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Phosphorus Balance in Sandy Soil Subjected to 12 Years of Successive Applications of Animal Manure and Mineral Phosphate Fertilizer in Subtropical Climate

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† This work is part of the doctoral thesis Phosphorus balance in 12 years and annual crops strategies to access phosphorus reserves built over 15 years by organic and mineral sources, presented at the Federal University of Santa Maria.

Abstract: Assessing the phosphorus (P) balance in agricultural soils is crucial for optimizing its use and reducing contamination risks. The objective of this study was to evaluate the impact of different animal wastes on the distribution of P in soil profiles subjected to 12 years of successive applications of animal wastes and mineral phosphate fertilizer, within a crop rotation system under no-till system. The study was conducted from 2004 to 2016 in the southern region of Brazil. The treatments were the applications of pig slurry (PS), cattle slurry (CS), pig deep-litter (PD), mineral fertilizer (MF), and a control treatment without application. The highest accumulation of P and its movement was observed in the 0–40 cm layer, in the soil submitted to applications of all P sources. The inputs of P via MF, CS, PS, and PD promoted the accumulation of 18, 42, 48, and 100 kg P ha^{-1} $year^{-1}$. The P mass balance showed that between 77 and 98% of the P added by animal manure and MF was accounted for in grain exports (17–34%), soil storage (41–72%), and post-harvest residues (<1%), with the remaining 2–33%, unaccounted for, which was attributed, especially, to P transfer at the soil surface.

Keywords: P accumulation; no-till farming system; P legacy; P contamination

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1. Introduction

Phosphorus (P) is one of the most important nutrients for maintaining or increasing the productivity of plants in agroecosystems. The demand for this nutrient for food and bioenergy production has intensified over the years as the world's population increases, raising concerns about its supply since P sources are limited and non-renewable [1,2]. Global swings in the price of phosphate fertilizers happen to accompany the depletion and scarcity of phosphate rock [3,4]. Therefore, it is critical to find viable alternatives that make it possible to meet food demand without relying solely on mineral phosphate fertilizers. An alternative could be the use of liquid animal waste, such as from pigs and cattle, or solid, such as pig deep-litter, as P sources in cropping systems [5,6].

The states in the southern region of Brazil (Rio Grande do Sul, Santa Catarina, and Paraná) are responsible for approximately 70% of Brazil's swine production and export more than 84% of the meat from these animals, with 695,000 tons of meat (18.5% of total national production) produced in Rio Grande do Sul [7]. Moreover, Rio Grande do Sul contributed 13% of the national milk production (812 thousand liters in the first quarter of 2019) [8]. These activities, when conducted in confined system, which represents almost the totality for swine production, generate a large volume of waste rich in nutrients, including P. This large volume of waste is used in agricultural soils as an alternative of disposal, but also as a way to reuse nutrients. However, successive applications of manure as fertilizer, in amounts commonly greater than those exported by crops, cause accumulation of P in soils over time [9].

However, long-term studies quantifying the role of legacy P in sustaining agricultural systems for food production are still scarce, especially in soils with higher adsorption capacity, such as those in tropical and subtropical regions. A field trial was established in Rio Grande do Sul, Brazil, in 2004 to evaluate the agronomic responses of grain with different types of animal wastes, along with changes in soil properties and associated potential environmental impacts. The objective of this study was to quantify the accumulation, distribution, and indicators of potentially adverse environmental impacts following the application of different types of animal manure and mineral phosphate fertilizer in a long-term no-till soil. Due to the surface application and high adsorption capacity of the soil, most of the P added via animal manure and mineral fertilizer was retained in the topsoil layers.

2. Materials and Methods

2.1. Location of Experiment and Treatments

The study was performed in a long-term experiment, with 19 years of conduction, located in the experimental area of the Federal University of Santa Maria (UFSM), in Santa Maria, Rio Grande do Sul (RS), in the southern region of Brazil (29°43′12″S and 53°43′4″W). The climate of the region is humid subtropical (Cfa 2), according to Koppen's classification, with annual averages of temperature, precipitation, and relative humidity of 19.3 °C, 1561 mm, and 82%, respectively. The soil is classified as Arenic Dystrophic Red Argissolo, according to Embrapa [10], corresponding to Typic Hapludalf [11], with 108 g kg^{-1} of clay, 183 g kg^{-1} of silt, and 709 g kg^{-1} of sand. The chemical properties of the soil before the installation of the experiment (2004) and at the time of the study (2016) are presented in Table 1.

Table 1. Chemical properties of soil collected at 0–10 cm before experiment implant (2004) and after 18 applications of organic and mineral N sources (August 2016). The values are the averages of the four plots.

Chemical Properties		**Treatments (2016)**				
	Initial Soil (2004) *	**Control**	**CS**	**PS**	**PD**	**FM**
pH$_{H2O}$	4.9	5.2	5.3	4.9	5.7	4.5
SOM (g kg^{-1}) $^{(1)}$	19	14.0	21.9	16.9	20.9	15.9
Available P (mg kg^{-1}) $^{(2)}$	20.3	9.7	76.5	72.9	93.2	55.1
Available K (mg kg^{-1}) $^{(2)}$	60	29.0	117.0	71.8	116.8	75.0
Exchangeable Ca (cmol$_c$ dm^{-3}) $^{(3)}$	0.8	2.2	3.3	2.1	4.4	1.4
Exchangeable Mg (cmol$_c$ dm^{-3}) $^{(3)}$	0.3	1.8	2.6	1.7	2.8	1.0
Exchangeable Al (cmol$_c$ dm^{-3}) $^{(3)}$	0.03	0.5	0.2	0.6	0.1	1.3
H+Al (cmol$_c$ kg^{-1})	3.7	3.7	3.7	3.3	1.9	4.1
CEC$_{effective}$ (cmol$_c$ kg^{-1})	1.3	4.6	6.4	4.5	7.6	3.9
CEC$_{pH\ 7.0}$ (cmol$_c$ kg^{-1})	5	7.8	9.9	7.2	9.5	6.7
Base Saturation (%)	25.4	53.7	63.5	56.3	80.0	38.8
Aluminum Saturation (%)	2	11.0	2.9	12.9	1.3	33.2

