Research Article

Risk of Leaching in Soils Amended by Compost and Digestate from Municipal Solid Waste

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New European directives have proposed the direct application of compost and digestate produced from municipal solid wastes as organic matter sources in agricultural soils. Therefore information about phosphorus leaching from these residues when they are applied to the soil is increasingly important. Leaching experiments were conducted to determine the P mobility in compost and digestate mixtures, supplying equivalent amounts to 100 kg P ha⁻¹ to three different types of soils. The tests were performed in accordance with CEN/TS 14405:2004 analyzing the maximum dissolved reactive P and the kinetic rate in the leachate. P biowaste fractionation indicated that digestate has a higher level of available P than compost has. In contrast, P losses in leaching experiments with soil-compost mixtures were higher than in soil-digestate mixtures. For both wastes, there was no correlation between dissolved reactive P lost and the water soluble P. The interaction between soil and biowaste, the long experimentation time, and the volume of leachate obtained caused the waste's wettability to become an influential parameter in P leaching behavior. The overall conclusion is that kinetic data analysis provides valuable information concerning the sorption mechanism that can be used for predicting the large-scale behavior of soil systems.

1. Introduction

In the EU, between 118 and 138 million tons of biowaste are produced each year, approximately 88 million tons of which are municipal waste. This latter value is projected to increase by 10% by 2020. European standards encourage the recovery of the organic fraction of municipal solid waste (MSW) by composting, anaerobic digestion, or incineration [1], reducing the amount of waste sent to landfills in accordance with the Landfill Waste Directive [2]. In 2006, the EU was host to 124 plants performing anaerobic digestion of biowaste, with an overall treatment capacity of 3.9 million tons y⁻¹ and a total annual compost production of 4.8 million tons.

The use of compost and digestate as soil fertilizer provides agronomic advantages, such as the improvement of several soil properties: structure, water infiltration, water-holding capacity, microorganism content (both amount and diversity), and nutrient content [3]. In particular, better phosphorus recycling may reduce the need for mineral fertilizers.

The most recent communication from the Commission to the Council and the European Parliament on future biowaste management steps in the European Union [1] proposed the recovery of both the digestate and compost from MSW anaerobic digestion as sources of stable degraded organic matter (approximately 45% of EU soils are characterized by low levels of humus [4], especially in southern Europe).

This new communication [1] promotes the digestate and the compost from MSW to be directly applied to the soil as valuable sources of organic matter and nutrients. Therefore, it is necessary to study the behavior and interaction of these biowastes with soils to evaluate their optimal use and potential environmental problems.
In many catchments around the world, agriculture is now the major contributor of P to surface waters [5]. The transport of P from agricultural soils to surface waters has been linked to eutrophication in fresh water and estuaries [6–8].

According to a recent document from the EU, "Sustainable phosphorus use" [9], 90% of the total phosphorus entering the food system (mineral fertilizer and organic manure) is lost before reaching the product, mostly via dissipation into the water system. Global losses from the soil to fresh water are estimated at 18.7 to 31.4 million tons per year [10]; in the EU-27, losses to leaching and runoff are estimated at 0.16 million tons per year [11].

Reuse of organic waste has been based on crop nitrogen (N) requirements and usually supplies P in excess of crop needs [12, 13]. Long-term phosphorus application to soils as fertilizer or manure can increase the potential for P loss to ground and surface waters.

These "excessive" soil P concentrations can be measured by environmental soil P-tests, such as water-soluble P (WSP) and FeO-P, which have been linked to P loss from agricultural land, or by agronomic soil tests, such as Mehlich-1 and Mehlich-3, which estimate the P available for crop growth [14–16]. The P source WSP test is a reliable mean for predicting the dissolved reactive P (DRP) concentrations in runoff from surface-applied manures and biosolids [17]. This phosphorus fraction should consist largely of the inorganic orthophosphate (PO$_4^{3-}$). The concentration of this fraction constitutes an index of the amount of phosphorus immediately available for algal growth. The WSP/TP ratio (the fraction of the total P that is water soluble) allows a more direct comparison of the environmentally relevant P in biosolids and manures with differing chemical and physical properties [18]. For leaching experiments, Sharpley and Moyer [19] found that the amount of P leached from six livestock manures and manure composts was significantly correlated with the water-extractable P in the materials (in the absence of soil). Other authors have suggested that the WSP of the P source materials is a good preliminary predictor for approximating the P leaching loss [20]; however, this may not account for continuous P release from added organic materials during continuous water infiltration.

