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Abstract: The effect of organic matter (OM) on soil phosphorus (P) sorption is controversial, as there is still no clear answer whether organic matter inhibits or increases P sorption. Despite the great need for renewable sources of available P and OM in agricultural soils, little is known about the interaction between P and dissolved organic matter (DOM) in natural soil systems. The aim of this research was to uncover if and how soil saturation with DOM derived from different types of abundant agricultural wastes (cattle manure, horse manure, biogas digestate, compost) affects the phosphate sorption. We examined the P sorption process in control and DOM-saturated sandy soils. The results indicated that OM introduced with agricultural waste did not always reduce P sorption, but certainly had an effect on impairing P fixation, and thus, may result in potentially greater P mobility in the soil, including P availability. Among these waste materials, DOM from horse manure had the most positive effect on P mobilization; thus, horse manure—if available—is recommended for spreading on soils with low P mobility.

Keywords: organic waste; organic matter; animal manure; compost; phosphorus retention; sorption isotherms



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1. Introduction

Research on the different types of natural and exogenous organic matter (OM) and their fractions and effects on phosphorus (P) retention in soil has become a consistent element within issues of global concern [1–9]. The progressive degradation of arable soils, i.e., drought, contamination, soil organic matter losses or the scarcity of plant nutrients (in particular P)—has stimulated strategies to utilize infertile and even degraded soils. The sustainable use of soil resources has become an important global problem in response to the increased demand for food production [10,11].

Phosphate rock is included in the Critical Raw Materials List by the European Commission in 2014, therefore reducing agriculture's reliance on mineral fertilizers and making greater use of renewable sources of macronutrients. At the same time, OM is an emerging direction for agriculture [12,13]. Crop residues, animal manures, composts, and other organic materials are frequently used to improve soil quality and enhance soil organic matter levels. An additional benefit to the agricultural ecosystems of these soil amendments may be an increase in the bioavailability of soil P. However, the impacts of these types of exogenic organic matter still need extensive studies [14,15].

The behaviour of P in soils remains problematic from both the agronomic and environmental perspectives. On the one hand, ensuring the sufficient availability of P poses particular problems regarding the very low efficiency of P mineral fertilization, rising up only 10–20% [16]. The frequent replenishment of bioavailable P forms is necessary but, at the same time, may cause negative environmental impacts. This leads to a continuous accumulation of strongly bound P accompanying fertilization

and the risk of its mobilization and release from soil into waterways once a certain safe value of soil P saturation is exceeded [17–19]. This critical value, known in the literature as a *change point* or *threshold point*, is documented in many studies [20–23]. The P sorption plays a key role in governing the P behaviour in the soil environment, including its mobility. This process occurs mainly on the mineral surfaces of soil colloidal particles having active sorption sites for P. A particularly important role in this process is attributed to metal oxides and hydroxides commonly found in soils (especially iron (Fe) and aluminium (Al), the activity of which is high, particularly under acidic conditions). At a soil pH above 7, calcium (Ca) ions can precipitate with phosphate anions, forming insoluble compounds which restrict plant P nutrition [24–26]. Silicate clay minerals (e.g., kaolinite, montmorillonite, illite) also participate in P sorption in soils; this occurs mainly at their edges via ligand exchange with surface OH⁻ groups [27–29].

Organic matter significantly contributes to soil sorption properties and also may affect phosphate sorption in soil as a consequence of various interactions with mineral soil components [30–34]. The great diversity in the origin, types, forms as well as fractions of OM makes it very difficult to obtain unambiguous results. Anion exchange between the simple coordinated OH groups of the OM is one important mechanism for the formation of strong organo-mineral associations, e.g., Fe-O-C bonds [35]. Ligand exchange between reactive inorganic hydroxyls (the OH groups of Fe-, Al-, Mn-hydroxides, and the edge sites of phyllosilicates) and organic carboxyl and phenolic OH groups is restricted to acid soils rich in minerals with protonated hydroxyl. OM forms strong complexes with Al and Fe oxides via ligand exchange, particularly in acidic soils [35,36].

It is generally believed that OM affects the P (ad)sorption in soils by competing for the same sorption sites [17,33]. Organic matter thus blocks P sorption sites, resulting in weaker P binding to soil particles. Therefore, an increase in the concentration of the available P forms in the soil solution, along with an increasing OM content, has been observed [4,24,29–32]. Consequently, this may be beneficial for agricultural needs but also, in certain conditions, can contribute to P leaching from the soil and promote the deterioration of surrounding water bodies through eutrophication.

There is a wealth of evidence for P sorption inhibition by the competitive effects and chelation interactions of low-molecular-weight organic acids (LMWOAs), which are common in soils [14,31,37–43]. LMWOAs are mainly derived from the decomposition of OM and the secretion of plant roots and microbes; they include oxalic acid, citric acid, tartaric acid, malic acid, succinic acid, salicylic acid, p-hydroxybenzoic acid, and maleic acid, among others [31]. All of those LMWOAs molecules are components of dissolved organic matter (DOM)—the water-soluble fraction of organic matter—which can be considered as the most active part of OM. This active part of OM contains components that are readily released into the soil solution, for example during rainfall events [30]. Dissolved organic matter is a complex mixture of organic compounds: humic and fulvic acids, amines, polysaccharides, and numerous other C compounds [17]. The ability of DOM to compete with P is mainly determined by the stability of LMWOA [33], and the quality and quantity of DOM is probably the main factor defining the influence of OM on the course of P sorption in soil [44].

The most popular studies using model organic compounds have also led to the suggestion that natural DOM could affect the P availability in soils through the competition between DOM and P for mineral adsorption sites [33,45]. However, some studies have found little or no evidence to support an effect of DOM on P sorption and the amount of phytoavailable P under realistic soil conditions [33,41]. Ohno and Crannell [46] have shown that animal manure-derived DOM was less capable of forming complexes with Al and Fe oxides and that they compete with P for adsorption sites in comparison to those from green manure. They found that green manure-derived DOM inhibited P sorption as a function of DOC concentration, whereas animal manure-derived DOM enhanced P sorption or had no significant effect [46]. The ability of green manure-derived DOM to inhibit P sorption was attributed to the lower molecular weight of these compounds in

comparison to animal manure-derived DOM. Other researchers also have shown that the extent of inhibition is closely related to the chemical structure of both the DOM and the sorbing surface [47]. There is also evidence that the formation of DOM–metal complexes can reduce the number of available sorption sites and alter surface charge chemistry (i.e., increase negative surface charge), thereby enhancing dissolution reactions [33]. In contrast, mineral-sorbed or particulate organic matter can also increase P sorption by increasing the number of sorbed cations available to form cation bridges with P [33]. Furthermore, the results with DOM isolated from crop residues, animal manures, and compost showed that the DOM initially leached or dissolved from these amendments was not likely to outcompete orthophosphate for mineral sorption sites [14]. Due to P bonding to FeOOH through a strong, inner-sphere mechanism, and DOM associating through weaker outersphere electrostatic interactions or multiple hydrogen bonds, P outcompetes DOM for adsorption sites at mineral surfaces [14]. Chase et al. [14] also presented results supporting their earlier studies, showing that only aromatic molecules >600 Da can compete with orthophosphate for mineral adsorption sites. Other findings [48] have shown that organic compounds with two or more functional groups are more capable of exerting a greater

Hence, the role of organic matter in P sorption is still inconclusive and unclear. Some results show no apparent effect of OM on P, but some also indicate an increase in P sorption under the influence of OM in soil [17,33,39,44,45,49–51]. These contradictory results could have various reasons. For instance, they could be related to the magnitude of the P adsorption capacity of soil, which depends also on the type of organic matter, soil genesis, or could be an average effect obtained in such a complex system as soil. On the one hand, the complex structure of OM, containing a variety of functional groups, gives many possible sorption sites for P. On the other hand, it can also block P sorption sites and compete with this component for the same sorption capabilities.

variety of interactions in soils than organic compounds carrying only one functional group.

