

# Soil phosphorus dynamics along a loess–limestone transect in Mihla, Thuringia (Germany)<sup>#</sup>

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## Abstract

The distribution of phosphorus (P) along a loess–limestone soil transect were investigated to delineate the spatial variation of the nutrient vertically in the soil profiles and laterally in the landscape. We hypothesized that spatial P patterns result from translocation caused by P mobilization, although P fixation would be expected along the slope. To depict this, three P fractions clearly differing in solubility were determined. Soil samples were treated with 0.1 M hydrochloric acid (HCl), with 12.1 M HCl, and with *aqua regia* (AR). In the profiles the spatial P distribution slightly corresponds to the occurrence of different bedrocks and substrata. Thus, a native “P loading” might not primarily explain the spatial P patterns. Especially the strong enrichment of the toeslope with easily soluble P indicates P translocation and prior mobilization. The enrichment is detectable throughout the profiles. Thus, superficial translocation (e.g., erosion) cannot sufficiently explain that pattern. Instead, underground processes must be the cause for this. They cause relatively high vertical and lateral variation in the spatial P distribution, e.g., within soil horizons and substratum layers. Hence, mixed sampling of soil sections might not produce data accurate enough for some kinds of P research and for P management. Also, the lateral P distribution should be detected more precisely prior to fertilization of agricultural land.

**Key words:** nutrient management / P fixation / P mobilization / P translocation / spatial P distribution / soil P fractions

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## 1 Introduction

Without mineral P fertilizers food could not be produced in amounts and quality large enough for modern demands (Vaccari and Strigul, 2011; Filippelli, 2017). The rapid decrease of these fertilizers' raw materials arouses fears of an upcoming global shortage of P-fertilizers within the current century (Cordell et al., 2009; Cordell and White, 2011). This has spawned interest in soil P research, because soils are the place where P enters the food chain (Cordell, 2010; Kruse et al., 2015).

While soil P research during the last decade strongly dealt with alternative P sources for agriculture (Eichler-Löbermann et al., 2007; Qayyum et al., 2015; Liu and Lal, 2017), spatial P patterns have rarely been investigated. Partly, this might be due to the common assumption in soil science that P quickly and strongly bonds to soil solids, thus, becoming spatially nearly immobile (Barber, 1995; Riddle and Bergström, 2013; Sharpley et al., 2014). Hence, P translocation is often considered a rather exceptional process, occurring mainly on the soil surface during erosion (Delgado and Scalenghe, 2008; Andersson et al., 2013; Bergström et al., 2015) and storm

runoff (Tunney et al., 2000; Kleinman et al., 2015; Bol et al., 2016). Underground P translocation is traditionally considered negligible because the soil solution and groundwater regularly contain small P concentrations (Heathwaite, 1997; Stewart et al., 2017). However, during the last decade several authors have documented the effects of underground P translocation by preferential flow (Ulén et al., 2013; Sharpley et al., 2014; Bergström et al., 2015; Bol et al., 2016). Thus, P migration in subsoils is increasingly considered possible.

Neglected before, we currently know little about underground P translocation and the emerging spatial P distribution in soil profiles and landscapes. This is reflected by the common methodological approaches of soil P research. Between January 1990 and October 2016, 62% of 177 P-related papers in *Plant Nutrition and Soil Science* (JPNSS) represent studies sampling exclusively topsoils. Further 15% do not even specify the spatial background of their samples. Subsoils are considered in 23%, complete soil profiles in 5% of the papers. Only 7% investigated the lateral distribution of P (e.g., in transects/catenae, slope sequences).

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Thus, in soil P research, soils have often been poorly represented as spatial continua. Knowing about the spatial P distribution seems necessary though, especially in times of growing scarcity. Spatial P patterns can help generate a better understanding of all the chemical and spatio-temporal processes (e.g., migration) affecting a soil's P status (= P dynamics). This is a prerequisite for a targeted and efficient P management in agriculture and may help to prevent ecological issues (e.g., eutrophication of lakes and coastal waters). Hence, we investigate if soil P is translocated without exceptional events like erosion. We hypothesize that the spatial P distribution in soils is (1) primarily caused by translocation processes (2) that mainly happen underground, and (3) are stimulated by prior P mobilization.