* Data adapted from Lourenzi et al. [12]. $^{(1)}$ Determined using modified Walkley–Black method + Van Bemmelen factor; $^{(2)}$ Extracted by Mehlich-1; $^{(3)}$ Extracted by KCl 1 mol·L^{-1}; Control = no fertilization; CS = cattle slurry; PS = pig slurry; PD = pig deep-litter; FM = mineral fertilizer.

The experiment was initiated in 2004, under a no-till farming system. The experimental design was randomized blocks, with four replicates. The plots had dimensions of 5 × 5 m The treatments were pig slurry (PS), cattle slurry (CS), pig deep-litter (PD), mineral fertilizer (urea + triple superphosphate—SFT + potassium chloride—KCl) (MF) and a control treatment, without the application of nutrients. The applications of PS, CS, PD, and mineral fertilizer were made on the surface, over the cultural residues of the previous crop and without incorporation. The treatments were applied only once a year and before summer crop sowing, in the period from 2004 to 2009. The treatments were not applied in winter. Starting in 2010, the treatments were applied twice each year, prior to winter and summer crop establishment. The crop succession used until August 2016 was black oats (*Avena strigosa S.*), corn (*Zea mays*), forage turnips (*Raphanus sativus*), black beans (*Phaseolus vulgaris*), and wheat (*Triticum* spp.). Further details about the experiment can be obtained in Lourenzi et al., Ferreira et al., Rodrigues et al., and Marhezan et al., [12–14].

2.2. Collecting Soil Samples

In June 2016, soil samples were collected in the layers 0–4, 4–10, 10–20, 20–30, 30–40, and 40–50 cm. For this, in each plot, two trenches with dimensions of 40 × 40 × 40 cmwere opened. In each trench, two soil samples per plot were collected, one in the longitudinal direction and the other transversal to the sowing line. To correct the soil attributes and concentrations in units of mass per area in hectares, in each sampled layer, the bulk density of the soil was determined by means of metal volumetric rings (78 cm^3), as described by Embrapa [15].

2.3. P Fractionation in Soil

The soil was air dried, ground, and passed through a 2 mm mesh sieve. It was then prepared and subjected to sequential P chemical fractionation with ammonium chloride (NH_4 Cl 1 mol l^{-1}), sodium bicarbonate ($NaHCO_3$ 0.5 mol l^{-1}, pH 8.5), sodium hydroxide (NaOH 0.1 mol l^{-1}), hydrochloric acid (HCl 1 mol l^{-1}), and a second extraction with NaOH (0.1 mol l^{-1}) [16–18]. Total soil P was determined in a soil subsample after digestion with concentrated sulfuric acid (H_2SO_4) and hydrogen peroxide (H_2O_2 at 30% v/v), according to Olsen and Sommers [19]. The inorganic P in the alkaline extracts of $NaHCO_3$ and NaOH was assigned to the reactive P-molybdate analyzed by the method of Dick and Tabatabai [20]. In the alkaline extracts, total P was determined by digestion with ammonium persulfate + sulfuric acid in an autoclave. Unreacted P-molybdate was considered as organic P and was obtained by the difference between total P and inorganic P. The P in acid extracts was determined according to the method of Murphy and Riley [21].

Total organic P extracted was determined as the sum of the organic P extracted with $NaHCO_3$ and the first and second NaOH extractions. Total extracted inorganic P was calculated as the sum of inorganic P in all extracted fractions. The difference between total soil P and total extracted P (inorganic + organic) was considered residual P. Soil P fractions were grouped into four P pools according to their relative lability: labile P (sum of NH_4 Cl and $NaHCO_3$ fractions (inorganic and organic)); moderately labile P (first NaOH (inorganic and organic)); stable P (HCl + second NaOH (inorganic and organic)); and residual P (non-extractable P) [22]. The amounts of total P, total inorganic P, and total organic P up to 50 cm were expressed in units of kg ha^{-1}.

The potential risk of P transfer was evaluated by P concentration values (mg P kg^{-1}) determined in the first extraction of sequential P fractionation (NH_4 Cl-P). For this, we considered the most superficial layers (0–4 and 4–10 cm), following the recommendations of McDowell and Sharpley [23]. Easily extractable P in soil can be used as an indicator of environmental risk, because this P fraction has a high potential for transferring from the soil by erosion processes and surface runoff [24–26]. When the reactive functional groups of soil surfaces can no longer adsorb P, the equilibrium point is established and, as a consequence, large amounts of weakly bound P will be present in the soil solution (in this study represented by NH_4 Cl-P), thus increasing potential P transfers [23].

2.4. P Added by Manure and Mineral Fertilizer

Liquid waste derived from pig and cattle farming was collected in open effluent tanks. The overlapping pig litter was collected in covered environments on the pig farms. The general physicochemical properties of the manure were analyzed on a representative sample before each application on all crops was performed. The applied dose of each organic waste was defined based on its N concentration and the N requirement needed by the crops, according to the regional recommendation of the Commission for Soil Chemistry and Fertility CQFS-RS/SC [27]. Therefore, the P inputs varied among the treatments. The total P inputs in the manure were calculated based on the P concentrations (kg m^{-3} for liquid manure and kg kg^{-1} for solid manure), present in the manure, multiplied by the amount of manure applied (m^3 manure ha^{-1} for CS and PS and kg manure ha^{-1} for PD) and, therefore, were expressed in kg P ha^{-1}. For MF, the P inputs were made according to the P contents present in the soil and the nutritional requirement of the crop. The P inputs (in kg P ha^{-1}) in the manure and MF, by crop season/year are presented in Figure 1.