The disagreements concerning the effect of OM on P sorption in the literature and the scarcity of studies on the effect of DOM derived from various agricultural waste materials on P sorption in natural soils [33,41,52], as well as the need for renewable sources of available phosphorus and organic matter in agricultural soils, calls for research on the interactions between P and DOM in natural soil systems.

The objective of the study was to clarify whether, and explain how, soil saturation with DOM derived from different types of abundant agricultural wastes (cattle manure, horse manure, biogas digestate, compost) affects the phosphate sorption. We assume that DOM introduced into P-fixing soils will interact with mineral surfaces, mainly with the active sites of Fe and Al oxides, thus blocking the potential P sorption sites and lowering the soil's capacity for P sorption. We also hypothesize that the different chemical composition of DOM would have a distinctive effect on P sorption due to the type of interactions between OM, Fe/Al oxides, and P. The overall aim of the research was to develop a practical approach to overcome the P-fixation in certain Northeastern European soils.

2. Materials and Methods

2.1. Materials

2.1.1. Soil Samples

This research was conducted on light textured soil materials characterized by different basic properties and diverse land uses. Soil 1 (S1)—classified as Brunic Arenosols, according to the FAO-World Reference Base [53]—located in central Poland (Spała Forest District), occurring in the fresh coniferous forest habitat (with *Pinus sylvestris* and *Picea abies*, with *Quercus petraea* in the undergrowth). Soil samples were taken from the *sideric* horizon (Bvs). This material showed also the features of *spodic* horizon. S1 is a sand-textured material characterised by pedogenic accumulation of immobile humic complexes with Al and Fe oxides and the presence of coatings of mineral grains composed of Fe and Al (not associated with humus), which were formed by silicate weathering and low humic acid synthesis. The features related to the illuvial accumulation of Fe and Al oxides as

a result of the podzolic process in the soil horizon are also visible in S1. Soils 2 and 3 (S2 and S3)—classified as Eutric Arenosols [53]—arable loamy sand-textured soils located in north-eastern part of Germany (S2—Prohn; S3—Poppendorf). Under natural conditions these soils have been podzols, but the original litter layer and leached A horizons have been incorporated into the tilled A horizon. Beginning in the early 1990s, deeper ploughing with stronger machinery incorporated the previous oxide-rich spodic horizon material into the surface Ap horizon, causing the problem of phosphate fixation. This Ap material has been sampled. These three soils represent large areas of the Weichselian Glaciation-derived lowlands in Northern, Central, and Eastern Europe, which currently are used for arable (mainly grain crops, special crops, and potatoes) and forestry.

2.1.2. Experimental Organic Materials and Soil Incubation

The different types of organic matter derived from agricultural waste materials—compost (CPT), biogas digestate (BD), cattle manure (CM), and horse manure (HM)—have been acquired locally and introduced into the soil in the form of dissolved and colloidal organic matter (abbreviated as DOM). The DOM was extracted using cold tap water at a ratio of 1:5 (*w*:*v*) of waste to water. After 24 h incubation, the suspensions were passed through a 1 mm sieve and were filtered through Macherey-Nagel, MN 616 G-filters. We have thus obtained a large (several kilo grams) mass of well-homogenized organic matter fraction from the waste materials. This fraction can be assumed as naturally the most active in soil, as it has the ability to be rapidly released into solution under natural conditions, such as during rainfall. Next, the air-dry well-mixed sandy soil samples were placed in beakers and slowly filled up with the dissolved organic matter until the applied solutions reached saturation levels in each soil. To minimize treatment variability, the soil samples were repeatedly and progressively wetted with DOM solution, which was introduced in small portions with simultaneous and thorough mixing of the soil. Repeating the addition several times resulted in relatively homogeneous, mixed samples, in which the enrichment in organic matter we verified by elemental analyses (C and N concentrations). Our intention was to fill the spaces between the soil particles with the solution of DOM to allow the introduced organic matter to occupy the active sorption sites at the soil particles, mainly derived from iron and aluminium oxides and hydroxides. The aim was to block potential sorption sites for P or even replace some P from those sorption sites with the introduced organic matter (DOM), which competes with P for the same sorption sites [33]. Thus, we aimed to lower the P sorption capacity of soil and, at the same time, to increase P availability to plants.

After saturation, the samples were incubated at 70 °C (relatively high temperature to prevent microbial decomposition of organic matter in samples), and then, the soil saturation procedure was repeated several times in the same way in order to achieve homogeneity of DOM saturation within the treated sample. After the drying of the samples, and testing the C contents as a measure of the organic matter addition, bulked samples were created, in which all measurements were made in three replicates. The final amount of DOM added into each soil is shown in Table 1. These soil samples saturated with DOM, derived from four types of agricultural waste materials, were used in P sorption studies in comparison to unamended soils, which were the reference (control) materials.

Table 1. The quantities of DOM additives introduced into the soils (in mL kg⁻¹ of soil)—the experiment variants.

Soil Sample/ DOM Type	No DOM (Control)	DOM-CPT * (mL kg ⁻¹)	DOM-BD (mL kg ⁻¹)	DOM-CM (mL kg ⁻¹)	DOM-HM (mL kg ⁻¹)
S1	0	300	300	300	300
S2	0	520	560	480	520
S3	0	400	480	480	480

* Abbreviations: DOM—dissolved organic matter; CPT—compost, BD—biogas digestate, CM—cattle manure, HM—horse manure.