Leaching and surface runoff experiments allow to evaluate the potential losses of soil P produced by the application of P sources to soils. Although P loss in runoff is considered to be the major contamination route, leaching also causes significant losses. Some authors have suggested that P leaching to groundwater is unimportant because the leaching is negligible [21]; however, other authors report that the downward movement of P from organic wastes is potentially significant in areas with shallow groundwater and coarse soil with low P-absorption capacity [22, 23].

Laboratory leaching tests are common tools for assessing the long-term impact of contaminated materials on the soil-groundwater pathway by determining the source term as an expression of the release potential of water-soluble contaminants during the use or disposal of waste materials. These tests provide a flow-through pattern similar to that found in field conditions and permit the basic characterization of waste materials [24]. The release of soluble components upon contact with water is regarded as a main mechanism of release, and this result in a potential risk to the environment during the reuse or disposal of such materials.

Although the soil characteristics are not generally considered to be important in surface runoff tests [25], soil type is relevant in leaching tests due to its effects on the behavior of P, especially in applications by incorporation. The soil adsorption index of P (PSI) was analyzed for its relationship to the P source losses.

The evaluation of the P speciation in biowaste is very important when determining the suitability of biowastes for land application or the optimum application rate.

The physical properties of biowastes are also important. The wettability of biowastes is an important property in the leaching process in soil. Several authors have observed the influence of soil wettability in aggregate stability and the decomposition of soil organic matter [26, 27]. Although strong water repellency has been shown to have negative effects on hydrological process (e.g., soil erosion), a slight increase in water repellency may reduce the breakdown of aggregates and consequently reduce surface sealing, overland flow, and erosion. Hydrophobicity, as a measure of water repellency, caused by organic substances, favors the formation and protection of stable aggregates [28] which, in turn, stabilize the encapsulated organic substances against microbial degradation and mineralization [29, 30].

Wettability may have an important effect on the stabilization of SOM due to a reduction of liquid adsorption rates, accessibility for microorganisms, and restricted accessibility of water and nutrients. Hydrophobic SOM is more stable against microbial decomposition.

In soluble species in the waste that could be lost by leaching, water must pass through the soil profile, wet the waste, and dissolve these species. Soil water repellency has been extensively studied and is mainly caused by organic compounds of various origins and structures [31]. The wettability of soil particles increases with the charge density and fraction of polar groups on the surface [32, 33]. Sorption of organic matter with nonpolar functional groups promotes nonwettatable surfaces [34], and long-chain amphiphilic organic compounds produced by a range of biota can induce hydrophobicity in soil [35]. When wetted, these compounds are usually hydrophilic, but drying can cause bonding of hydrophilic (polar) ends of amphiphilic molecules to each other or to particle surfaces, resulting in the exposure of hydrophobic (nonpolar) functional groups to the pore space [36]. This effect can be observed on biowastes after the application of various treatments.

The goals of this work were (a) to quantify the phosphorus leaching losses from organic waste generated in MSW treatment plants and (b) to evaluate the differences in the behaviors of digestate and compost on the potential mobility and P availability in three soil types. To that end, a first-order kinetic model was used to estimate digestate and compost wettability and leaching behavior.
2. Materials and Methods

2.1. P Source Samples. The biowastes applied as P sources were a compost and a digestate from an anaerobic MSW digestion plant, located in southern Madrid. Ten samples (5 kg each) were collected for the residues on different days and combined to produce a single homogenous sample representing each residue.