## 2 Material and methods

### 2.1 Soil sampling

A sequence of 19 soil profiles was cored with an auger (length: 1 m; core diameter: 2 cm). The transect was placed on a slope West of Mihla (51°04' N, 10°18' O), Wartburg district, Thuringia, Germany (Fig. 1). Here, mean annual rainfall of 598 mm and an average annual temperature of 8.3°C were recorded (*ClimateData*, 2016).

We established P profiles and reference profiles. P profiles were sampled completely in 5-cm-steps to depict the spatial P distribution in detail. Reference profiles were sampled according to soil horizons (classified after *AG Boden*, 2005) to illustrate local soil conditions. Samples were air-dried, ground, passed through a 2-mm sieve and afterwards stored airtight in plastic bags until analysis.

### 2.2 Chemical analyses

We determined texture (DIN ISO 11277:2002-08), humus content (DIN 19684–3:2000-08), pH (with KCl) and electrical conductivity (DIN ISO 10390:1997-05), and content of Ca carbonate (DIN 10693:1997-05) for the reference samples. P profile samples were treated according to *Weihrauch et al.* (2016b) to determine their P contents. We distinguish three

arbitrary P fractions differing in solubility, but not assigned to discrete P forms (e.g., Ca-bound P). Easily soluble  $P_{dHCl}$  was extracted with 0.1 M HCl. This procedure has been used in geoarchaeological research before to determine plant-available P, as well as P forms which are already stronger bound and, thus, not directly bioavailable (*Weihrauch et al.*, 2016a). Furthermore, slightly soluble  $P_{cHCl}$  was extracted with 12.1 M HCl and recalcitrant  $P_{AR}$  with *aqua regia*.  $P_{dHCl}$  and  $P_{cHCl}$  were measured on a spectrophotometer,  $P_{AR}$  by ICP-MS (*Weihrauch et al.*, 2016b).

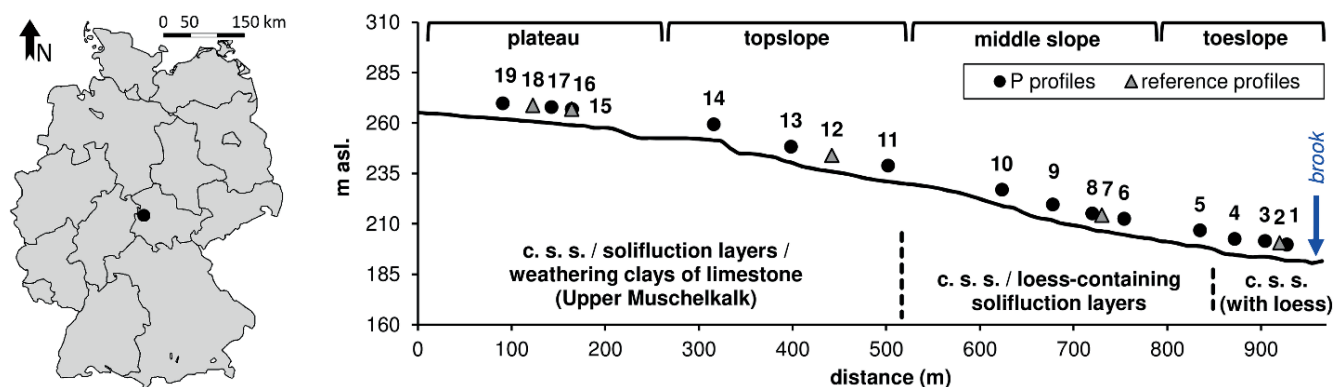
### 2.3 Statistical analyses

Samples were measured three times and averaged. For simplification, data are given as “mg P kg<sup>-1</sup>”, meaning mg P kg<sup>-1</sup> of dried, ground and sieved soil. Basic statistical operations were performed in Microsoft Excel 2013 (Microsoft, Redmond, WA, USA). Means of grouped data, their standard errors and significance of differences between means (Tukey-HSD-test) were determined in SPSS 24 (IBM Corporation, Armonk, NY, USA). Significances were tested for the  $p < 0.05$  level.