2.2. Analytical Methods

2.2.1. Basic Soil Chemical Characteristics

The following basic properties were determined in soils under study (in fraction < 2 mm): soil texture by the hydrometric method [54]; total organic carbon content (C) using a CS-MAT 5500 analyser (Ströhlein GmbH & Co., Kaarst, Germany, currently Bruker AXS Inc., Madison, WI, USA); total nitrogen (N) by the Kjeldahl method using a Buchi Labortechnik GmbH N analyser (BÜCHI Labortechnik AG, Postfach, Switzerland); pH was determined potentiometrically in 1 m KCl solution at 1:2.5 soil:solution ratio. The total content of Al, Fe, and P extracted after digestion in aqua regia (3:1 ratio of concentrated HNO₃ and HCl acids) with microwave-assisted digestion were measured by inductively coupled plasma optical emission spectroscopy (ICP-OES, Perkin-Elmer Optima 8300 DV, Waltham, MA, USA [55]). The amounts of non-crystalline and poorly crystalline oxides of Al (Al_{ox}), Fe (Fe_{ox}), and P (P_{ox}) were extracted with the solution of oxalic acid and ammonium oxalate at pH 3.0 [56] and measured with the ICP-AES analyzer. Then, the P sorption capacity (PSC) and degree of P saturation (DPS) of soil were calculated according to the following equations [57–59]:

$$PSC = 0.5 ([Fe_{ox}] + [Al_{ox}])$$
 (1)

$$DPS = ([P_{ox}]/PSC) \ 100 \ [\%]$$
(2)

where PSC and P_{ox} , Al_{ox} , and Fe_{ox} are expressed in mmol kg⁻¹.

All determinations were conducted in three replicates. The average results and standard deviation from the mean are shown in the tables and figures.

2.2.2. P Sorption Isotherms

Phosphorus sorption process was carried out in soils not saturated (S1, S2, S3) and saturated with DOM derived from waste organic materials: compost (S1-CPT, S2-CPT, S3-CPT), biogas digestate (S1-BD, S2-BD, S3-BD), cattle manure (S1-CM, S2-CM, S3-CM), horse manure (S1-HM, S2-HM, S3-HM). For this purpose, a sorption isotherm was constructed based on 10 doses of phosphorus: 0.0, 0.5, 1.0, 1.5, 2.0, 3.5, 5.0, 10, 15, 20 mg P·dm⁻³ in the form of KH₂PO₄ with 0.01 mol·dm⁻³ CaCl₂ as a background solution [49]. All the P doses were introduced into 1 g of soil in volume of 25 mL, in 3 repetitions. The samples were equilibrated for 24 h on an end-over-end shaker, then centrifuged for 15 min at $4500 \times g$ and filtered. Phosphorus in the supernatants was determined by inductively coupled plasma optical emission spectroscopy (ICP-OES, Perkin-Elmer Optima 8300 DV, Waltham, MA, USA).

2.2.3. Statistical Analyses

The amount of P (ad)sorbed (Q) was calculated from the difference between the amount of P added into the sample (P dose) and the amount of P in the supernatant of equilibrated solution according to Equation (3), where Q is the amount of P sorbed (mg kg⁻¹), C_P—the initial concentration of P in solution; P added (mg·dm⁻³); C_{eq}—the concentration of P after 24h-equilibration (mg·dm⁻³).

$$Q = C_P - C_{eq} \tag{3}$$

The Freundlich (4) and Langmuir (5) isotherm equations were applied to describe P adsorption to the soils:

$$Q = K_f C_{eq}^{nt}$$
(4)

$$C_{eq}/Q = C_{eq}/Q_{max} + 1/K_L Q_m$$
⁽⁵⁾

where Q is the amount of P adsorbed to soil at equilibrium concentration (mg kg⁻¹), K_f is the Freundlich unit capacity, C_{eq} is the P concentration in the equilibrated solution (mg dm⁻³), n_f is the Freundlich exponent describing the nonlinearity of the adsorption,

 Q_m is the maximum adsorbed P concentration (mg kg⁻¹), K_L is a constant related to the binding strength of P at the adsorption sites (dm³ mg⁻¹).

The maximum adsorption buffering capacity (MBC, $dm^3 kg^{-1}$) was calculated on the basis of the following:

$$MBC = K_L \cdot Q_m \tag{6}$$

Phosphorus adsorption isotherm indices (K_L , K_f , n_f) and maximum adsorbed P concentration (Q_m) were calculated using the linearized form of the isotherm. The results obtained were fitted to the isotherm models. The standard error (SE), sum of SE, and variance were calculated to determine R-squared values to evaluate the fit of the results to the sorption models. Adsorption isotherm data were fitted using the non-linear solver function [60] and also the normal linearization method. Fitted parameters using both methods were compared based on goodness of fit measures and the best-fit parameters are finally presented here. It is noteworthy to mention that solver results were found to be more appropriate compared to the linearization technique.

The results obtained were statistically verified using Statistica 13 software [61]. The normal distribution of data was verified using the Kołomogorov–Smirnov test and the chi-square test. The majority of results did not indicate the nature of the normal distribution. Therefore, non-parametric tests were used for further tests of the data. The differences between groups in the experimental variants were tested using the ANOVA Kruskal–Wallis test. Correlation analysis between data was performed using Spearman's correlation coefficient. The analysis of principal components (PCA) was conducted and the relationships between the selected parameters were expressed as a correlation coefficient at a statistically significant level of p < 0.05.

3. Results and Discussion

3.1. Soil Chemical Characteristics

3.1.1. Initial Properties and P Status

The soils S1–S3 differ in their chemical properties. Among the main factors contributing to the different characteristics of these soils are the type of bedrock and land use. Soil S1 (B horizon) is characterized by acidic reactions, typical for the conditions found in coniferous habitats, conducive to the podzolization process. This is a soil of low quality, formed from sand as a bedrock. The low soil quality is reflected by its low C content, wide C:N ratio, and narrow C:P and N:P ratios. The arable soils (S2 and S3) taken from the surface layer (Ap) have markedly different properties, as evidenced by higher pH, C, N, and P values, a narrower C:N ratio, and wider C:P and N:P ratios, most likely originating from arable use, liming, and fertilization. Also, these soils have higher total Al and Fe contents (Table 2). The Al_{ox} and Fe_{ox} contents of the soils are shown in Table 3, and the PSC was estimated from it, which is between 43 mmol kg⁻¹ in S1 and 55 mmol kg⁻¹ in S2. These values are narrow in a relatively low range as compared to data from northwestern Germany [62].

When the phosphorus bound to Fe_{ox} and Al_{ox} (P_{ox}) is taken into account, the degree of phosphorus saturation (DPS) can be estimated (Table 3). The DPS is frequently used as an indicator of the threshold for environmental safety, which, if exceeded, can mean a significant increase in the concentration of the soluble forms of P in the soil and increased environmental risk associated with P mobility. It has been assessed that this threshold usually occurs at a value of about 25% DPS in sandy soils with high groundwater levels [20–23,37]. In S1, the poor forest soil, the DPS is low (21.3%), while in arable soils (S2 and S3) it reaches higher values of 37–42%, which means that the "change point" might have been already exceeded. Furthermore, at ongoing P accumulation, downward movement towards groundwater and DPS >25% throughout the soil profile of these soils may pose a risk for freshwater pollution with phosphate and eutrophication.

