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Environmental risks of increasing phosphorus addition in relation to soil sorption capacity

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Abstract

Phosphorus content of soil profiles, interstitial water, and shallow groundwater was determined in a semiurban region of Buenos Aires city in order to examine phosphorus leaching. Soils from different places and position in the slope were characterized and analysed for their P sorption– desorption capacities in order to understand transport processes and to assess potential environmental risks. Adsorption increases with depth and decreases with increasing additions of phosphorous. Interstitial water at 0.6 m depth showed greater P concentrations than shallow groundwater (9 to 30 m depth) in accordance with the importance of clays for P adsorption. © 2006 Elsevier B.V. All rights reserved.

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1. Introduction

The transport of P to surface waters sensitive to eutrophication has been a worldwide environmental concern for more than 30 years (Sims et al., 1998; Sharpley et al., 2003). Accumulation of bioavailable P in surface waters leads to algae proliferation, an increase in oxygen demand, and deterioration of water quality (Sharpley et al., 1993; Lemunyon and Gilbert, 1993; Indiati and Sharpley, 1995; Sharpley et al., 1996). However, because P leaching to groundwater has normally been considered to be inconsequential in most soils the effect of accumulation in groundwater has been less studied even though, there are combinations of soil properties, agricultural management practices and climatic conditions that may result in significant P accumulation in subsoil and leaching to groundwater (Sims et al., 1998). Groundwater may be influent to surface water thus enhancing eutrophication processes. It is known that the P concentration in groundwater and the amount of P retained by the soil are related to the soil's ability to sorb P (Weaver, 1993) and hence in order to assess the local envi-

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ronmental effects on groundwater of P addition e.g. through agrochemicals, the relationship between P transport/soil accumulation and P content in groundwater should be established for specific soil types and regions.

Generally, within an ecosystem, P can be leached to the shallow groundwater, transported in soluble forms, or through erosion, in combined form with the colloidal particles (particulate P), following which it can be deposited in sedimentation areas to finish its cycle in the water system. The period over which a soil can assimilate applied nutrients without breakthrough depends on the soil's sorption capacity for the nutrients, the rate at which nutrients (in this case P) are applied and the depth of soil above the groundwater table (Weaver, 1993). Because the sorption capacity of soil is limited, some national environmental policies (Breeuwsma et al., 1995) have incorporated the concept of a critical degree of P saturation e.g. german legislation states the saturation percentage that should not be exceeded to prevent adverse effects on groundwater quality, with the specific goal that the phosphate concentration in groundwater should not exceed 0.10 mg l^{-1} of ortho-P at the level of the mean high water table.

In order to fully assess the risk of contamination of drainage water by P leaching measurement of P accumulation must be accompanied by a measure of desorbability. Provin et al. (1995)

found that cumulative water-soluble P concentration in soils and subsoil are well correlated with Bray P1 (Davedere et al., 2003; Dalv and Casev. 2005: Vadas et al., 2005), suggesting that routine bioavailable P soil testing may be a useful tool in predicting the potential for P loss in leaching and drainage waters. Djodjic et al. (2004) have shown that incorporating other routinely measured soil properties (e.g. texture) into regression equations with P concentration measured from soil testing can improve the ability to predict cumulative watersoluble P from multiple consecutive extractions. The development of P soil testing methods that can identify soils likely to exacerbate the environmental impact of nutrient effects on water quality is an area in need of continued research especially in relation to the development of standardized methods that can relate the measured concentration of P to the desorption and downward movement of P and the potential for P loss in subsurface runoff.

In respect of this it seems likely that at least two hydrologic pathways are operative in the subsurface transport of P from soil to water (Sims et al., 1998). The first is the gradual, downward movement of P in percolating waters that interact with the bulk of the soil profile and eventually with tiles or subsurface waters flowing laterally and discharging into ditches or streams. The second is bypass flow, the rapid movement of dissolved and particulate P via macropores that extend from the soil surface to tile drains or subsoil horizons where accelerated lateral flow to surface waters occurs. The slow, continual leaching of P by the first process should be of greater concern in soils with extremely high P concentrations from long-term fertilization and manuring (e.g. intensive animal agriculture). Bypass flow of P, however, could occur in any soil where physical or biological macropores exist and would be more or less independent of soil P concentration although it would clearly be of greater environmental significance in high P soils. The two processes are obviously not mutually exclusive and could simultaneously contribute to P loss by subsurface runoff in many situations (Sims et al., 1998).