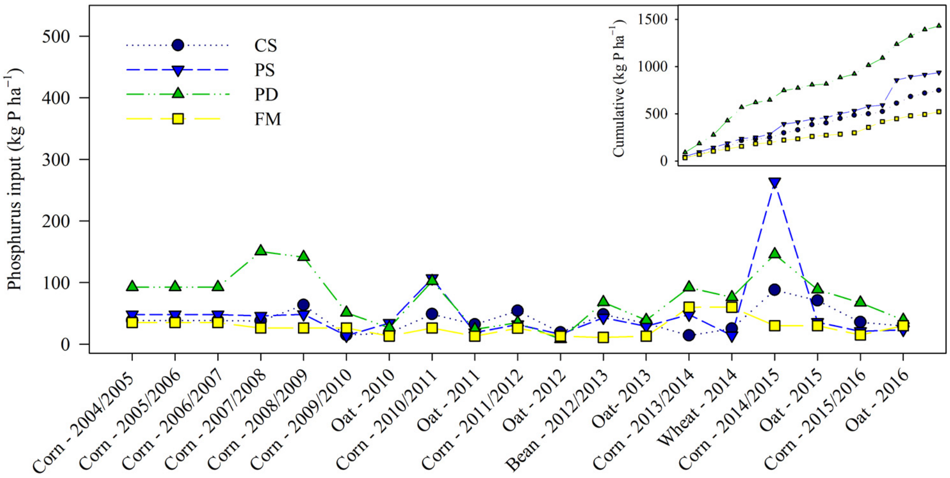

Figure 1. Total P applied (expressed in kg P ha^{-1}) to field plots receiving cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF) from 2004 to 2012 under no-tillage system.

2.5. P in Residue and Exported by Crops

Plants were collected at flowering (winter and summer crops). For this, the shoot of the plants was cut close to the soil surface. Plants were dried in a forced air oven at ±65 °C to constant mass for dry matter determination. The tissue was ground, prepared, and subjected to nitric-perchloric digestion (ratio 4:1) [15]. P was determined in the extract according to Murphy and Riley [21]. This procedure was repeated for the determination of P present in grains and cereals. For the calculation of the P balance in the system, the $P_{residue}$, consists of the P present in the cultural residues after harvest. For this, we used the data of P contained in the dry matter of the oat crop established at the time of soil collection.

2.6. P Balance

The P balance in the soil–plant system (kg ha^{-1}) was estimated considering the 50 cm layer, the amount of the nutrient added in fertilization (kg ha^{-1}), the amount of P exported by grain crops (kg ha^{-1}), and the change in the amount of P in the soil. To determine the P budget of the system, we used a similar approach to that described by Boitt et al. [28].

The P mass balance was calculated empirically as follows (units in kg P ha^{-1}) (Equation (1)):

$$P_{added} - \Delta P_{soil} - P_{exported} - P_{residue} - P_{unaccountedfor} = 0 \quad (1)$$

where: P_{added} is the cumulative sum of P added by the treatments; ΔP_{soil} is the change in P storage in the soil profile ($P_{initial} - P_{final}$) calculated by summing the amounts of P determined in each soil layer (0 to 50 cm layer); $P_{exported}$ is P exported by grains and cereals; $P_{residue}$ is the P content of crop residues and $P_{unaccounted\,for}$ is the difference between P_{added} and the sum of Pcounted for pools, which was attributed to some accumulated uncertainties in P accounting in the mass balance. For the control treatment, P_{added} was zero and $P_{unaccounted}$ was considered insignificant, given the absence of P addition and minimal soil losses through erosion and runoff in this experiment [29]. Therefore, the soil from the control treatment was used to calculate $P_{initial}$ and was considered equal throughout the experiment area. This made it possible to determine the ΔP_{soil} of all treatments.

Total P inputs accounted for ($P_{accounted}$; Equation (2)) in the treatments fertilized with manure and mineral fertilizer are given by the difference in the sum of all P pools (given by $\sum P_{Pool}$; Equation (3)), minus the sum of the P pools of the control treatment (no P inputs) as follows:

$$P_{acconted}\left(\mathrm{kgha}^{-1}\right) = \sum P_{pools} treat.fertilized - \sum P_{pools} control \quad (2)$$

Being,

$$\sum P_{pools} = P_{soil} + P_{exported} + P_{crop\ residue} \quad (3)$$

Thus, the P accounted for in the system (for each treatment that received fertilization) was calculated according to Equation (4):

$$P_{acconted}(\%) = \frac{P_{acconted}[\mathrm{kgha}^{-1}]}{P_{input}[\mathrm{kgha}^{-1}]} \times 100 \quad (4)$$

The cumulative agronomic efficiency of the P balance of the system was calculated according to Equation (5):

$$P\ balance\ efficiency(\%) = \frac{P_{exported}}{\left(P_{exported} + \Delta P_{soil}\right)} \times 100 \quad (5)$$

where ΔP_{soil}, in units of kg P ha^{-1}, is the change in soil P storage in the 0 to 50 cm soil layer calculated using Equation (6):

$$\Delta P_{soil}\left(\mathrm{kgha}^{-1}\right) = P_{final} - P_{initial} \quad (6)$$

2.7. Statistical Analysis

Differences in mean soil P fractions between treatments and depths, as well as crop yields, were determined using ANOVA and when significant were compared by the Scott–Knott 5% test. In addition, linear correlations (Pearson correlation coefficients) between the data were determined using R software [30].