Semula	pН	C *(%)		C *(%) N (%)		Р	P (%) A		Al (%) Fe (%)		(%)	CN	C D	N D	C 11	0.5
Sample	\tilde{H}_2O	x	SD	$\overline{\mathbf{x}}$	SD	$\overline{\mathbf{x}}$	SD	$\overline{\mathbf{x}}$	SD	$\overline{\mathbf{x}}$	SD	CIN	CP	N:P	C:AI	C:Fe
S1	4.81	0.36	0.01	0.03	0.007	0.02	0.0005	0.37	0.008	0.26	0.000	13.9	15.3	1.3	0.98	1.37
S1-CPT	5.21	0.39	0.02	0.02	0.000	0.02	0.0000	0.33	0.015	0.24	0.008	19.5	17.3	0.9	1.17	1.61
S1-BD	6.28	0.49	0.01	0.05	0.007	0.03	0.0058	0.35	0.006	0.25	0.013	10.0	18.6	1.9	1.40	1.98
S1-CM	6.17	0.59	0.02	0.03	0.001	0.02	0.0002	0.40	0.012	0.28	0.003	17.5	23.8	1.2	1.47	2.13
S1-HM	6.11	0.46	0.02	0.03	0.002	0.04	0.0009	0.40	0.005	0.27	0.003	18.0	11.1	0.7	1.16	1.72
S2	6.03	1.29	0.03	0.12	0.021	0.05	0.0005	0.58	0.029	0.60	0.008	10.6	25.4	2.4	2.22	2.14
S2-CPT	6.32	1.32	0.04	0.11	0.014	0.05	0.0001	0.48	0.001	0.53	0.005	11.7	27.5	2.3	2.72	2.50
S2-BD	6.55	1.40	0.05	0.14	0.014	0.05	0.0008	0.48	0.036	0.55	0.034	10.3	27.4	2.7	2.90	2.55
S2-CM	6.79	1.59	0.05	0.14	0.021	0.05	0.0002	0.51	0.021	0.56	0.008	11.4	30.7	2.7	3.13	2.84
S2-HM	6.76	2.23	0.05	0.17	0.021	0.07	0.0011	0.45	0.019	0.47	0.000	12.8	33.4	2.6	4.97	4.78
S3	6.61	1.07	0.05	0.11	0.021	0.05	0.0001	0.48	0.018	0.54	0.003	9.7	21.4	2.2	2.21	1.97
S3-CPT	6.66	1.14	0.11	0.11	0.007	0.05	0.0009	0.47	0.043	0.50	0.031	10.4	23.5	2.4	2.40	2.26
S3-BD	6.72	1.21	0.04	0.12	0.014	0.05	0.0018	0.45	0.019	0.50	0.006	10.1	25.4	2.5	2.72	2.40
S3-CM	6.83	1.25	0.05	0.11	0.014	0.05	0.0003	0.54	0.074	0.53	0.024	11.0	26.1	2.3	2.31	2.37
S3-HM	6.80	1.70	0.06	0.15	0.014	0.07	0.0002	0.51	0.005	0.51	0.007	11.4	25.4	2.2	3.31	3.34

Table 2. pH and basic chemical properties of unamended soils (initial properties—S1, S2, S3) and amended with different types of DOM derived from compost (S1-CPT, S2-CPT, S3-CPT), biogas digestate (S1-BD, S2-BD, S3-BD), cattle manure (S1-CM, S2-CM, S3-CM), horse manure (S1-HM, S2-HM, S3-HM).

* Explanation: C-total organic carbon; N-total nitrogen; P-total phosphorus; Al-total aluminum; Fe-total iron.

Table 3. Content of acid ammonium-oxalate extractable Fe, Al, P (Fe_{ox}, Al_{ox}, P_{ox}); phosphorus sorption capacity (PSC) and degree of phosphorus saturation (DPS).

Sample	Fe (mmol	Fe _{ox} (mmol kg ⁻¹)		Al_{ox}		ox $ \mathbf{k}\sigma^{-1} $	PS (mmol	5C kg ⁻¹)	DPS		
Sample	x	SD	x	SD	x	SD	x	SD	x	SD	
S1	6.71	0.68	14.45	0.40	2.25	0.14	10.6	0.54	21.3	0.20	
S1-CPT	5.80	0.01	13.25	0.11	2.08	0.16	9.5	0.05	21.9	1.54	
S1-BD	5.75	0.66	13.18	1.35	2.04	0.13	9.5	1.00	21.5	0.91	
S1-CM	5.91	0.67	13.54	1.58	1.89	0.11	9.7	1.12	19.4	1.15	
S1-HM	5.88	0.23	14.00	0.49	4.00	0.10	9.9	0.36	40.2	0.47	
S2	15.15	0.15	15.41	0.28	6.44	0.04	15.3	0.21	42.2	0.34	
S2-CPT	14.28	0.36	13.12	0.12	5.85	0.09	13.7	0.12	42.7	0.30	
S2-BD	13.82	0.74	12.80	0.49	5.74	0.15	13.3	0.61	43.1	0.86	
S2-CM	13.98	0.48	12.68	0.31	5.81	0.41	13.3	0.09	43.6	3.34	
S2-HM	13.02	0.24	12.37	0.12	8.39	0.04	12.7	0.18	66.1	0.62	
S3	13.91	1.07	11.44	0.37	4.70	0.22	12.7	0.72	37.1	0.39	
S3-CPT	12.43	0.13	10.33	0.05	4.59	0.06	11.4	0.09	40.3	0.18	
S3-BD	12.24	0.22	10.42	0.14	4.59	0.06	11.3	0.04	40.5	0.36	
S3-CM	11.69	1.86	9.79	1.49	4.10	0.48	10.7	1.67	38.2	1.46	
S3-HM	12.19	0.14	10.12	0.04	6.93	0.14	11.2	0.09	62.1	0.77	

3.1.2. DOM Impact on Selected Soil Chemical Properties

The saturation of soils with different types of DOM caused a significant increase in the pH values and C contents in all cases (Table 2). The highest pH values were observed in S3 and the lowest in S1. The DOM derived from compost (DOM-CPT) had the weakest effect on the pH increase in all soils. The introduction of DOM-CM (cattle manure) caused the highest pH changes in the cultivated soils (S2 and S3), while DOM-BD (biogas digestate) caused the highest pH changes in soil S1. The DOM saturation increased the C contents. The greatest differences were obtained after DOM-CM introduction in S1 (S1-CM) and after the DOM-HM addition into S2 and S3 (S2-HM, S3-HM).

The saturation with DOM had a diverse effect on the total content of basic nutrient elements (Table 2). The P content increased significantly only after the addition of DOM-HM. The effect of the other types of DOM on P content either did not cause significant changes or caused a slight decrease in its content, which was observed under the effect of DOM-CPT in all soils. Similarly, slight changes were observed in the Al and Fe contents after DOM addition. Only in soils S1 and S3 was the Al content increased after the addition of DOM-CM and DOM-HM, in other cases the values were rather low compared to the control soil. In the case of Fe, a slight increase was observed in soil S1 under the influence of DOM-CM and DOM-HM, while in the other cases, the content of this component decreased.