A question that may arise particularly in areas dominated by intensive agricultural activities is whether the P sorption capacity of some soil profiles is saturated to the extent that continuous, environmentally significant leaching of P occur. Since in Argentina intensification of agricultural activities has occurred in the last two decades, it is of utmost interest to perform studies on P groundwater content because groundwater is an important water source for shallow lakes (pampasic ponds), which are hypertrophic in the region of greatest agricultural importance in the country (Miretzky et al., 2000). In the present paper, P content in soil profiles, interstitial water, and shallow groundwater was determined in a semiurban region presenting mixed activities in the surroundings of Buenos Aires city (Escobar, province of Buenos Aires) in order to examine P leaching processes. Phosphorus sorption-desorption properties of different soil profiles were studied in order to understand transport processes and to assess likely environmental risks.

Eutrophication restricts water use for fisheries, recreation, industry, and drinking, due to increased growth of undesirable

algae and aquatic weeds, followed by oxygen shortages as the biomass decomposes. Also, many drinking water supplies throughout the world undergo periodic massive surface blooms of cyanobacteria. These blooms contribute to a wide range of water-related problems, including summer fish kills, unpalatability of drinking water, and formation of trihalomethane, a known carcinogen, during water chlorination. Recent outbreaks of the dinoflagellate *Pfiesteria piscicida* in the eastern U.S. have also been linked to excess nutrients in affected waters.

Neurological damage in people exposed to the highly toxic volatile chemical produced by this dinoflagellate has dramatically increased public awareness of eutrophication and the need for solutions. In most cases, phosphorus (P) accelerates the eutrophication of fresh waters. Consequently, controlling algal blooms and eutrophication mainly requires reducing P inputs to surface waters (Pierzynski and Sharpley, 2000).

Argentina has more than 400 lakes with surface area >5 km², but for some of them not even major ion data are available. The Chaco–Pampa Plain lakes are usually lightly impacted by agricultural operations. All lakes in the Chaco–Pampa Plain are very shallow and range from eutrophic to hypertrophic or salt lakes. Most of the lakes situated in the central-western and northwestern arid regions are reservoirs or salt lakes, and range from mesotrophic to eutrophic (Quiros and Drago, 1999).

2. Materials and methods

The soil profiles, interstitial and groundwater under study are located between 34° 18' S and 58° 51' W to 34° 24' S and 58° 44' W, in the geomorphologic unit Rolling Pampas (Pampa ondulada) in Buenos Aires province of Argentina. The province has a wet climate and a mean temperature of 16 °C. The mean annual precipitation is 900 mm. Although precipitation is higher in the summer, the precipitation balance is negative in this season.

Soil samples were collected in soil trenches during January 2004 (summer) from undisturbed sites, located in different places in Escobar county (Garín, Maschwitz, Belen de Escobar, Loma Verde and Matheu). They were air dried and sieved to pass 2 mm sieve and analysed for extractable phosphorus (Bray–Kurtz 1 method), pH (1:2.5 soil water relationship), organic carbon (OC) (Walkley–Black method), electrical conductivity (CE) and clay content (Bouyoucous method) as previously described (Sparks, 1996; USDA, 1996).

In order to assess the response capacity of the analysed soils to increasing fertilization, sorption–desorption experiences with increasing additions of phosphorus to dry soil samples were performed. Adsorption was determined for every horizon in each of the soil samples, since both the water transport mechanism through the soil and the subsoil properties are more important than P content in the topsoil for P leaching (Djodjic et al., 2004). A solution of CaCl₂ 0.01 M (25 ml) containing increasing quantities of P: 0 (P0), 12.5 (P12.5), 25 (P25), 50 (P50) and 250 (P250) μ g P (as K₂HPO₄), was added to dry soil (2.5 g). These quantities are equivalent to an addition of: 0 (P0), 5 (P5), 10 (P10), 20 P20) and 100 (P100) μ g P g⁻¹ of soil. After addition of chloroform (1% in volume) to eliminate microbial

activity, the suspension was shaken for 2 h, no difference was found when shaking time was larger than 2 h in our own experience working with Mollisol soils from Rolling Pampas (Giuffré de López Camelo and Heredia, 1988). The quantity of sorbed P is expressed as a percentage, and arises from the difference between the added P and the remaining P in the soil solution. P in soil solution for an addition equivalent to $5 \ \mu g \ g^{-1}$ soil is referred to P5sn and similarly for other concentrations. P was determined using the colorimetric molybdenum blue method (Murphy and Riley, 1962), and is expressed in mg l⁻¹. The Freundlich isotherms were used to describe the process of P sorption (Ho et al., 2001).