## 3 Results

### 3.1 Study site and local soils

The examined slope is comprised of agricultural land (profiles 1–14), grassland (profiles 15, 16), and deciduous forest (profiles 17–19). The agricultural sites have been under conventional cultivation since at least the late 1970s under the direction of the agricultural cooperative (LPG) in the former German Democratic Republic. No documentation of agricultural measures exists for that time. For the last 20 years the field has also been under conventional farming, including crop rotation, mineral and organic fertilization, and tillage with or without the plough (according to previous crop) to reduce erosion. During the investigation the slope was cropped with winter rape (2012), winter wheat (2013, 2015), and bean (2014). According to soil tests, the field had an average P content of 69 mg kg<sup>-1</sup> in 2008 (*LUM*, 2017), after extraction with calcium-acetate-lactate (CAL; *Schüller*, 1969), which is



**Figure 1:** Geographical location of the study area (left) and organization of the soil transect (right). The investigated slope is 2–9° inclined to the Northeast. Soil substrata for the slope sections are given from top to bottom (c.s.s. = Holocene colluvial slope sediments). (Sources: EuroGeographics 2017 for the administrative borders; Thüringer Landesamt für Vermessung und Geoinformation for height asl data.)

one of the official procedures to determine plant-available P in Germany. Thus, no annual P fertilization is necessary here and the field only received small amounts of additional P by liquid biogas manure in 2012 and 2015 (12.5–15.0 kg P ha<sup>-1</sup>; LUM, 2017).

To facilitate discussion, the slope is divided into four segments (Fig. 1). The plateau and topslope comprise transitional stages of Cambisols (Braunerden) with Mollic Leptosols (Rendzinen) and Chromic Cambisols (Terrae fuscae). On the middle slope, Cambisols and mixtures of Cambisols and Luvisols (Parabraunerden) were found. On the toeslope, Cambi-

sols were identified, with stagnic features throughout the profiles 1–3 (Cambic Planosols or Braunerde-Haftpseudogleye). General characteristics of the soils are shown in Tab. 1.

## 3.2 Spatial P patterns

### 3.2.1 Plateau

The plateau sites are low in P<sub>dHCl</sub>, but relatively high in P<sub>CHCl</sub> and P<sub>AR</sub> (Tab. 2). Higher values of P<sub>dHCl</sub> tend to occur in

**Table 1:** General characteristics of the reference profiles, sampled according to soil horizons (AG Boden, 2005).

	Depth (cm)	pH <sub>KCl</sub>	EC (μS/cm)	Humus <sup>a</sup>	Carbonate <sup>a</sup>	Sand <sup>a</sup>	Silt <sup>a</sup>	Clay <sup>a</sup>
<b>Profile 2</b>								
M°Ap-Sg	0–30	7.6	168.5	3.82	2.2	15.2	66.8	18.0
M°Bv-Sg	30–60	7.5	155.0	2.64	1.9	16.5	65.5	18.0
II M°Bv-Sg	60–90	7.6	163.5	2.33	2.1	19.1	62.3	18.6
III M°Bv-Sg	90–100	7.6	173.0	2.48	2.1	21.7	59.2	19.1
<b>Profile 7</b>								
M°Ap	0–26	6.6	111.0	3.85	0.0	13.7	67.2	19.1
M°Ah-Bv	26–40	6.6	64.5	2.94	0.0	13.3	68.6	18.1
II Bv	40–84	6.3	48.8	2.29	0.0	11.5	69.0	19.5
III Bv	84–100	6.1	46.0	1.78	0.0	10.1	70.3	19.6
<b>Profile 12</b>								
M°Ap	0–28	6.4	104.5	4.56	0.0	8.1	54.9	37.0
II Bv-Tv	28–60	6.2	59.5	3.22	0.0	6.5	49.9	43.6
III Bv(-Tv)	60–85	6.3	50.0	2.50	0.0	8.6	51.5	39.9
IV Bv(-Tv)	85–100	6.1	51.0	2.13	0.0	6.6	51.1	42.3
<b>Profile 16</b>								
M-rAp°Ah	0–7	6.0	71.0	9.14	0.0	9.8	66.2	24.0
M-rAp°Bv	7–26	5.3	39.0	4.12	0.0	15.9	63.3	20.8
II Bv	26–44	5.4	33.0	3.12	0.0	7.1	60.2	32.7
III fBv-Tv	44–70	7.2	249.0	3.66	0.0	12.5	42.4	45.1
IV cCv	70–100	7.5	202.0	2.76	18.2	23.1	38.1	38.8
<b>Profile 18</b>								
M°Ah	0–21	4.5	68.0	9.48	0.0	6.2	73.7	20.1
II Ah-Bv	21–37	4.9	37.0	3.03	0.0	6.2	70.9	22.9
III cCv-Bv	37–77	6.8	72.0	2.66	0.0	8.1	66.6	25.3
IV cCv	77–93	7.0	150.0	3.91	0.9	4.8	44.7	50.5
V cCv	93–98	7.5	264.0	n.d. <sup>b</sup>	11.0	7.1	31.3	61.6