The soils differed in the contents of the oxalate-extracted Fe and Al, which are considered as being main binding partners of phosphate [23] (Table 3). For Fe_{ox} and PSC, the order was S3 < S1 < S2. The two agricultural soils S2 and S3 had a larger DPS than the forest soil S1, reflecting previous P fertilizer applications to optimize crop production. The DOM addition with HM had the strongest positive effect on DPS, yielding values in the range of 62–66% in the arable soils. Such high DPS scores were reported for soils in an area of intensive husbandry in northwestern Germany [62], indicating a risk for P leaching [23,57]. Either the HM contained more P_{ox} than the other organic materials, or the organic matter of HM mobilized P to become oxalate extracted.

The relation between C and some components in the tested samples are noteworthy (Table 2). In general, the value of the C:P ratio increases (Table 2) in the studied soils after the DOM saturation according to the following order: O < CPT < BD < CM < HM. An exception is the effect produced after the treatment with DOM-HM of S1 (S1-HM), in which the C:P value is much lower than in O (S1); also, in soil S3, the mentioned tendency is less pronounced. The C:Fe and C:Al ratios follow a similar pattern to the C:P relationship, as they increase in all soils according to the order presented above. However, exceptions are observed for DOM-HM in S1—in which C:Fe and C:Al were lower than for DOM-CM. For the C:Al ratio in S3, some divergences are also seen. All values of the C:P, C:Fe, and C:Al ratios remained in similar relationships in the tested soils and followed the following order: S1 < S3 < S2.

3.2. Process of P Sorption

3.2.1. P Sorption in Unsaturated Soils (S1, S2, S3)

The results obtained for P sorption in S1–S3—soils with different P sorption properties—are the reference for the soils saturated with DOM. The standard deviations of the equilibrium solution concentrations from three replicates were in the range of 0.001 mg L⁻¹ to 1.844 mg L⁻¹. For the sorption isotherm, the calculations of the means were used and plotted in Figure 1 for control soils, and in Figure 2 for DOM-saturated soils. The standard deviations do not exceed the size of the symbols.



Figure 1. Phosphorus sorption in soils under study (S1, S2, S3). Explanation: Q—the amount of P adsorbed to soil at equilibrium concentration; C_{eq}—the P concentration in the equilibrated solution.



Figure 2. Phosphorus sorption after DOM saturation of soils S1 (**A**), S2 (**B**), and S3 (**C**). Symbols: S1, S2, S3—control soils; DOM types: CPT—compost, BD—biogas digestate, CM—cattle manure, HM—horse manure; symbols for soil 1: S1-CPT—soil 1 with compost DOM addition; S1-CM—soil 1 with cattle manure DOM addition; S1-HM—soil 1 with horse manure DOM addition; S1-BD—soil 1 with biogas digestate DOM. The same pattern was used for other soils. Q—the amount of P adsorbed to soil at equilibrium concentration; C_{eq}—the P concentration in the equilibrated solution.

The early stage of sorption (at the low P doses) in S1 (B horizon) indicates a very high affinity for P, which weakens considerably with the sorption of further P doses (Figure 1). The slope of the sorption isotherm decreases before reaching the highest P adsorption value (Q), of approximately 0.2 mg P g⁻¹ soil. The P sorption in S2 and S3 is similar between these arable soils at the lower equilibrium concentrations. Their much smaller slopes compared to S1 indicate a substantially smaller affinity for P, which is associated with the higher DPS value of both soils (Table 3); this observation is typical for overfertilized arable soils [62]. The relative shapes of the isotherms for S2 and S3 reflect the similar magnitudes of these soils' P sorption capacity (Table 3, Figure 1). The sorption capacity of S2 can be compared to S1, while Q is lower in S3 at 0.13 mg P g⁻¹. These results confirm a strong influence on Q of the type of land use of these soils, which seems to be the main factor determining the course of P sorption here. Arable soils (S2 and S3), regularly fertilized with P, have a much higher DPS value; as such, their affinity for P is much weaker in contrast to forest soil (S1), with a much lower DPS value (Table 3).

The P sorption isotherms obtained in the studied soils are typical examples of so-called "L" isotherms, in which the concentration ratio of the element remaining in solution and adsorbed on the solid decreases with the increasing concentration of the solute, providing a convex curve [63,64]. The P adsorption by soil is regarded as being a multi-stage kinetic process involving an initial fast adsorption stage (chemical) and then a slower adsorption stage (physico-chemical, physical), and possibly further stages [30,65]. At relatively low P concentrations, the adsorption process is completed rapidly and ion exchange and ligand exchange are probably the dominant mechanisms contributing to the high adsorption rate [30,66]. This quick chemical phase of the adsorption process slows down promptly at high concentrations of added P because the available adsorption sites quickly become saturated and P becomes physico-chemically and physically adsorbed from the liquid to the soil at a slower rate; the process at this point represents the slow adsorption phase [30,65,66]. Ultimately, the "L" curve may reach a strict asymptotic plateau when the solid has a limited sorption capacity, or the curve does not reach any plateau in cases where the solid does not clearly show a limited sorption capacity [63]. This is often observed in the case of sorption processes in natural soils, which are polydisperse and highly heterogeneous; in these situations, it may be difficult to clearly classify the isotherm as subtype "L".

3.2.2. DOM Effect on P Sorption in Soils

The saturation of soil samples (S1, S2, S3) with DOM derived from waste organic materials (CPT, BD, CM, HM) resulted in different effects on P sorption (Figure 2).

An increase in P adsorption (Q) at higher P doses was observed as DOM effects in all combinations except for soils saturated with DOM-HM. The highest Q reached about 0.25 mg P g⁻¹ and occurred as a result of DOM-CPT and/or DOM-BD saturation. The enhanced P sorption was also observed as an effect of the DOM-CM treatment in all studied soils. In S2, the DOM-BD and DOM-CM saturations slightly increased the sorption at the highest P dose, but the P sorption process occurred in a very similar way to the control soil in these both cases (S2). Comparable P sorption isotherms were also observed after saturation with DOM-CPT and DOM-CM in S3 (the Q was higher by about 0.05 mg P g⁻¹ than in S3). The soils saturated with DOM-HM behaved undoubtedly differently. In each soil, we observed P desorption at the initial stages of sorption (low P doses) and the significant weakening of P sorption (Figure 2). A smaller angle of the inclination towards the OX axis indicated a much lower affinity for P in the DOM-HM soil samples than in the control soils, and the decrease in Q was also observed (the Q was lower by about 0.05 mg P g⁻¹ than in the control soils), unlike with the other DOM treatments: CPT, BD, CM.

The initial stage of the sorption is determined mainly by conditions related to the properties of reactants important in chemical sorption, i.e., the number of sorption sites on mineral surfaces (including the bonds related to Al and Fe oxides, clay minerals and other soil colloids), PSC, or pH. These conditions explain the differences in the soils' affinity for P, as indicated by the slope at the beginning of the sorption curves (Figures 1 and 2). The affinity for P weakens in the following order: S1 > S2 > S3, and is similar for all cases even after saturation with DOM materials, excluding DOM-HM, which significantly diminished the affinity for P in every soil. At a later stage, when the sorption mechanism is no longer chemical binding, the type of adsorbent becomes important, and clearly reveals the differences specific to the type of DOM introduced. The different behaviour of DOM-HM at an earlier stage may be linked to the introduction of increased P amount with this DOM type, which could change the equilibrium of the reaction and lead to previously sorbed P becoming desorbed.