This is an empiric model that can be applied to nonideal sorption on heterogeneous surfaces as well as multilayer sorption and is expressed by the following equation:

$$q_{\rm e} = a_{\rm F} C_{\rm e}^{b_{\rm F}} \tag{1}$$

Where q_e is the amount of phosphorus sorbed at equilibrium (µg g⁻¹), a_F is the Freundlich constant (l g⁻¹), C_e is the solution ion concentration at equilibrium (µg ml⁻¹) and b_F the Freundlich exponent.

This isotherm assumes an exponential decaying sorption site energy distribution. Eq. (1) can be linearized

 $\ln q_{\rm e} = b_{\rm F} \ln C_{\rm e} + \ln a_{\rm F}$

A plot of $\ln q_{\rm e}$ versus $C_{\rm e}$ gives a straight line of slope $b_{\rm F}$ and intercept $\ln a_{\rm F}$ (Ho et al., 2001).

Correlation between the different sorption indexes, the edaphic variables (clays, pH, Cox, CEC) and extractable P was explored using InfoStat 1.1 (2002).

Interstitial water was collected from nursery soil (U-soil 10), which was cultivated and amended for the 12 months before the experiment was performed. Soil amendment was organic and consisted of bovine manure compost in a $30\% \text{ v v}^{-1}$ proportion in the A horizon. Water samples were taken monthly during 6 months (from June to November) by means of a pressure-vacuum soil water sampler at 0.6 m depth. Groundwater samples were collected from wells in the Pampeano aquifer (34 samples, 9 to 30 m depth). Interstitial and groundwater analysis for P was carried out by the Murphy and Riley method (1962).

The saturation index was determined in interstitial and groundwater by PhreeqcI (USGS, 2005).

3. Results and discussion

3.1. P adsorption-desorption

Soils were classified according to Soil Taxonomy (Soil Survey Staff, 2003), as shown in Table 1, corresponding to Mollisols and Entisols orders. The soils are in two slope positions: the upper (U) position, between 8 to 28 amsl (U-soils 1, 2, 4, 5, 6, 7 and 10), and lower position (L), between 4 to 0.6 amsl (L-soils 3, 8 and 9).

Soil P content (Bray P1) is shown in Table 2. An irregular P distribution with depth was observed. Anomalously high extractable P values are observed for soil 2 (sandy soil) and 10 (soil with organic amendments due to a previous nursery).

P isotherms for the A horizon and the highest clay content horizon are shown for soils 2, 4 and 9 (Fig. 1A,B) as an example. U-soils 2 (sand) and 4 (clay loam) are characteristic of this slope position, being the first one of sandy class, while L-soil 9 (silty clay loam) is representative of lower soils. Soils 1, 5, 6, 7 and 10 showed a similar behavior to soil 4, while L-soils 3 and 8 behaved like soil 9. In general, it was observed that adsorption in the A horizon was lower than for the clayey horizon, e.g. adsorption increases with soil depth as may be expected. On the other hand, adsorption tends to decrease with increasing P addition. Among the A horizons, considering all applied doses, the soil with higher adsorption was U-soil 5 while no adsorption was observed for U-soil 10, which is the soil from the nursery. Among the B horizons, the higher adsorption P values were observed in U-soil 1 and the lowest in L-soil 8 in accord with clay content.