<sup>a</sup>Data in mass-%.

<sup>b</sup>n.d. = not determined.

**Table 2:** Statistical overview of the P data according to slope positions.<sup>a</sup>

		Plateau	Topslope	Middle slope	Toeslope
<b>min</b>	P <sub>dHCl</sub>	0.0	0.0	6.0	37.0
	P <sub>cHCl</sub>	187.0	372.0	297.0	482.0
	P <sub>AR</sub>	850.0	620.0	591.0	904.0
<b>max</b>	P <sub>dHCl</sub>	1070.0	360.0	380.0	687.0
	P <sub>cHCl</sub>	2144.0	2849.0	720.0	1196.0
	P <sub>AR</sub>	8732.0	8493.0	3326.0	7654.0
<b>focus</b>	P <sub>dHCl</sub>	<25 (64%)	0–200 (82%)	6–200 (79%)	201–400 (76%)
	P <sub>cHCl</sub>	201–500 (64%)	401–800 (65%)	301–600 (89%)	501–700 (59%)
	P <sub>AR</sub>	1501–2500 (50%)	751–1500 (50%)	751–1500 (84%)	1251–3250 (74%)

<sup>a</sup>Data in mg kg<sup>-1</sup> of soil. Percentages in parentheses refer to number of samples with the according P contents.

> 40 cm of depth (Fig. 2), but no significant depth-specific trend was observed. P<sub>cHCl</sub> instead is significantly more abundant in > 60 cm of depth. A comparable, but not significant trend shows for P<sub>AR</sub>. We also found no significant trends based on substratum or soil horizons (data not shown).