The digestate was obtained after the anaerobic digestion (21 d, 39°C, and constant agitation) of the organic fraction of MSW followed by dehydration with the addition of flocculants and centrifugation.

Compost was obtained by the composting in tunnels (14 d, 55°C, periodic watering and forced aeration) of a mixture of digestate of MSW and defibered plant matter, with a subsequent static stabilization period and sieving.

The digestate had a pasty texture, forming large clumps, whereas the compost was a powder with a much smaller particle size (<5 mm).

2.2. Chemical Analyses. The study was conducted with the surface horizon (0–20 cm) of three soils, from Madrid (40°32' N, 3°17' W), Guadalajara (40°28' N, 4°0' W), and Ciudad Real (39°0' N, 3°56' W) (Spain). All soil samples were collected in plots from agricultural research stations. These locations were chosen because their histories of P application are well known. The soils selected had not suffered applications of phosphorus in the last five years.

Soil organic matter (SOM) was determined by dry combustion at 540°C for 4 h. Soil pH was determined in a 1:5 (v/v) water extract. Soil Olsen-P was extracted with 0.5 M NaHCO3, pH 8.5, for 30 min and analyzed using the Murphy and Riley [37] spectrophotometric method. Calcium (Ca) and iron (Fe) concentration in soil were determined by inductively coupled plasma atomic optical emission spectroscopy (ICP-OES) following USEPA Method 3050A acid digestion. Total P in filtrates of water, NaHCO3, and NaOH extracts were determined by digesting aliquots of filtrates in an autoclave at 103.5 kPa with acidified (NH4)2S2O8 [44]. P in all the extracts was analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-OES) following USEPA Method 3050A digestion. Labile P includes the sum of IP and OP from water, resin, and NaHCO3 fractions, whereas refractory or unavailable P includes the remaining fractions. The procedure was performed in triplicate on each biowaste sample.

A Sigma force tensiometer and the Washburn technique for the wetting of porous solids were used to estimate the biowaste wettability, with the mass of adsorbed liquid (water, g) measured by weight difference every second for 20 minutes. Finally, biowaste particle size was determined by a granulometric analysis.

2.3. Column Leaching Experiment. The leaching tests were performed according to the European standard CEN 14405:2004 “Characterization of waste—Leaching behavior test—Up-flow percolation test” [45].

Thirty glass columns (6 cm in diameter × 21 cm in height) were prepared by adding a layer of 21 cm of each of the three types of topsoils (0–15 cm) from Spanish research centers: soil A (Calcic Haploxeralf), soil B (Petrocalcic Palexeralf), and soil C (Typic Haploxeralf) (Soil Survey Staff, 2010). Columns were prepared in triplicate for each experiment. The soils were air-dried, crushed, sieved (<2 mm), and homogenized.

Each soil (600 g) was amended with different P sources at an application rate of 100 kg P ha−1 (25.5 g of compost and 11.6 g of digestate, resp.), which is considered too high for agricultural needs. Due to the widespread practice of fertilizer application based on N content, in general, these high rates of P are very common in the field. Appropriate controls of soils without waste application were included, and all treatments were performed in triplicate. The columns were carefully packed to avoid the formation of preferential water paths. Fiberglass was placed in the bottom and top of each column to prevent soil loss.
The columns were saturated with distilled water from the bottom upwards, and the saturation was balanced at room temperature for 72 h. The distilled water was then allowed to flow out of the bottoms of the columns. The leaching was transported continuously for 20 d with a constant flow of 22 mL h⁻¹ induced by a peristaltic pump from the bottom of each column. The leachate was collected from the top of the columns. Sixteen stages of leachate per column were collected, with liquid-solid (L/S) ranges of 0.1 to 10 L kg⁻¹.

Leaching fractions were analyzed for dissolved reactive phosphorus (DRP), determined from the filtered samples (0.45 μm) without digestion. The P content was determined by ICP-OES.