3.3. The P Adsorption Indices

The P adsorption isotherms were quantitatively described by the two most commonly fitted models—Langmuir and Freundlich equations. The fitted parameters and R^2 values for the models are summarised in Table 4. In most cases, the Freundlich and Langmuir equations are characterized by very high R^2 values. The only noteworthy exception ($R^2 = 0.67$) was the S3-HM treatment.

Soil Sample/		Freu	indlich Isot	herm			MBC				
Amendment	K _f	n _f	sum SE	Variance	R ²	KL	Q_{m}	sum SE	Variance	R ²	${ m mg}{ m g}^{-1}$
S1 (CONTROL)	0.1	0.27	0.001	0.0053	0.99	3.68	0.18	0.003	0.0053	0.95	0.662
S1-CPT S1-BD S1-CM S1-HM	0.12 0.1 0.09 0.03	0.34 0.43 0.40 0.67	0.003 0.002 0.002 0.001	0.0089 0.0085 0.0069 0.0035	0.97 0.98 0.97 0.98	2.78 1.05 1.25 0.12	0.24 0.26 0.23 0.26	0.003 0.003 0.002 0	0.0089 0.0085 0.0069 0.0035	0.97 0.96 0.97 0.99	0.667 0.273 0.288 0.031
S2 (CONTROL)	0.05	0.56	0.001	0.0053	0.98	0.23	0.27	0	0.0053	0.99	0.062
S2-CPT S2-BD S2-CM S2-HM	0.06 0.05 0.05 0.008	0.63 0.70 0.66 1.12	0.001 0.002 0.002 0.005	0.0083 0.0068 0.0064 0.004	0.98 0.98 0.97 0.88	0.22 0.13 0.16 0.01	0.37 0.39 0.34 1.34	0.002 0.001 0.001 0.005	0.0082 0.0068 0.0064 0.004	0.98 0.99 0.99 0.87	0.081 0.051 0.054 0.012
S3 (CONTROL)	0.03	0.49	0	0.002	0.99	0.27	0.15	0	0.002	0.99	0.041
S3-CPT S3-BD S3-CM S3-HM	$\begin{array}{c} 0.04 \\ 0.04 \\ 0.04 \\ 0.002 \end{array}$	0.55 0.73 0.64 1.51	0.001 0.001 0.001 0.004	0.0037 0.0064 0.004 0.003	0.98 0.98 0.98 0.85	0.24 0.09 0.15 0.07	0.22 0.45 0.26 0.2	0 0.002 0.001 0.009	0.0037 0.0064 0.004 0.003	0.99 0.97 0.98 0.67	0.053 0.041 0.039 0.010

Table 4. The P isotherm parameters.

3.3.1. Freundlich Equation Parameters

The capacity factor (K_f). The K_f value ranged in the control soils from 0.03 (S3) to 0.1 (S1). The almost ten times higher value of K_f in S1 than in the arable soils reflects a significantly higher P binding capacity of this soil, as we observed in the P sorption curves for the control soils (Figure 1). The S1 neither has more Al and Fe nor more Al_{ox} , Fe_{ox}, and PSC than the other two soils but its DPS is less than half of soils S2 and S3 (Tables 2 and 3). This indicates that K_f is determined by the available sorption sites in soil, as is also confirmed in the literature [67,68].

The addition of DOM revealed diverse effects on the K_f parameter (Figure 3A). In each soil, a significant increase (by 20–35%) of the K_f parameter was observed after DOM-CPT addition, while a substantial reduction in the K_f values (up to 90%) was caused by DOM-HM addition in all soils compared to the control soils. The saturation with DOM-CM and DOM-BD increased the K_f value only in S3 (of about 30%), which indicates the importance of soil properties influencing the value of K_f. No other effects on K_f were observed except for a decrease of about 10% in S1 after DOM-CM saturation (Figure 3A). The K_f parameter shows significant negative correlations with the contents of C, N, and with pH (Table 5). This confirms the important role of organic matter content and pH in building the P sorption capacity in soils and its binding strength and has been observed by other researchers [43,67–69]. This may explain such a strong decrease in the K_f value after DOM-HM application and the increase in this value in all soils as an effect of the saturation with DOM-CPT.



Figure 3. Changes of Freundlich isotherm parameters in S1, S2, S3 soils after DOM application: (A). Changes of K_f parameter; (B). Changes of n_f parameter. Changes (expressed in %) for the values of each parameter were related to the values of the control samples, which are displayed here as level 0.

Table 5. Correlation coefficients between the sorption isotherm parameters and soil properties. Marked (*) and bold correlation coefficients are significant with p < 0.05 (N = 15).

Variable	С	Ν	pH H ₂ O	Al	Fe	Р	C:Al	C:Fe	Al:P	Fe:P	C:P	N:P
K _f	-0.53 *	-0.60 *	-0.66 *	-0.46	-0.33	-0.66 *	-0.52 *	-0.50	0.82 *	0.43	-0.28	-0.30
n _f	0.78 *	0.80 *	0.69 *	0.46	0.41	0.78 *	0.78 *	0.81 *	-0.91 *	-0.59 *	0.61 *	0.58 *
KL	-0.74 *	-0.76 *	-0.67 *	-0.43	-0.36	-0.73 *	-0.73 *	-0.76 *	0.90 *	0.60 *	-0.58 *	-0.53 *
Qm	0.54 *	0.50	0.23	0.14	0.28	0.38	0.53 *	0.59 *	-0.44	-0.32	0.71 *	0.62 *
MBC	-0.57 *	-0.63 *	-0.71 *	-0.45	-0.31	-0.67 *	-0.55 *	-0.56 *	0.86 *	0.54 *	-0.34	-0.35

In addition to the basic soil properties, also, the total P content has a very strong effect on the course of P sorption. This is related to the number of available sorption sites for P in a given soil, as reflected in the DPS values. The statistically significant negative correlation coefficients between P content and the K_f parameter supports this explanation (Table 5), and is consistent with the soils saturated with DOM-HM having the largest P contents (Table 2). The lack of significant correlations between sorption isotherm variables and the total contents of Al and Fe are plausible. Essentially, the total contents include atoms in the internal crystal structures that are not directly involved in any interaction with phosphate. The linearity parameter (n_f). The value of the n_f exponent is a measure of the linearity of the function [70,71]. In control soils, the n_f value is lower than unity, ranging from 0.27 to 0.56 (Table 4). These values obtained are in line with the results of other authors [72]. After the saturation with each type of DOM, the exponent n_f was significantly increased. The effects on the n_f value follow the following order: HM >> BD > CM > CPT. The highest increase in the n_f value after the DOM-HM saturation (Figure 3B) suggests additional processes such as surface complexation and precipitation [73] that occurred preferably in the DOM-HM samples.