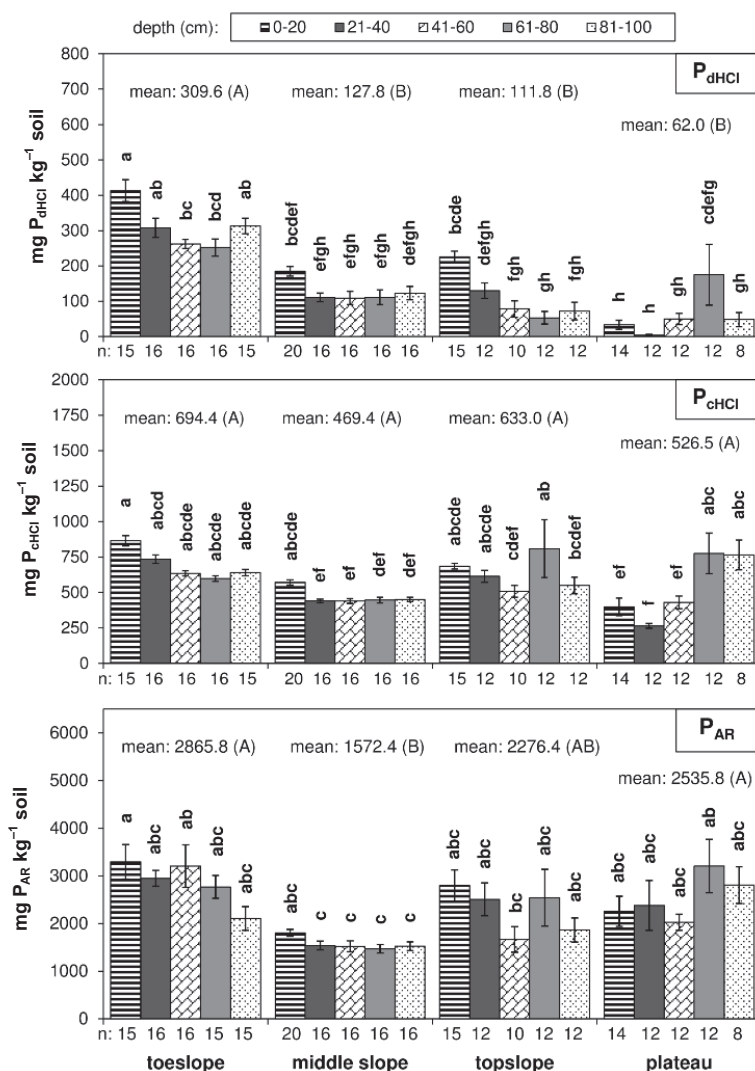
### 3.2.2 Topslope

The topslope soils contain a mean of 90% more P<sub>dHCl</sub> and 27% more P<sub>cHCl</sub> than the plateau sites (Tab. 2). P<sub>AR</sub> occurs on average 15% less. Mean P<sub>dHCl</sub> values are highest in the top 20 cm of soils and tend to decrease with depth (Fig. 2). The periglacial solifluction layers derived from limestone have the lowest values (profiles 13, 14; Fig. 3). Instead, loess-containing solifluction layers and colluvial slope sediments are significantly higher in P<sub>dHCl</sub> (profile 11; data not shown).

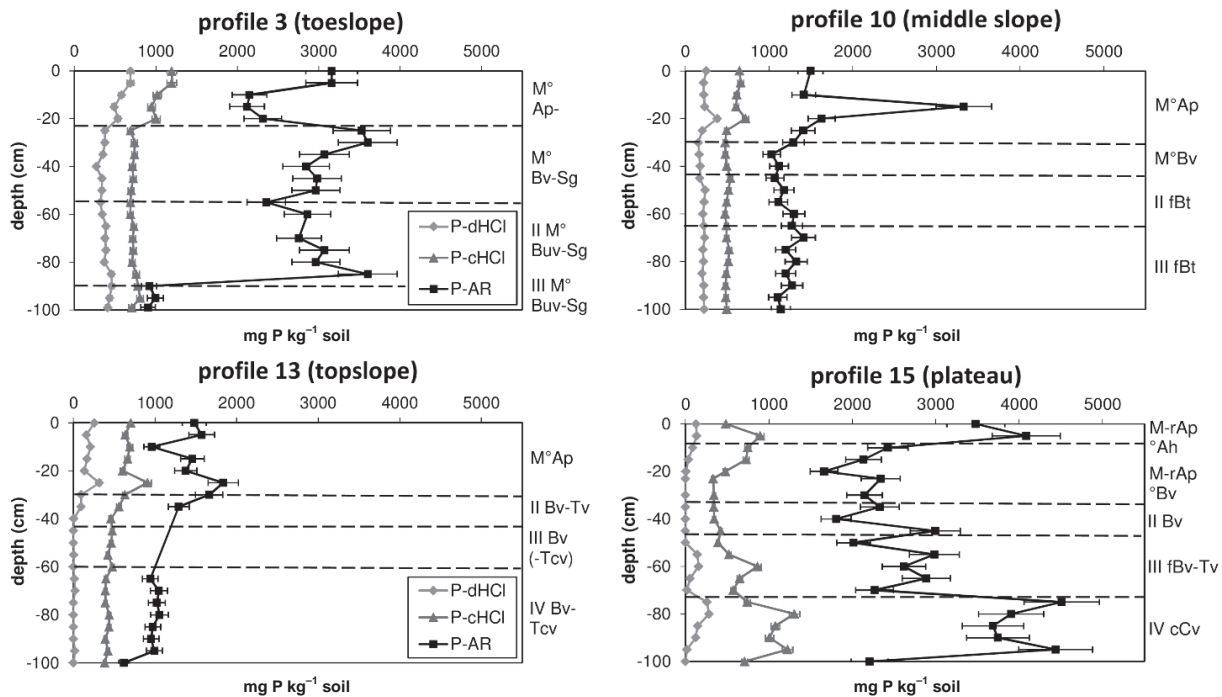
P<sub>cHCl</sub> shows no such substratum-specific trend. No significant effect of depth was found either (Fig. 2). Still, in some soils the plough layer contains slightly more P<sub>cHCl</sub> than deeper profile sections (e.g., profile 13; Fig. 3). This also applies for P<sub>AR</sub>, without a significant depth-specific trend (Fig. 2). In contrast to P<sub>dHCl</sub>, P<sub>AR</sub> is significantly higher in profile 14 than in profiles 11 and 13, without substratum specificities (data not shown).

