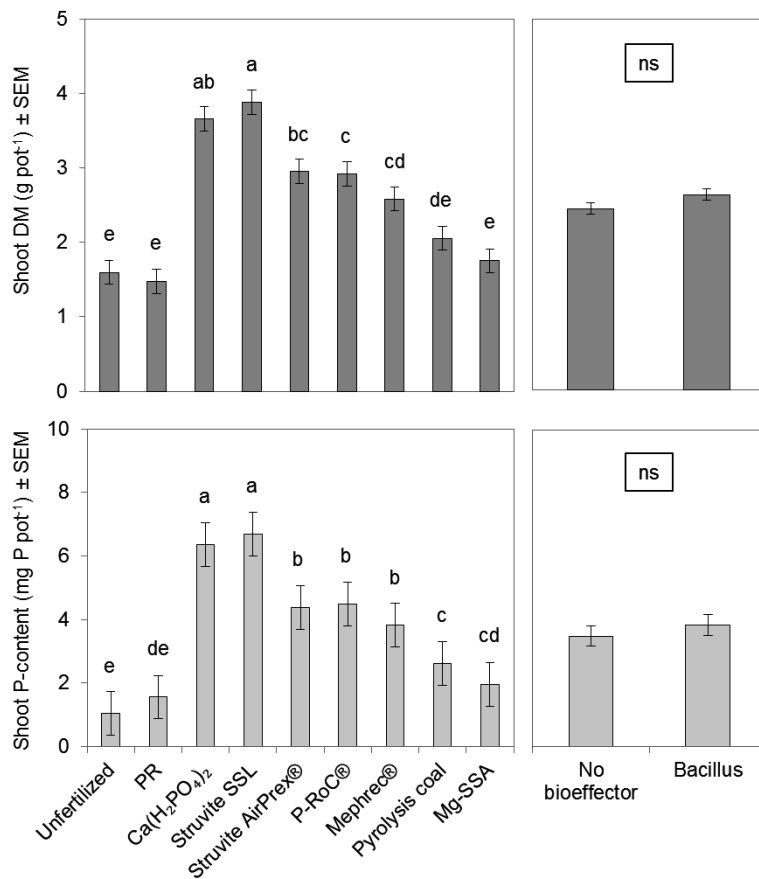


**Figure 1:** Shoot DM ( $g\ pot^{-1}$ ) and shoot P content ( $mg\ P\ pot^{-1}$ ) of clover at harvest dates 1, 2, and 3, maize and ryegrass, grown in Experiment 1, fertilized with recycled P fertilizers and control fertilizers (PR = phosphate rock, Struvite SSL = struvite obtained from Stuttgart Sewage 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  $9.11\ g\ pot^{-1}$  [ $Ca(H_2PO_4)_2$ ] 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  $15.4\ mg\ P\ pot^{-1}$  (Mg-SSA)– $20.8\ mg\ P\ pot^{-1}$  [ $Ca(H_2PO_4)_2$ ] in Experiment 1 and only  $1.04\ mg\ P\ pot^{-1}$  (unfertilized)– $6.69\ mg\ P\ pot^{-1}$  (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  $Ca(H_2PO_4)_2$  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<sup>-1</sup>) and shoot P content (mg P pot<sup>-1</sup>) 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 ≤ 5%).

## 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(H<sub>2</sub>PO<sub>4</sub>)<sub>2</sub> 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

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.

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 CaCl<sub>2</sub>. 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  $\text{CaCl}_2$  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; Condon and Newman, 2011). For a comprehensive P speciation and interpretation of results, a combination of different methods (e.g., solution  $^{31}\text{P}$  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  $\text{NaHCO}_3$ . 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.

## Acknowledgments

This work was carried out within the *EU-project IMPROVE-P*. The authors gratefully acknowledge the financial support for this project provided by the *CORE Organic II Funding Bodies*, being partners of the *7<sup>th</sup> Framework Program ERA-Net project, CORE Organic II* ([www.coreorganic2.org](http://www.coreorganic2.org)). Parts of this study were funded by the *European Community's Seventh Framework Programme (FP7/2007–2013)* under grant agreement no. 312117 (*BIOFECTOR*). Special thanks also go to *Anke Ehbrecht, Carsten Meyer, Joachim Mallon, Prof. Thomas Appel, Kevin Friedrich* and *Outotec* for providing their recycled P fertilizers, and to *Sourcon Padena* and *ABiTEP* for providing their bioeffector products.

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