3. Results

3.1. P in Soil

The additions of CS, PS, PD, and MF over 12 years increased the amounts of P in all fractions (Figure 2 and Supplementary Table S1). The highest values of labile P were observed in the superficial layers in soils submitted to animal manure applications (CS, PS, PD). However, among the manure sources, the highest values of labile P up to 40 cm layer were observed in the soil with PD applications (Figure 2a). The moderately labile P fractions increased up to 40 cm, especially in soils with animal manure applications, and the highest amounts were observed in soils with PD application (Figure 2b). The highest values of stable P were observed up to 30 cm, in soils submitted to residue and mineral fertilizer applications (Figure 2c). The highest values of residual P (not extractable by chemical P fractionation, Figure 2d) were observed up to 30 cm in the control and MF soils.

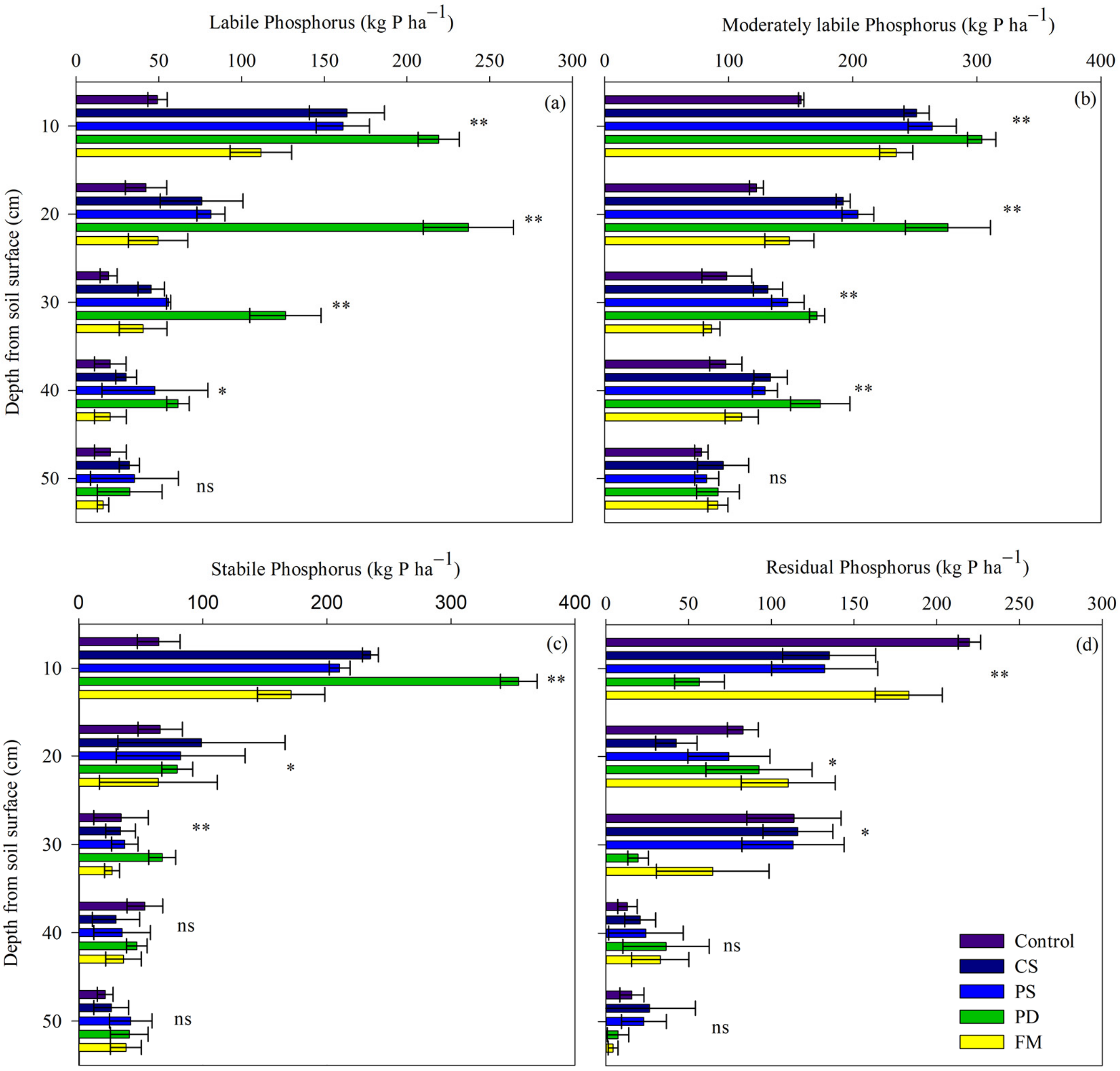

Figure 2. Average P quantities (kg P ha−^{1}) determined in P fractions for soils sampled from different depths after 12 years under applications cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF): labile P (PiAM + Pibic + Pobic) (**a**), moderately labile P (PiOH_I + PoOH_I) (**b**), stable P (PiHCl + PiOH_II + PoOH_II) (**c**), and residual P (Pres) (**d**). * $p < 0.05$; ** $p < 0.01$ and ns: not significant (Scott–Knott test at 5%). Standard deviation is presented on the surface of the bars. Data presented are means (n = 4).