### 3.2.3 Middle slope

The middle slope soils comprise 11% more P<sub>dHCl</sub>, but 26% less P<sub>cHCl</sub> and 48% less P<sub>AR</sub> than the topslope sites (Tab. 2). In each P fraction, higher contents tend to occur in the top 20 cm of the soils (Fig. 2). The lower sections of the profiles show no significant vertical variation (e.g., profile 10; Fig. 3).



**Figure 2:** Average P levels for 20-cm-soil-sections and slope positions. Columns represent means of *n* samples with standard errors. Different letters in the same style of writing indicate significant differences between means ( $p < 0.05$ ).



**Figure 3:** Vertical P distribution for one example profile per slope section. Error bars are estimated ranges of analytical uncertainty, calculated as the sum of the average standard deviation of the method plus the average standard deviation of measured data from their fitted mean.

### 3.2.4 Toeslope

The toeslope soils contain 136% more  $P_{dHCl}$ , 46% more  $P_{cHCl}$ , and 148% more  $P_{AR}$  than the middle slope sites (Tab. 2). Thus, this slope position is on average richest in P (Fig. 2). The highest values occur in the top 20 cm of the soils. There is no significant vertical variation in the lower profile sections. Also, no substratum- and soil-horizon-specific trends were found.

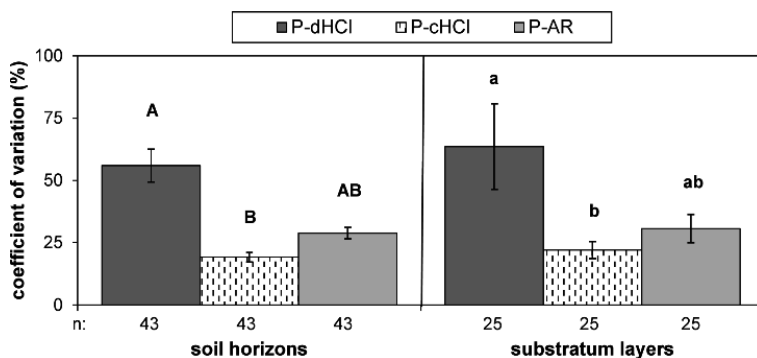
### 3.3 Spatial P variation

$P_{dHCl}$  values differ most from the mean P level of single soil sections (Fig. 4). The average coefficient of variation (CV) is 56% for soil horizons and 64% for substratum layers.  $P_{cHCl}$

shows less variation, but still has a mean CV of 19% and 22%, respectively. For  $P_{AR}$  29% and 31% were calculated.

The vertical P variation within whole profiles was also determined. Figure 5 (top) shows the variation of the P data from their mean for each P fraction in each profile.  $P_{dHCl}$  differs on average by 72% from its mean values,  $P_{cHCl}$  and  $P_{AR}$  by 30% and 35%, respectively.

The lateral variation between P contents in different profiles is even higher. Profiles were split in 30-cm-sections, and the corresponding mean values were calculated for each profile. Within one section (e.g., 0–30 cm) the average of the profiles' means was calculated. The variation from these averages is shown in Fig. 5 (bottom).  $P_{dHCl}$  has the highest lateral variation between soil sections in different sites (mean CV: 86%).  $P_{AR}$  and  $P_{cHCl}$  data also differ greatly from site to site (65% and 40%, respectively).