2.4. Kinetic Model. A variety of kinetic equations including zero-, first-, and second-order, fractionation-power, and parabolic-diffusion and Elovich equations have commonly been employed over the years to describe the kinetics of soil chemical phenomena [46–48].

In the case of the phosphorus leaching experiment data, P leaching (P) was adjusted to a first-order kinetic model:

\[
P = P_{\text{max}} \cdot \left(1 - e^{kr}\right).
\]

Here, \(P_{\text{max}}\) (mg kg⁻¹) is the maximum phosphorus leaching expected, \(k_l\) (h⁻¹) is the rate of phosphorus leaching, and \(t\) (h) is the time.

The behavior of biowaste wettability (W) as a function of time is expressed as follows:

\[
W = M_w \cdot \left(1 - e^{-brt}\right).
\]

Here, \(M_w\) (g) is the maximum wettability expected, \(br\) (s⁻¹) is the adsorption rate, and \(t\) is the time (s).

2.5. Statistical Analysis. Analysis of variance (ANOVA) using the \(F\)-test at a significance level of 0.05 was performed to establish the possible significant differences between the mean values of leached phosphorus among the different treatments and soil types.

3. Results

Table 1 provides details of some characteristics of the soils used in this study. The main differences between the soils are the higher clay content of soil A (28%) relative to the other soils, which are sandy (<70%). Soils A and B are slightly basic (pH 7.5–7.9), whereas soil C is acidic (pH 5.9). All three soils have low levels of organic matter. The Fe and Ca concentrations are higher for soil B (5.0% Ca, 1.4% Fe). Values of P(III) indicate the retention capacity of P for each soil. Soil B has the highest P adsorption capacity (112 mg kg⁻¹), and soils A and C have similar P adsorption capacities (30 and 27 mg kg⁻¹, resp.).

A summary of the chemical properties of the digestate and compost used in the study is provided in Table 2.

Total solid content is higher in compost (81.9%) than in digestate (28.2%). The digestate has a lower OM content (45.3%) than the compost (62.0%). With respect to acidity, the compost is neutral (pH = 7.5), whereas the digestate is slightly basic (pH = 8.5). The TP concentration of the digestate (7.49 g kg⁻¹) is about twice that of the compost (3.09 g kg⁻¹). The TP concentration, obtained after the calcination and acid extraction of biowaste, is a useful overall indicator of pollution but provides no information about the solubility of P species, which depends on their chemical forms.

Table 2 shows that IP was higher than OP in both digestate (94%) and compost (91%). The Olsen-P and WSP are much higher for the digestate (55% and 49% of the TP) than for compost (20% and 22% of the TP). Significant differences were observed in the calcium content, which was 5 times lower for digestate than for compost (0.8% and 4.5% for digestate and compost, resp.). The hydrosoluble organic matter content was also lower for digestate than for compost. The analyzed biowastes exhibited low concentrations of heavy metals and trace elements (data not shown).

Percentages of WSP, membrane-P, NaHCO₃-P, NaOH-P, and HCl-P with respect to total P for digestate and compost are given in Figure 1. The main P fraction for both biowastes was HCl-P, at 49.6% in digestate and 58.0% in compost, followed by NaHCO₃-P (16.4%) in digestate and NaOH-P in compost (19.5%). WSP and membrane-P were the smallest fractions for both biowastes.

The sum of the percentages of TP composed of WSP and membrane-P, designated as "loosely bound-P", is higher for digestate than for compost (18.5% and 6.7% of TP, resp.). The sum of the percentages of loosely bound-P and NaHCO₃-P, designated as "labile P", is also higher for digestate than for compost (34.8% and 22.4% of total P, resp.). The large amount of this fraction in digestate indicates high vulnerability for both P leaching and availability to plants.

Digestate has a much higher content of P that is easily lost. In contrast, the compost has high P content in the NaOH-P and HCl-P fractions, indicating that the extracted P is more recalcitrant and therefore more difficult to dispose of.