The highest values of inorganic P were observed in the soil fertilized with PD, followed by PS, CS, and MF, with increases of 73, 54, 50, and 40%, relative to the control soil (0–50 cm layer) (Figure 3a). The additions of PD, CS, PS, and MF increased surface inorganic P contents by 79, 69, 65, and 52%, respectively, relative to the control (0–10 cm layer), with observed amounts greater than 580 kg P ha^{-1} in the soil with PD addition. The contribution of inorganic P to total soil P varied from 26% in the control soil to 54, 41, 39, and 37% in the soils with PD, PS, CS, and MF applications, respectively (layer 0–50 cm, Figure 3a,c and Supplementary Table S2). On the other hand, the highest amounts of organic P were observed in soil with PD, followed by CS, PS, and MF, with increments of 36, 30, 29, and 6%, relative to the control (layer 0–50 cm Figure 3b). The additions of PD, PS, CS, and MF increased the amounts of organic P on the surface with values 50, 47, 43, and 42% higher than the control (layer 0–10 cm). The proportion of soil organic P varied from 37% in the soil with PD and MF additions, to 42% in the control soil and with PS and CS applications (layer 0–50 cm, Figure 3b,c and Supplementary Table S2).

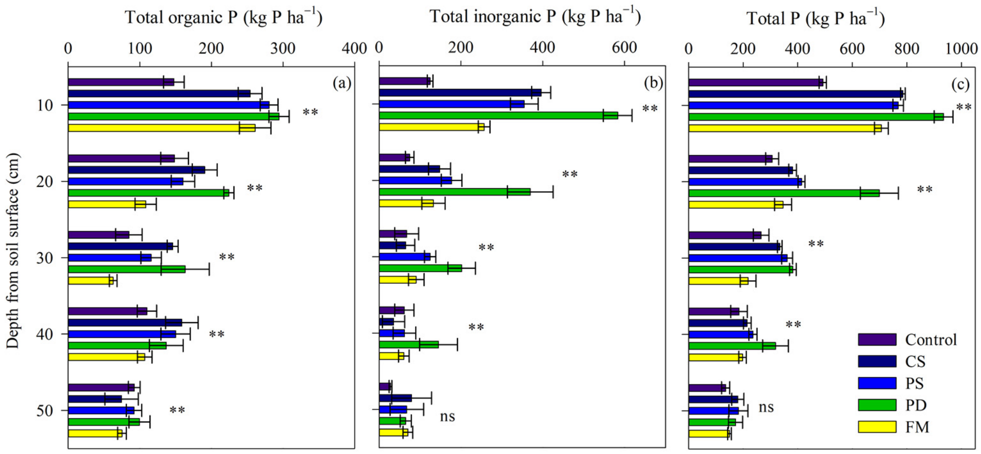

Figure 3. Average quantities (kg P ha^{-1}) of total extracted inorganic P (Pi), total extracted organic P (Po), and total P determined for soils sampled from different depths after 12 years under applications cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF) (total extracted inorganic P = sum of PiAM, Pibic, PiOH_I, PiHCl, PiOH_II (**a**), total extracted organic P = sum of Pobic + PoOH_I + PoOH_II) (**b**) and Total P (**c**). ** $p < 0.01$ and ns: not significant (Scott–Knott test at 5%). Standard deviation is presented on the surface of the bars. Data presented are means (n = 4).

The increase of total P and its distribution in the soil profile were proportional with the amounts of P added by the treatments. The highest amounts of total P were observed in the soil with PD applications, followed by PS, CS, and FM, with increases of 79, 41, and 37.16%, respectively, relative to the control soil (layer 0–50 cm, Figure 3c and Table 2). The highest values of total P in depth were seen in the soil with PD applications, with increment until the 40 cm layer. On average, applications of PD, PS, CS, and MF increased the amounts of total P by 91, 48, 42, and 18 kg P ha^{-1} year^{-1} in the soil up to the 50 cm layer, relative to the control.

Table 2. P mass balance in the no-tillage cropping system receiving long-term P inputs in cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF). The P pools presented are the cumulative amounts (in units of kg P ha^{-1}) after 18 successive applications of the treatments from 2004 to 2016. Data presented are means (n = 4).

P Budget in the System		**Treatments**				
		Control	**CS**	**PS**	**PD**	**FM**
		kg P ha^{-1}				
Inputs	P added via treatments [a]	0	747.5	938.2	1431.1	538.71
Pools	Soil P pools [b]	1383.9 e *	1892.9 c	1971.1 b	2417.1 a	1605.5 d
	Crop P residue [c]	4.8 d	10.3 a	10.3 a	9.5 b	8.7 c
Outputs	Grain P export [d]	98.88 d	218.08 b	242.28 a	243.87 a	184.22 c
Total P accounted [e]		1487.6 e	2121.3 c	2223.7 b	2670.5 a	1798.4 d
P unaccounted [f]		0	10.1 b	98.4 a	144.4 a	124.2 a
Accounted P in the system (%) [g]		0	98.6 a	89.5 b	89.9 b	76.9 c
Added P accumulated in soil (%) [h]		0	68.1 b	62.6 c	72.2 a	41.1 d
P balance efficiency (%) [i]		0	30.0 b	29.2 b	19.1 c	45.5 a

* Letters identify significant differences in the means between treatments for a given P pool (Scott–Knott test at 5%). [a] Total P added via pig slurry after 12 years (2004–2016). [b] Total soil P in the layer 0–50 cm depth. [c] Amounts of P stored in crop residues. [d] Total amount of P exported in grain (corn and beans [in 2016]) in 12 years as a function of P concentration in grain (% P) and total quantities of produced grain (kg ha^{-1}). [e] Sum of P accounted for in pools (soil and residues) and P outputs (P exported in grain). [f] Punaccounted (kg P ha^{-1}) = Pinput − (ΔPsoil + Poutput + Presidue). [g] Accounted P (%) = [1 − (Punaccounted:Pinput)] × 100. [h] Accumulated P in soil (%) = (Δpsoil:Pinput) × 100. [i] P balance efficiency (%) = [Poutput:(Poutput + ΔPsoil)] × 100.