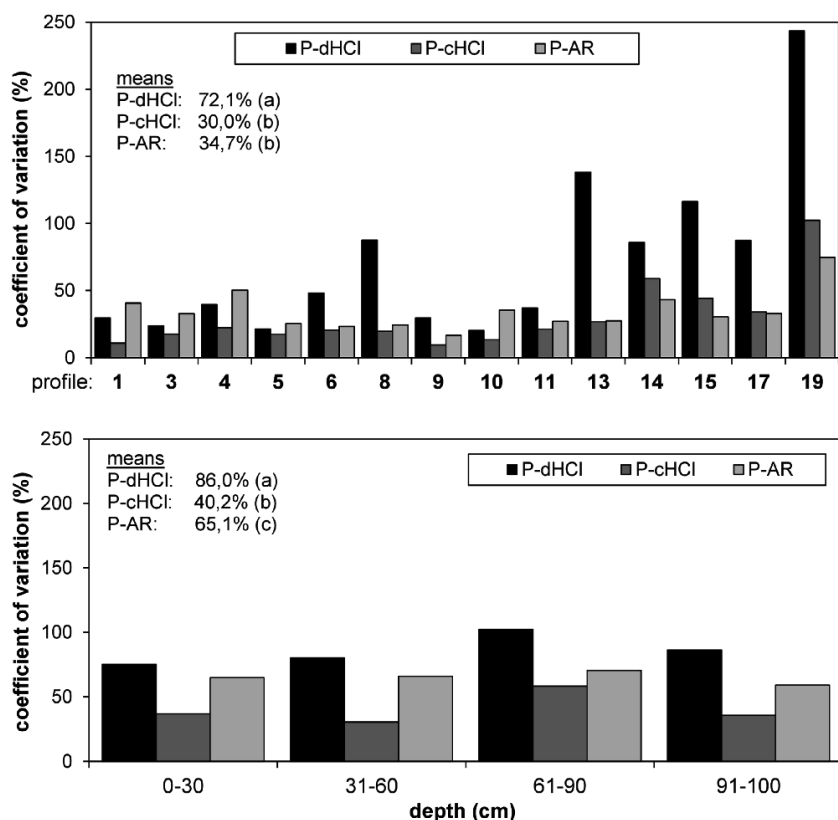
**Figure 4:** Vertical variation of P contents within soil sections. Columns represent average CV of the CV of each soil section's mean P content, and standard errors of the average CV. Different letters in the same style of writing indicate significant differences between average CV ( $p < 0.05$ ).

## 4 Discussion

### 4.1 Plateau

We interpret  $P_{dHCl}$  as a kind of proxy for soil solution P, because it can easily be adsorbed and exchanged back into the solution according to local conditions (e.g., moisture). On the plateau, the colluvial slope sediments and upper solifluction layers are nearly  $P_{dHCl}$ -free (Fig. 3). Easily accessible P could have been taken up by plants, but this is unlikely because plants would probably cause a constant supply of bioavailable P by root exudation of organic acids and





**Figure 5:** Top: Vertical P variation within soil profiles. Columns represent CV from the mean P content of each profile. Bottom: Lateral P variation between 30-cm-sections of the investigated profiles. Columns represent CV from the mean P content of the average P levels of each profile in the according soil section. Different letters indicate significant differences between mean CV of the P fractions ( $p < 0.05$ ).

resulting P mobilization during the vegetation period (Oburger et al., 2011; Alamgir and Marschner, 2013). On the other hand, vertical translocation of dissolved P within the silty upper parts of the profiles and lateral translocation from higher up the slope could have occurred. As a result, the clayey, compact solifluction layers and the limestone weathering layers would have become enriched with  $P_{dHCl}$ .

Furthermore, replacement of P anions by  $OH^-$  and  $HCO_3^-$  is probable in these substrata (Binner et al., 2015; Prietzel et al., 2016). The deeper solifluction layers and the weathering layers are neutral to slightly alkaline, and high in carbonate (Tab. 1). Weathering of carbonate enriches the soil solution with  $Ca^{2+}$  and  $HCO_3^-$ . Thus, P sorption would be reduced. Resulting high concentrations of P anions in the soil solution then could precipitate with the also quite abundant  $Ca^{2+}$  in the form of discrete Ca-P-minerals (Barber, 1995; Samadi, 2010; Oburger et al., 2011). Hence, high amounts of  $P_{cHCl}$  and an even higher percentage of  $P_{AR}$  were detected in the lower parts of the soils (Fig. 3). This implies that recalcitrant P minerals (e.g., hydroxyapatite, Octa-Ca-phosphate) precipitate more often in these layers than more easily soluble Ca-P-minerals (e.g., Mono-Ca-phosphate).