The n_f exponent is positively correlated with C, N, P, and pH (Table 5). This implies that saturation with DOM and the increase in the C content (including elements related to the presence of organic matter: N and P content) and the pH conditions have a dominant effect on the n_f values. This may be related to the quantity, but also to the quality of the organic matter introduced into the soil, since it is the structure of the organic molecules that will determine the characteristics of the functional groups active in P sorption. This may also explain the major changes in the n_f values observed under the influence of DOM-HM, whereby additional to a significant increase in the C content, we also observe the introduction of a large amount of phosphate ions. All these components (DOM, P, Al, and Fe), important for P binding in soils, could probably have interacted with each other differently, depending on the pH and other reaction conditions such as the contribution of these components. These issues are unfortunately beyond the scope of our study and require further analysis, with emphasis on the qualitative characterization of the organic matter introduced.

3.3.2. Langmuir Equation Parameters

The maximum adsorption capacity for P (Q_m). The Q_m reflects the number of P adsorption sites per unit weight of soil and is therefore commonly used to estimate the P adsorption capacity of soils [30,50,74]. The value of the Q_m ranged from 0.15 mg P g⁻¹ (S3) to 0.27 mg P g⁻¹ (S2) in control soils. This value is comparable for S1 and S3 and lower than Q_m in S2. After the saturation with DOM, the Q_m value increased in all cases (Figure 4A). All types of DOM amendments (CPT, BD, CM, HM) had a comparable effect on the Q_m in S1, giving about 30–40% higher values than in the control soil (Figure 4A). The DOM materials gave similar effects in S2, excluding the DOM-HM amendment, which greatly increased the calculated Q_m value (by as much as 400% compared to the control sample). This indicates that DOM-HM initiates binding processes that are different from the other soils. A stronger impact on the Q_m parameter was observed after the saturation with DOM-BD, DOM-CM, and DOM-CPT in S3. The magnitude of this effect followed the order: BD > CM > CPT. In contrast, the DOM-HM impact was negligible compared to the control soil (S3).





The comparable effect of the applied DOM types in S1 and S2 suggests that the origin of DOM is unlikely to have a significant effect on the value of the Q_m parameter. Also, the different properties of S1 and S2 did not affect the significant differences in the Q_m between the tested variants of the experiment. The increase in the Q_m values (Table 4) is attributed to an increase in organic matter (OM) content (Table 2), which is confirmed by a statistically significant (p < 0.05) correlation coefficient (r = 0.54) between the Q_m and total organic C content (Table 5), and remains in line with the results of others [30,50,74,75]. It is likely that DOM has occupied sorption sites potentially available for P-mainly in association with soil Al and Fe—but providing some new sorption sites for P on its own organic particles, hence the increase in observed Q_m. There are also results indicating that OM does not have a direct effect on Q_m and the influence of other soil properties is also important [74]. Previous research has shown the relationship between Q_m and OM to be dependent on soil pH [30], after Zhao et al. [76], who found that at pH > 6.0, increasing the OM content increased the amount of P adsorbed by soils but decreased the amount of P adsorbed by soils at pH < 6.0. In our study, we observed this relation as an increase in the Q_m value, with an increase in the C content at pH > 6.

The values of the K_f and Q_m parameters show disparate trends after saturation with HM. This material caused a marked decrease in K_f in relation to the control samples, while increasing the value of Q_m . We can probably explain these inconsistencies with reference to the assumptions that are implicit in the Langmuir and Freundlich isotherms [63,77]. In particular, the assumptions of the Langmuir isotherm may pose interpretation problems in soil materials (especially with the sorption of high concentrations of a given substance [26]). Essentially, the Langmuir isotherm assumes monolayer (ad)sorption, during which the reactants are characterized by a homogeneous, monodisperse nature, in which no additive interactions between the sorbents take place. This is an assumption that is impossible to meet in natural soils (or fractions thereof). This was confirmed by the weakest values of the fit of the theoretical curve to the obtained data (R2, Table 5) precisely for the Langmuir isotherm for the variants with HM. The Q_m parameter also shows a much weaker correlation with basic soil properties.

Numerous studies have reported the effect of soil organic matter on inhibiting P adsorption in soil by blocking adsorption sites on metal oxides, such as Al and Fe oxides, or by forming metal–SOM-P complexes with flexible structures [30,31,33,39,42,71]. Nevertheless, our results conducted on different types of DOM do not confirm this based on the Q_m value analysis. A Q_m value higher than in the control soils indicates that new sorption sites for P are likely to be generated after soil saturation with DOM. This is clearly demonstrated in S2 after DOM-HM saturation, in which a significantly higher number of sorption sites can be related to the higher amount of introduced DOM and the higher content of Al and Fe (especially amorphous Al and Fe—Al_{ox}, Fe_{ox}; Tables 2 and 3). Possibly, in this case, the sorption sites for P were not blocked, but a different sorption mechanism/s prevailed. It is not known clearly what the role of the high concentration of P introduced together with DOM-HM might have been. Possibly, in the presence of a high concentration of P, the behaviour of organic molecules was different than in other cases. Competition and the mutual blocking of these components may have occurred. Although, it is also possible that DOM interacted with the surface functional groups of Al and Fe, and/or generated additional sorption sites for P associated with the organic molecules' sorption active functional groups. It is possible that the high P concentration may also have favoured the formation of monodentate, rather than bidentate, bonds with P on active surfaces, which tended to bind P more weakly in the soil, thereby increasing the P pool that can more readily pass into solution. The probability of multilayer P sorption, the formation of monodentate bonds on sorptive surfaces, or increased P leaching under high P loading conditions has been repeatedly observed in the literature [78-80]. In the presence of high P content, bi- (or more) layer bonding and co-precipitation may occur as well. Another possibility is that DOM-HM could support the disintegration of Al-/Fe-oxide microaggregates, making more sorption sites available. All these mechanisms [73] may have resulted in such a large increase in the

maximum P sorption capacity (Q_m) calculated from our results and the correlations (Q_m :C) obtained (Table 5). However, questions remain on the effect of high P concentration on the activity of the introduced organic matter, and also on the ability of DOM to block/enhance P sorption as a function of its qualitative nature [32,81].

The P bonding energy constant (K_L). The K_L parameter reflects the P bonding energy and is one of the most important indicators describing the soil affinity for P. A higher K_L value indicates stronger P sorption, and spontaneous sorption may occur more readily as the P supply becomes less intense [65]. The K_L parameter values calculated for control measurements can be ordered as follows: S2 < S3 << S1. The comparable K_L values lower than 0.3 were calculated for arable soils (S2, S3), while the forest soil had a K_L value at least ten times higher (Table 4). These differences reflect various soil properties; primarily, the mineralogy that governs P-binding sites and the land use/management history, e.g., the former P fertilization at the arable soils S2 and S3. This view is supported by the apparent much higher DPS values in the arable soils compared to forest soil S1.