3.2. P Mass Balance

Cumulative P inputs were 746, 938, 1431, and 538 kg P ha^{-1} in soils with CS, PS, PD, and MF applications, respectively (Table 2). On average, applications of CS, PS, PD, and MF corresponded to inputs of 62, 78, 119, and 45 kg P ha^{-1} year^{-1}, respectively. The variation of P inputs among the crops (Figure 1) was correlated with the dry matter concentrations in the manure (Pearson's correlation coefficient = 0.41, p value < 0.001) and also by the N content present in the manure, since the amounts of animal manure applied were established considering the N concentration.

Average grain and cereal yields (corn, beans, and wheat) over 12 years increased significantly in soils with PD, PS, CS, and FM applications, relative to the control (Figure 4a). On average, corn yield (10 crops) grown on soils with FM, PS, CS, and PD applications were 1.8, 2.2, 2.4, 2.0, and 2.5 times higher, respectively, relative to yield on the control soil. The yield of beans grown on soils with MF, PS, CS, and PD applications increased by 1.5, 2.7, 3.0, and 4.1, times respectively, relative to the control. The highest wheat yield was observed in soils grown with MF applications followed by PS, PD, and CS with increments of 2.6, 2.3, 2.3, and 2.2 times, respectively, relative to the control. More information on the grain yield responses of the crops in this study can be seen in Bacca et al., Marchezan et al., and Ferreira et al. [13]. The highest cumulative P exports (sum of corn, bean, and wheat exports) over 12 years were observed in crops grown on soils subjected to manure and mineral phosphate fertilizer applications (Figure 4b). Exports of P by crops grown on fertilized soils averaged 150, 143, 119, and 85 kg P ha^{-1}, higher than that observed in the control soil (Figure 4b and Table 2).

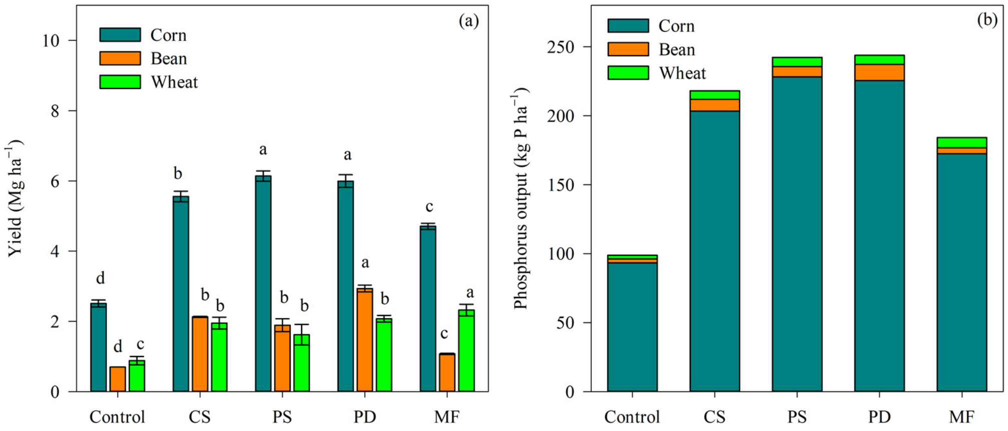

Figure 4. Response of the production of grains of corn, beans, and wheat (**a**) and dry mass maize, beans, wheat, oats, and turnips (**b**), depending on the successive applications cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF). The results represent an average of 12 years (2004–20016) of crop rotation management with 18 treatment applications. Corn was cultivated in 10 seasons, while beans and wheat were grown in the 2012/2013 and 2014 cropping seasons, respectively. For cover crops, oats were grown for 8 cropping seasons and O. Radish in three cropping seasons (2005, 2007, and 2009). Different letters between treatments at the same depth indicate significant difference (Scott–Knott test at 5%). Standard deviation is presented on the surface of the bars.

The applications of MF, CS, PS, and PD promoted the accumulation of 18, 42, 48, and 100 kg P ha^{-1} year^{-1} (Table 2). The mass balance calculations showed that between 41 and 72% of the total P added by manure and MF was accumulated in the 50 cm layer. Furthermore, most of the P added in the cropping system can be explained by soil storage, exports by grains and crop residues (98, 89, 89, and 77%, for treatments CS, PS, PD, and MF, respectively). The P balance efficiency was highest in the soil with FM, at 40%, decreasing to 30, 29, and 19% in soils with CS, PS, and PD applications, respectively.

4. Discussion

4.1. Soil P Balance

Additions of CS, PS, PD, and MF in a no-till system for several years increased the amounts of P in the soil, with its vertical movement proportional to the amount of P applied by each source [31–33]. The difference in the increment in the amounts of total P compared to the control was observed in all treatments up to 40 cm depth, with a higher proportion in the upper soil layer (0–20 cm). This happened because, in the no-till farming system, the soil is not disturbed, and the fertilizer sources are applied on the surface. In addition, the rapid adsorption of the phosphate ion by the functional groups of reactive soil particles decreases the availability of P in solution, preventing it from moving to depth [34,35].

The fraction with which P was accumulated in the soil differed among treatments and in depth, with preferential accumulation of inorganic P in PD. In the soil layers with PS, CS, and MF applications, the accumulated amounts of organic and inorganic P were similar. The accumulation of P in the inorganic fraction in PD may have happened, especially, because of the higher concentration of P per kilogram of PD (Figure 1). This is because PD waste has animal feces and urine and food leftovers, which are deposited for several months in pig breeding stalls [36]. Therefore, PD has a high concentration of nutrients, among them, P. Moreover, the concentration of inorganic P in swine manure is higher in relation to the manure from other livestock farms, due to the absence of the enzyme phytase in the digestive tract, which prevents the degradation and utilization of compounds that contain P, excreting in the feces the vast majority of the P consumed via grains [37,38].