Since it was observed that adsorption diminished with increasing additions of P, profiles are shown on addition of 100 μ g g⁻¹ soil (the maximum dose applied). The adsorption varied between 0 to 75% in horizon A, mostly between 25 and 70%, as shown in Table 3. It can be observed that in general adsorption is greater in the deeper horizons. Only L-soil 3 shows a significant decrease in adsorption with depth. Soils 2 and 10 showed abnormal profiles, as previously observed

Table	1				
Main	physicochemical	parameters	of the	studied	soils

Identification	Location	Coordinates	Taxomic classification	pН	Extractable P (mg kg ⁻¹)	Organic carbon (g kg ⁻¹)	Clay (%)	CEC (cmolc kg ⁻¹)
1 (U)	Garín	34° 24′ 53.9″ S 58°44′ 36.7″ W	Typic Argiudoll	6.4	5.9	13.5	27.5	16.7
2 (U)	Maschwitz	34° 22′ 42.3″ S 58°44′ 40.6″ W	Entic Hapludoll	6.4	73.8	8.1	15.0	7.7
3 (L)	Maschwitz	34° 21′ 46.9″ S 58°44′ 05″ W	Aquic Hapludoll	6.5	13.7	17.5	56.3	15.6
4 (U)	Bélen de Escobar	34° 20′ 11.9″ S 58°48′ 06.7″ W	Typic Argiudoll	6.7	12.7	12.4	27.5	17.3
5 (U)	Bélen de Escobar	34° 20′ 34.4″ S 58°46′ 53.9″ W	Typic Argiudoll	6.2	4.8	13.2	38.8	17.1
6 (U)	Bélen de Escobar	34° 20′ 05.2″ S 58°47′ 15.6″ W	Udarent	7.9	2.7	7.8	42.0	18.0
7 (U)	Loma Verde	34° 18′ 53.4″ S 58°51′ 07.9″ W	Typic Argiudoll	6.9	6.5	23.6	22.5	13.1
8 (L)	Loma Verde	34° 18′ 38″ S 58°51′ 18.8″ W	Typic Natraquoll	7.4	11.4	26.9	25.0	15.9
9 (L)	Matheu	34° 23′ 36.8″ S 58°49′ 06.1″ W	Mollic Fluvaquent	8.8	9.2	30.0	50.0	20.0
10 (U)	Matheu	34° 22′ 48.3″ S 58°48′ 48.3″ W	Typic Argiudoll	6.7	38.6	19.5	32.5	17.2

Table 2 Extractable phosphorus concentration at different soil depth

Soil number										
Depth	1	2	3	4	5	6	7	8	9	10
(cm)	Extractable P ($\mu g g^{-1}$)									
10	5.9	73.8	13.7	12.6	14.8	2.7	6.5	11.4	9.2	38.6
30	10.3	73.8	12.6	9.2	14.8	3.8	6.5	13.7	8.6	23.5
50	10.9	48.7	18.5	9.2	16.1	7.0	7.0	3.8	4.8	23.0
70	7.0	19.1	8.1	7.0	23.5	7.0	8.1	3.3	3.8	12.6
90	5.9	43.9	9.2	7.0	13.2	8.1	8.1	7.0	4.8	
110	9.2			8.1	12.0		6.5	13.2		
140				10.3						

Table 3 Adsorbed phosphorus on addition of 100 $\mu g \; g^{-1}$ of P at different soil depth

Soil number										
Depth	1	2	3	4	5	6	7	8	9	10
(cm)	Adsorbed P ($\mu g g^{-1}$)									
10	45.2	30.8	75.5	36.4	69.1	65.5	48.9	23.8	50.5	0.0
30	62.6	18.8	76.0	45.6	69.1	67.9	62.5	48.5	52.7	50.5
50	90.6	36.6	69.8	55.6	81.2	29.0	62.5	48.5	67.7	67.7
70	76.7	7.9	54.3	83.6	81.6	29.0	56.1	53.3	67.3	80.3
90	72.9	80.4	45.1	83.6	80.3	65.9	56.1	62.6	48.9	
110	72.8			79.6	74.4		39.5	67.9		
140				55.7						

for the Bray 1 phosphorus profile (Table 2). In the former case, this is probably due to irregular genetic clay content, while in the latter it is likely due to the organic amendment applied for a long time. No further adsorption was observed in the upper horizons.

The relationship between P adsorption and edaphic properties was examined. A significant correlation was observed between adsorbed P and clay content for all the added P concentrations: r=0.36 (p<0.05) for P5, r=0.495 for P10 (p<0.001), for P20 (r=0.62) and P100 r=0.77 (p<0.001). A positive correlation between the soil CEC and P adsorption was also observed. The organic carbon content and P0sn showed a highly significant positive correlation suggesting that the organic matter would favour the desorption of P to the soil solution. Moreover, P adsorption at low concentrations of added P (P5 and P 10) negatively correlates with organic carbon suggesting that organic matter may compete with P for adsorption sites (r=-0.53 and -0.396 respectively, p<0.01) in accord with previous findings for other soils in Argentina (Heredia, 1997).