To characterize the wetting of each residue, Figure 2 shows the amount of water absorbed (g) versus time for the two biowastes studied, digestate and compost. The two residues exhibited totally different behaviors. Digestate initially absorbed water rapidly (up to 100 s); later, the amount of water absorbed was fairly constant, indicating a lower wettability. The compost absorbed a large amount of water relative to its weight, with water absorption increasing steadily throughout the trial (1200 s). Wettability results for digestate and compost were well described by a first-order kinetic model (Figure 2). The water adsorption rate (br) for digestate (17.9 · 10⁻³ s⁻¹) was higher than for compost (1.02 · 10⁻¹ s⁻¹) but the maximum wettability (Mw) was much higher for compost (1.97 ± 0.09 g of adsorbed water per g of waste) than for digestate (0.82 ± 0.05 g of adsorbed water per g of waste).

Figures 3 and 4 represent the DRP (mg P kg⁻¹ of soil) lost by leaching as a function of time (h) for digestate and compost in each soil. The total DRP concentration (mg P kg⁻¹ of soil) accumulated in 10 L of leachate varies for each type of soil, from 43 ± 1.8 mg P kg⁻¹ to 49 ± 1.5 mg P kg⁻¹ for the mixtures.
Table 1: Chemical characteristics of the soils used in this study.

| Parameters                  | Soil A             | Soil B             | Soil C             |
|-----------------------------|--------------------|--------------------|--------------------|
| USDA classification         | Calcic Haploxerupts| Petrocalcic Palexeralfs | Typic Haploxeralfs |
| Texture                     | Clay loam          | Sandy clay loam    | Sandy loam         |
| Sand, %                     | 55.0               | 70.4               | 71.0               |
| Silt, %                     | 17.0               | 8.0                | 11.0               |
| Clay, %                     | 28.0               | 21.6               | 18.0               |
| pH, w extract 1:5           | 7.5                | 7.9                | 5.9                |
| OM, %                       | 1.41               | 2.22               | 0.06               |
| Ca, %                       | 0.7                | 5.0                | 0.6                |
| Fe, %                       | 1.1                | 1.4                | 0.6                |
| Olsen-P, mg kg⁻¹            | 18.8               | 17.9               | 10.1               |
| PSL, mg kg⁻¹                | 29.9               | 112.2              | 26.5               |

Table 2: Biowaste chemical characterization: compost and digestate.

| Parameters      | Digestate | Compost |
|-----------------|-----------|---------|
| Particle size, mm | 10–25     | <5      |
| Total solids, %  | 28.2      | 81.9    |
| OM, %           | 45.3      | 62.0    |
| Hydrosoluble OM, % | 1.1      | 1.9    |
| EC, dS m⁻¹       | 6.8       | 4.7     |
| PH              | 8.5       | 7.5     |
| Kjeldahl-N, %   | 3.2       | 2.8     |
| TP, g kg⁻¹      | 7.49      | 3.09    |
| WSP 1: 250, g kg⁻¹ | 3.64    | 0.69   |
| IP, g kg⁻¹      | 7.04      | 2.80    |
| OP, g kg⁻¹      | 0.39      | 0.25    |
| Olsen-P, g kg⁻¹ | 4.11      | 0.61    |
| Ca, %           | 0.8       | 4.5     |
| Fe, %           | 1.4       | 0.5     |

*Dry weight basis.

of soil and digestate and from 42 ± 2.1 mg P kg⁻¹ to 62 ± 2.3 mg P kg⁻¹ for the mixtures of soil and compost. Losses of P vary from 33% to 37% and from 32% to 48% for digestate and compost, respectively.

Losses of DPR in columns amended with compost were more variable and lower for soil B than for soils A and C (Figure 3), which is consistent with the PSI values for each soil type. Soil B has the largest capacity to absorb P (112 mg kg⁻¹) and thus a lower capacity for retention of P, while soils A and C have lower PSI (30 mg kg⁻¹ and 27 mg kg⁻¹, resp.). Results in columns amended with digestate were similar for the three soils (Figure 4). Columns of soil with compost, with a lower WSP content (22% of the TP), lost large amounts of the P applied (from 32% to 47%). In contrast, the DRP losses from the columns treated with the digestate, which have a very high WSP content (49% of the TP), were lower than expected and varied less with the soil type (from 33% to 37%).