The higher amounts of organic P observed in the topsoil layers with CS, PS, PD, and MF applications can be explained by the higher biomass production of the soil cover plant species and grain crops [32]. Plants during their growth and development absorb P only in the orthophosphate (Pi) form and at the end of the crop cycle the P, after being incorporated into the tissues, returns to the soil in the form of organic P [32,39,40]. The replacement of P to the soil happens because of deposition and decomposition of the aerial part of the crops [35], as well as by root senescence [41].

The successive Inputs of P by CS, PS, and PD significantly increased the labile P fractions, especially the concentrations of weakly bound P (P-NH4 Cl) in the topsoil (0–4 and 4–10 cm), with the highest values observed in the soil with PD, because of the greater amount of P added (Figures 1 and 5). The increase in P-NH4 Cl concentrations in the topsoil layers may potentiate possible transfers of soluble P or P adsorbed to functional groups of solid particles in the surface runoff solution [42,43]. Because of the high affinity of the phosphate ion to reactive soil particles and the small movement of P in the profile observed in this study, we believe that it is unlikely that P was transferred by leaching [23,44,45].

In a study conducted in southern Brazil, Ceretta et al. [44] observed losses ranging from 6.4 and 14.4 kg P ha^{-1} by surface runoff, in an experiment conducted on sandy soil (53% sand and 4% slope), subjected to applications of doses from 20 to 80 m^3 ha^{-1} of liquid pig manure for 8 years. In the same experiment, Tiecher et al. [45] quantified during 6 crops (2.5 years) the forms of P transferred by leaching. The authors concluded that less than 0.2 kg P ha^{-1} year^{-1} applied at a dose of 80 m^3 ha^{-1} of liquid pig manure was leached. When compared to P losses by surface runoff, leaching losses are economically and agronomically negligible. However, due to the numerous precipitations and high volume of rainfall in subtropical climate, which may increase the water flux in the profile, the leached P concentrations may exceed the limits recommended by the environmental legislation, especially in soils with high P contents. Thus, although the amounts of P transferred by leaching are small, these losses can have a significant environmental impact in groundwater contamination, especially when the water table is shallow [45–48].

Soil P accumulation in the inorganic fractions and P movement in the soil profile are similar to other studies that evaluated the effects of long-term P fertilization by phosphate fertilizers and different animal manure sources [28,49–52]. Studies conducted on soils and climatic conditions similar to those of the present study (<20% clay), with a history of swine manure application, also observed a movement of labile P greater than 30 cm [29,53,54].

While in more clayey soils (>40% clay), P movement observed in experiments with long application history was less than 20 cm [28,55]. The lower migration of P to deeper layers in clayey soils compared to sandy soils is directly related to the higher capacity of P adsorption in soils, especially in free functional groups on the surfaces of clays and Fe and Al oxides and hydroxides [25,56].

Most of the P added by CS, PS, PD, and MF was accounted for by mass balance calculations. However, it was not possible to calculate 0.8, 8.0, 12, and 10 kg P ha^{-1} year^{-1} in soils that received applications of CS, PS, PD, and MF, respectively (these values correspond respectively to 1, 13, 10, and 22% of the P added annually). The highest amount of unaccounted P in the PD soil (144 kg P ha^{-1}) may be related to the highest concentration of weakly bound P among the treatments evaluated, which indicate potential P losses (Table 1 and Figure 5).

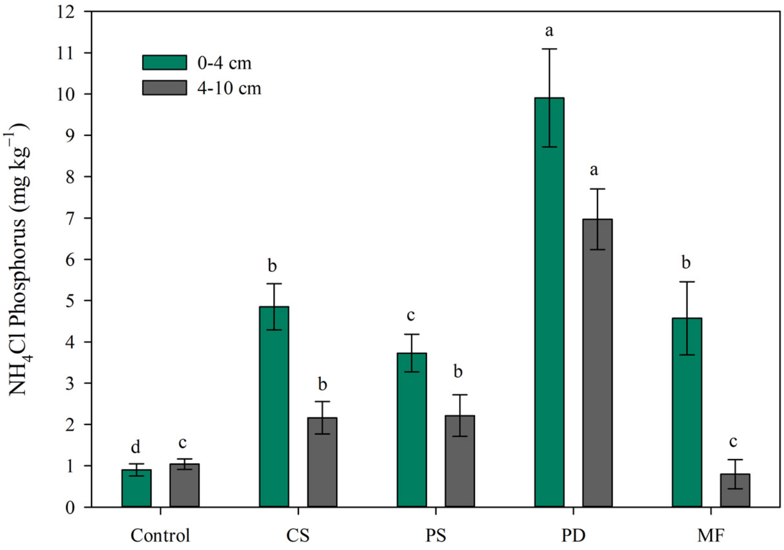

Figure 5. Representation of weakly bound P extracted with 1 mol L^{-1} NH_4Cl after long-term animal slurry and mineral fertilizer additions in the topmost soil layers (0–10 cm). Substantial increases in this labile fraction pose potential environmental risks, mainly following soil erosion by runoff. Vertical bars represent standard deviation of averages (McDowell et al. [46]). Different letters between treatments at the same depth indicate significant difference (Scott–Knott test at 5%).