The extractable P (Bray P1) was negatively correlated with the sorbed P (p < 0.01) for all the concentrations of added P.

When correlation analysis was performed separately for soils in the upper (U) and lower (L) parts of the field different trends for extractable P and organic carbon were observed, showing the importance of the position of the soil in the slope. In U-soils,



Fig. 1. Adsorption P isotherm. A: A horizons. B: clayey horizons.

Bray P1 is correlated in a negative way with P adsorption (p < 0.01) suggesting that this parameter is a good index of P leaching. Nevertheless, in L soils no significant correlation was observed. The U soils showed no correlation between adsorbed and desorbed P with organic carbon, while in the L-soils this correlation was very strong.

Freundlich isotherm fitting is shown in Table 4 for U-soils. The Freundlich constant (a_F) show strong correlation with clay

Table 4				
Freundlich	isotherm	fitting	for	U-soils

Soil	Horizons	$a_{\rm F} \ (1 \ {\rm g}^{-1})$	b_{F}	r^2	р
1	А	18.93	0.548	0.984	< 0.001
	A_2	24.15	0.593	0.990	< 0.001
	B_t	103.99	0.690	0.954	< 0.01
	BC	55.34	0.372	0.999	< 0.001
	IIA _{b1}	43.05	0.482	0.792	< 0.1
	IIC	52.48	0.364	0.974	< 0.01
2	A_1	5.75	0.560	0.760	< 0.10
	AC	6.92	0.520	0.970	< 0.01
	IIA _{b1}	10.62	0.718	0.974	< 0.01
	IIC_1	2.73	0.468	0.997	< 0.001
	IIIC	57.94	0.639	0.923	< 0.01
4	A ₁₁	14.45	0.553	0.959	< 0.01
	A ₁₂	22.80	0.441	0.993	< 0.001
	AB	30.48	0.383	0.999	< 0.001
	B_{t1}	62.79	0.600	0.822	< 0.05
	B _{t2}	58.08	0.466	0.997	< 0.001
	BC	29.72	0.432	0.999	< 0.001
5	A_1	38.11	0.541	0.987	< 0.001
	AB	59.29	0.644	0.972	< 0.01
	B_{t1}	53.58	0.674	0.993	< 0.001
	B _{t2}	57.15	0.539	0.987	< 0.001
	BC	30.69	0.602	0.819	< 0.05
6	1	30.20	0.613	0.988	< 0.001
	2	36.98	0.452	0.984	< 0.001
	3	6.76	0.333	0.888	< 0.05
	4	32.73	0.480	0.971	< 0.01
7	A_1	22.65	0.500	0.986	< 0.001
	B_{t1}	34.28	0.479	0.991	< 0.001
	B _{tk1}	26.92	0.493	0.997	< 0.001
	B _{tk2}	18.78	0.443	0.980	< 0.001
10	А	nd	nd	nd	
	AB	5.82	1.534	0.806	< 0.10
	B_{t1}	34.83	0.646	0.975	< 0.001
	B _{t2}	59.70	0.551	0.988	< 0.001

nd: not determined.

 $(r=0.614 \ p<0.01)$, organic carbon $(r=-0.339 \ p<0.05)$ and CEC $(r=0.461 \ p<0.01)$.

In general $a_{\rm F}$ is bigger in clays horizons. The $b_{\rm F}$ values range between 0.3 and 1.5 and show no correlation with soil parameters.

3.2. P transport to groundwater

It has been previously reported that the transport of soluble P may be estimated from the desorbible P in CaCl₂ 0.01 M or distilled water (Dorich et al., 1985; Pierzynski et al., 1994). Therefore, the soil solution P profiles for all the studied soils were determined. It can be observed that P concentrations varied between 0 to 0.33 mg l⁻¹ in the A horizons, decreasing in subsurface horizons by an order of magnitude (from 0 to 0.04 mg l⁻¹). Since these values may be representative of the quantity of bioavailable P, they could be related with P content in interstitial and groundwater.