The K_L parameter decreased considerably as a general effect of the saturation with DOM materials (Figure 4B). This indicates that the introduced organic materials weakened the P binding energy in all three soils. The new sorption sites likely generated under the DOM's influence are energetically weaker in holding P than the Al and Fe bonds [35,73], thus leading to the marked reduction in the K_L parameter, which was greatest in S1 compared to other soils. A comparable effect of soil organic matter on the K_L values was observed by Yang et al. [30] in studies on phaeozem soils that received an addition of humic substances. The largest negative effect on K_L was observed after the saturation with DOM-HM, followed by DOM-BD, DOM-CM, and the weakest effect was observed after the introduction of DOM-CPT (Figure 4B). The strongest impact of DOM-HM is probably related to the large amount of P incorporated into the soil (Table 2), so P fixation is much weaker in such a material and probably leads to the formation of single bonds with P that are definitely more easily desorbable from soil.

The Maximum Buffering Capacity (MBC). The MBC is a compound parameter that combines the effects of the Q_m and K_L values [30,65]. The higher MBC means that more P can be sorbed by soil. We observed an increase in the MBC value (Figure 4C) solely as an influence of DOM-CPT in S2 and S3 (S1-CPT had no effect). The other DOM types predominantly caused a decrease in the MBC value (Figure 4C), which was the steepest after DOM-HM saturation in all soils. No influence was observed in S3-BD and S3-CM.

The different response to the DOM-CPT introduction may be related to the specific nature of the raw organic waste composted and, above all, to the transformation of OM during the mineralization and humification processes [82,83]. This probably allowed for the production of a material with a stronger retention of P in the soil, e.g., DOM with a more aromatic character than the other inputs. It is well-known that aromatic moieties exert the largest binding energies for phosphate among various organic functionalities [73]. Although the effect of CPT on Q_m is similar for other types of DOM (Figure 4A), it stands out as having the weakest effect on the reduction in the K_L parameter (Figure 4B) compared to other DOM materials. However, further studies on the nature and composition of CPT organic matter are required to confirm these mechanisms.

3.4. Relationships Affecting P Sorption in DOM-Treated Soils

The PCA displays the relationship among the analyzed data for soil samples saturated and unsaturated with DOM materials, and estimates which parameters or parameter combinations best characterize the variability (Figure 5). Each axis, in a two-dimensional representation, explains a certain percentage of the total variability that exists between samples. The principal components 1 (PC1) and 2 (PC2) explained, together, 82.01% (68.74 + 13.27) of the total variability among the selected properties of the DOM-amended soils and control soils and gives the two-dimensional representation of the components to describe the sample characteristics. The total content of P, C, N, pH, and the C:P, C:Al, and Al:P ratios, and the values of K_f, K_L, and MBC are the main contributors in PC1, while the Fe:P ratio seems to be the main contributor in PC2. The parameters with vectors situated closer to each other indicate parameters that may have a high positive correlation. These include the sorption parameters K_L , K_f , MBC, and the Al:P ratio. In contrast, they are negatively correlated with some basic soil properties, such as pH, C, N, P, and the C:P, C:Al, C:Fe ratios. These results are confirmed by statistically significant, strong coefficient correlations (Table 5). The vectors that form an angle close to 90°, such as the Fe:P and sorption parameters (K_L , K_f , MBC) may not present a significant correlation. We observed statistically significant correlation coefficients for the Fe:P ratio with the n_f exponent, KL, and MBC (Table 5).



Figure 5. Projection of variables onto the factor plane (1×2) .

The position of the soil samples displayed in Figure 6 reveals that all the variants of S2 and S3 (arable soil), are located in two quadrants of the graph, indicating the similar character of these samples. They are comparable in their basic chemical properties, and strongly differ from soil S1. Soil S1 and its DOM-treated variants, having different characteristics, are situated in the same area in which the Al:P, K_L, K_f, and MBC parameters appear. This is undoubtedly related to the different land use as forest of soil S1, but also to the different genetic material of the collected soil. Within these two separated groups of soils, a certain type of differentiation due to the type of DOM applied is also interesting: the DOM-HM group stands out the most clearly in the top of the chart, followed by the DOM-BD group. The subsequent variants already show a much weaker specificity, but DOM-CPT is usually slightly higher than DOM-CM and the control samples, which (apart from S1) appear in the lowest position in Figure 6.

The PCA and strong significant correlation coefficients obtained between the total organic C content and all sorption parameters confirm the important role of soil organic matter in P sorption process in soils. It is worth noting that the Q_m parameter, that was the weakest correlated sorption parameter, showed significant positive relations with the C content and C-related ratios. The proportion of the Al and Fe content to the total organic carbon (C:Al, C:Fe) and total phosphorus content (Al:P, Fe:P) are of great importance in explaining P sorption in the studied soils, as evidenced by the high correlation coefficients revealed between these indices and the isotherm parameters (Table 5). This supports our hypothesis that an increasing amount of organic matter in the form of added DOM interacts with active sorption sites on Al and Fe (hydr)oxides. Specifically, by blocking the sites in

this manner, DOM affects P sorption in the soil. The strong positive correlations between n_f (and negative for K_f , K_L , and MBC) with the C:Al and C:Fe (and C:P) ratios, together with the significant negative correlations between n_f (and positive for K_f , K_L , MBC) with the Al:P and Fe:P ratios provide evidence for this thesis. Evidently, both the increasing proportion of the total C content relative to Al and Fe (as well as to P) and the decreasing Al and Fe content relative to the total P content are highly important in the P sorption process.



Figure 6. Projection of cases onto the factor plane (1×2) .

4. Conclusions

Our study indicates that dissolved organic matter (DOM) introduced with agricultural wastes did not always reduce P sorption, but certainly had an effect on impairing P fixation and, thus, may result in potentially greater P mobility in the soil, including P availability.

Our hypothesis, that P sorption sites, derived in sandy soils mainly from Fe and Al oxides, would be blocked by introduced organic matter in the form of DOM, was only partially confirmed. As a consequence of the saturation with DOM, we observed a substantial increase in the heterogeneity of the energy of P sorbing sites (n_f) and a weakening of the P bonding energy (K_L), while at the same time an increase in the sorption capacity (Q_m). The DOM (excluding the impact of DOM-CPT) affected the maximum buffering capacity (MBC) in the studied soils and decreased the potential soil activity in P retention.

Based on the P sorption parameters, DPS and P sorption isotherms, and multivariate statistical data evaluations, we recommend horse manure to at least partially overcome the P fixation in arable soils, because it had the most positive effect on the P concentrations in the equilibrium solutions. The molecular basis of the positive HM-DOM effects requires further studies. The effect of DOM saturation that we observed is likely an average result of the activity of the functional groups of DOM, and the predominant nature of these groups will determine the soil P binding capacity and its mechanism. For these reasons, qualitative studies of the different types of DOM and their ability to compete with P for the same sorption sites are an important direction for further research on the application of organic wastes in terms of the sustainable management of P availability in soil.

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