The dynamics of phosphorus leaching in the column experiments fitted a first-order kinetic model. Values of kl and Pₘₐₓ are shown in Table 3. According to these results, leaching rates (kl) were higher for the soils with digestate (2.3 × 10⁻³ to 6.7 × 10⁻³ h⁻¹) than for soils with compost (1.7 × 10⁻³ to 1.8 × 10⁻³ h⁻¹).

In all cases, the calculated values for maximum phosphorus leached (Pₘₐₓ) were higher for soils with compost (72 to 108 mg kg⁻¹). In soils amended with compost, Pₘₐₓ values were significantly higher in soils A and C than in soil B.
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Figure 3: DRP losses by leaching in soil-compost mixtures. Error bars indicate the standard error of the mean \((n = 3)\).

Figure 4: DRP losses by leaching in soil-digestate mixtures. Error bars indicate the standard error of the mean \((n = 3)\).

Table 3: First-order kinetic constant \((k_l)\) and \(P\) maximum leached estimated \((P_{\text{max}})\) for each biowaste and amended soil type.

| BIOWASTE | Soil | \(k_l \times 10^{-3}\) (h\(^{-1}\)) | \(P_{\text{max}}\) (mg kg\(^{-1}\)) |
|----------|------|-----------------------------------|----------------------------------|
| Compost  | A    | 1.7                               | 108.0\(^{\text{a}}\)             |
|          | B    | 1.8                               | 72.3\(^{\text{b}}\)              |
|          | C    | 1.7                               | 99.5\(^{\text{a}}\)              |
| Digestate| A    | 4.4                               | 54.6\(^{\text{a}}\)              |
|          | B    | 6.2                               | 47.8\(^{\text{a}}\)              |
|          | C    | 2.3                               | 61.9\(^{\text{a}}\)              |

\(^{\text{a}}\) Same capital letters within a soil type indicate that there were no significant differences between the biowaste type at \(\alpha = 0.05\). Same lowercase letters within the same biowaste type indicate that there were no significant differences between soil types.

5. Conclusions

As recent works have demonstrated, leaching losses of \(P\) by application of organic wastes cannot be neglected. Measuring...
biowaste for P indices (WPS, TP, or Olsen-P) that determine soluble or labile P are not useful for assessing P loss by leaching, though this is a practice currently applied in runoff experiments. Instead, it is necessary to evaluate other characteristics of the biowastes. In this experiment, wastewater wettability has been useful for explaining P losses.

The soil PSI is important for assessing potential P losses by leaching; however, the influence of soils is different for the two types of biowaste. The maximum phosphorus leached in soils amended with compost is significantly dependent on soils and consistent with the PSI values. In contrast, soils amended with digestate present lower total P losses, and the effect of soil is not significant. This phenomenon may be due to the lower soil-waste interaction as a consequence of digestate wettability.

Finally, our overall conclusion is that the P leaching rate depends mainly on the biowaste type and is less dependent on the soil type. However, the maximum amount of P leached depends on both the type of waste and the soil characteristics.

**Abbreviations**

MSW: Municipal solid waste  
WSP: Water-soluble phosphorus  
DRP: Dissolved reactive phosphorus  
TP: Total phosphorus  
FeO-P: Phosphorus extracted by iron oxide  
PSI: Soil adsorption index of phosphorus  
TS: Total solid  
OM: Organic matter  
W: Wettability  
Mw: Maximum wettability  
Br: Biowaste rate  
P\text{max}: Maximum phosphorus leaching  
k\text{l}: Rate of phosphorus leaching.

**Conflict of Interests**

The authors declare that there is no conflict of interests regarding the publication of this paper.

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