Contents of P in interstitial water during the 6 month experiment varied between 0.12 to 1.18 mg I^{-1} with an average value of 0.60 mg I^{-1} , indicating pulses of P solubilization and desorption. Although these values are higher than those determined in the soil solution, they are within the range previously reported for other soils (Mengel and Kirkvy, 1987). The highest value was found only once, being the other values more in accordance with soil solution concentrations. Taking into account P contents determined in the soil solution for U-soil 10 during the adsorption experiment, interstitial P concentrations are within the range determined for the soil solution. Without P addition, the P content in the soil solution varied between 0 and 0.33 mg I^{-1} , while with added P, those values varied between 0.32 to 6.31 mg I^{-1} for U-soil 10.

Since in Argentina maximum allowed P groundwater contents have not been established, a value of 0.05 mg l^{-1} , as suggested by USEPA (1992) for incoming water to surface water bodies, was used to evaluate the effect of P groundwater concentrations in the study area.

The P concentration in shallow groundwater was between 0.014 to 0.571 mg l^{-1} , with an average value of 0.13 and median value of 0.06 mg l^{-1} , with 53% of the samples over 0.05 mg l^{-1} . Some of the studied soils may desorb P in concentrations similar to those found in groundwater. Retention ability diminishes with increasing P addition, being a risk for shallow groundwater. The P content in interstitial water is higher than groundwater because its content diminishes due to clay retention.

In sandy soils or soils with high extractable P and low P adsorption such as soil 2 and 10, the highest P groundwater contents were observed. On the other hand soils with high clay content, high P adsorption and low extractable P such as soils 1, 4 or 5, lower P groundwater concentrations were determined.

Saturation Index (SI) were estimated for hydroxyapatite both in interstitial and groundwater in order to analyse the possibility of phosphorus being dissolved from the soils or precipitated from water. Interstitial water is saturated in hydroxyapatite (SI=2.37) while groundwater is not (SI=-3.18), confirming the importance of clays in P adsorption.

With increasing addition of P through fertilizers, detergents, domiciliary residues, sewage sludge, blind wells or industrial

products with P, the studied soils show important rates of retention of 90–95% in the U-soils (e.g. U-soil 5), but less than 80% in L-soils (e.g. L-soil 3) or even lower when the contents of sand are high and in consequence low in clays (e.g. soil 2) where the adsorption can be from 31 to 42% of the added P. This retention capacity diminishes when added P increases, thus progressing through the unsaturated zone to groundwater, with high risk of contamination when it is influent to surface water.

In the pampean region in Argentina there is a strong relation between groundwater and surface water. The water bodies of the humid pampean region are eutrophic and the different activities in the area contribute to P concentration in groundwater. Therefore the study of the adsorption–desorption processes, and the vertical and subsurface movements of P are important tools for environmental risk assessment. P input through groundwater has been up to now not considered in eutrophication studies and should be taken into account.

4. Conclusions

The importance of knowing the physical and chemical processes that attenuate the transport of phosphorus and their passage to aquifers and from the aquifer to other water bodies is of great importance since the accumulation of bioavailable P in surface water results in water quality deterioration.

Phosphorus adsorption increases with soil depth, in accordance with the positive correlation found between P adsorption and clay content.

The Freundlich constant (a_F) shows a high correlation with clay content, CEC and organic carbon. In general a_F is higher in clayey horizons.

The phosphorus interstitial water content has its origin in the processes of P solubilization and adsorption–desorption. The high values found are in accord with the previous organic ammendments applied to the nursery and with the fact that a negative correlation was found between organic carbon content and P adsorption. Hydroxyapatite saturation index is indicative of the saturation of interstitial water in phosphorus. The fact that groundwater samples are not saturated in this mineral phase, confirms the importance of clay content in phosphorus adsorption.

Extractable phosphorus as determined by Bray 1 is a good index for dissolved P, especially for U-soils.

The fact that P adsorption decreases with increasing P addition reinforces the importance of the study of P sorption– desorption processes within the soil and unsaturated zone, since fertilizers and animal slurry applications to the land from farms and sewage sources lead to an important input of inorganic and organic forms of phosphorus. These compounds progress through the unsaturated zone to the water table with high risk of contamination when groundwater is influent to surface water.

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