Accumulated P in the soil represented the main P compartment in this agricultural system, even in a sandy soil (10.8% clay), which has low P adsorption capacity [53,55]. In the soils subjected to the additions of CS, PS, and PD, this may have happened because of the constant additions of P many times higher than the demands of the crops. Added to this, the maintenance of cultural residues on the surface and the cultivation of cover crops over the years may have collaborated in the cycling and maintenance of P in this compartment. As discussed above, the transfer of P by leaching may be insignificant in this soil and, therefore, 2.5 to 23% of the P inputs, which could not be accounted for by the mass balance, may represent P lost by surface runoff.

4.2. Environmental and Agronomic Implications

The applications, especially of liquid (CS and PS) and solid (PD) animal manure, significantly increased the P content of the soil compared to the control, causing concern about the continued use of these nutrient sources. Over 12 years, the amounts of animal manure were defined considering the efficiency index of each manure, the concentration of N, and the need of each crop, as established by the regional recommendation on the use of animal manure [27]. Probably, these variables are efficient in defining the amount of manure in the first years of applications. However, when the manure is applied repeatedly over several years, it can greatly increase the P contents in the soil, exceeding the extraction capacity of the plants, causing significant accumulation of this nutrient in the soil [28,32,49]. A study by Gatiboni et al. [25] established the environmental safety limits for P in soil in the southern region of Brazil. The method is based on the concentrations of bioavailable P in soil (Mehlich-1 extractor) in the 0–10 cm layer, soil P adsorption capacity, and clay content, according to the equation: P threshold (mg P kg^{-1}) = 20 + clay content (%). When employing this prediction for the soil of the present study (clay content of 10.8%), the P threshold (30.8 mg P kg^{-1}) was exceeded by 79, 135, 148, and 202% for MF, PS, CS, and PD applications, respectively. These values correspond to the significant increment in the concentrations of weakly bound P (NH4 Cl-P Figure 5) in the topsoil layer (0–4), representing potential risk of P losses by surface runoff [23,26]. In the absence of measures to mitigate excess soil P, ongoing applications could escalate environmental hazards, compromising surface water quality [24,56,57].

The grain yield data obtained for corn, beans, and wheat over the years showed the increase in productivity achieved through applications of CS, PS, and PD. The improvement in soil nutrient availability demonstrates the potential of using animal manure as a source of fertilizer [13,58]. However, ongoing monitoring of critical soil P levels is essential. This study's P mass balance results indicate an imbalance in P additions among treatments, resulting in significant P accumulation in the soil compartment and low P efficiency for plant uptake [48,59,60]. In addition, the differences in the amounts of P added by the manure over the seasons may have been a consequence of the composition of the manure, especially the effect of dry matter concentration per kilogram of manure added. These factors contributed to the excessive addition of P in the PD treatment, thus decreasing the overall P mass balance efficiency of the culture system to unsatisfactory levels. These results raise concerns not only about the amount of animal waste that can be applied per area, but also about the chemical composition of the added manure in agricultural areas.

Continued additions of P and the accumulation of P legacy in soils after long periods of animal manure applications in the same areas can present a serious environmental risk, as was widely demonstrated in this long-term study. Similarly, applications of mineral phosphate fertilizers (MF) also implied a worrisome accumulation of P in the soil, especially in the topsoil. However, the amounts of plant-available P (Mehlich-1 extractor) is within the adequate range (30–50 mg P kg^{-1}), requiring annual maintenance fertilization. We observed that the addition of MF increased the fractions of P not available to plants, with 51% of the total P in the stable and residual form in the surface layer (Figure 2d). This may have happened because of the high affinity of the phosphate ion that is rapidly adsorbed to the surface functional groups of reactive soil particles [34]. This effect was more pronounced in soil with MF applications, probably because of the lower pH value, caused by acidification, caused by the release of H+ coming from the solubilization of N fertilizers [61,62]. The decrease in pH causes the protonation of functional groups of reactive soil particles, thus increasing the binding energy of P with the soil solid phase, increasing the more stable P fractions [63]. Low efficiency and accumulation of P in the surface layers in soils with mineral phosphate fertilizer additions is very common in most highly weathered soils.

Considering the obtained results, research is needed to find strategies to mobilize recalcitrant P fractions present in the soil and decrease the dependence on constant phosphate fertilizations [40,64,65].

5. Conclusions

The additions of PD, PS, CS, and MF to the soil over 12 years caused the accumulation of 100, 48, 42, and 18 kg P ha^{-1} $year^{-1}$, respectively.

All P sources caused significant P accumulation in the surface layers, especially in the inorganic P fraction. However, in the soil with PD, the P moved up to 40 cm and with applications of CS, PS, and MF, the movement was observed up to 30 cm.

Soils with animal manure applications, especially PD, have a worrisome potential for P transfer. Thus, threshold levels of P should be constantly monitored, and the fertilizer recommendation system should consider the history of nutrient source applications in the cropping system to establish the need and dose for a new application of a P-containing source.

The P mass balance revealed that 77 and 98% of P input by animal manure and MF was accounted for in grain exports (17–34%), soil storage (41–72%), and post-harvest residues (<1%), with the remaining 2–33% unaccounted for because of transfers by surface runoff. This reinforces the need for conservation farming practices to avoid runoff and soil losses.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/agriculture13091762/s1, Table S1. Distribution of P pools in the soil profile after 18 successive applications (2004–2016) of cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF). Data presented are means (n = 4); Table S2. Quantities of inorganic, organic, and total P (kg P ha-1) accumulated in the soil profile after 18 successive applications (2004–2016) of cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF). Data presented are means (n = 4); Table S3. P contents in the different forms of Hedley fractionation and sum of the P contents extracted from the soil profile after 18 successive applications (2004–2016) of cattle slurry (CS), pig slurry (PS), pig deep-litter (PD), and mineral fertilizer (MF). Data presented are means (n = 4).

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