Furthermore,  $P_{cHCl}$  and  $P_{AR}$  contents could be increased by the sorption of multiple P ion layers to calcite (Samadi, 2010). According to Holford and Mattingly (1975), P first is chemi-

sorbed to calcite in a monolayer at P concentrations of  $< 0.5 \mu g mL^{-1}$ . Higher concentrations instead foster the physical, low-energy sorption of multiple P layers on the monolayer, from which finally Ca-P minerals (e.g., octa-Ca-phosphate) precipitate. The latter processes thus occur when the soil solution is severely oversaturated with P and Ca ions (Alvarez et al., 2004; Reddy et al., 2005), which could be the case in the investigated soils at certain times during the year (e.g., dry spans in summer). Additionally, high  $P_{cHCl}$  contents may result from the specific sorption of P anions to Fe-/Al-oxyhydroxides in the clay fraction (Holzmann et al., 2016). As shown in Tab. 1 the lower parts of the profiles are rich in clay.

Its high P content can hardly be explained by a “geogenic loading” derived from the original bedrock. Carbonate rocks are often low in P. Pecoroni et al. (2014) report  $131 mg P kg^{-1}$  dry matter for carbonate rocks in Hesse. Others give  $300\text{--}400 mg P kg^{-1}$  dry matter (Rösler and Lange, 1965; Blume, 2004; Linhardt and Zarbok, 2005), which is in the range of sandstones (Pecoroni et al., 2014). Thus, the deeper profile parts on the plateau are enriched with P more than the bedrock seems to explain. This could be due to P translocation, which might occur vertically in the profiles and laterally with the sloping of the plateau. In this case,

P-depleted profiles should be found somewhere further up the plateau. Yet, another possibility must be taken into account. The weathering of limestones which are poor in P creates a proportionately smaller mass of residual clay in which the original amount of P might possibly be retained almost completely. Thus, a concentrating effect could have taken place next to limestone weathering, which would have enriched the plateau soils with P originally stemming from bedrock. Thus, the P content in the emerging soil would be explained not by the bedrock’s P content, but rather by secondary features originating from bedrock weathering. Based on our approach no final decision could be made about this question.

## 4.2 Topslope

The high average amounts of  $P_{dHCl}$  and  $P_{cHCl}$  on the topslope might be due to fertilization. Yet, only small amounts of liquid biogas manure have been applied on the field recently and strong enrichment of plant-available P forms is unlikely. Furthermore, P contained in liquid fertilizer could easily leave the topslope via surface runoff. Slightly soluble  $P_{cHCl}$  could instead stem from past land management in times before fertilization was adjusted to soil P status. Still, it is surprising that  $P_{AR}$  did not become proportionately enriched if this was the case. In general, the P fractions are dispersed quite heter-

ogeneously within the topslope soils, so land use and fertilization do not appear to be the main drivers behind that.

On average,  $P_{AR}$  is probably less common on the topslope because soils here are mostly low in carbonate (Tab. 1) and, thus,  $Ca^{2+}$  for precipitation of Ca-P-minerals is less abundant. Only profile 14 contains 66% more  $P_{CHCl}$  and 183% more  $P_{AR}$  than the other topslope soils. Its limestone-derived substrata correspond to the plateau sites. They are rich in Ca and carbonate and can therefore adsorb and precipitate P in recalcitrant forms, thus decreasing easily soluble P (Alvarez et al., 2004; Reddy et al., 2005).

Profile 13 is low in  $P_{dHCl}$  beneath the topsoil, but low in  $P_{AR}$  as well.  $P_{dHCl}$  may have been decreased here by lateral P translocation down the slope. This also seems to apply for  $Ca^{2+}$  from the weathering of carbonate. Profile 13 contains on average 89% less Ca than profile 14 (data not shown). Hence, the reagents for adsorption and precipitation of P are scarce, thus explaining the low  $P_{AR}$ .

The loess-derived materials in profile 11 are free of carbonate and 86% lower in Ca than profiles 13 and 14 (data not shown). Thus, more dissolved P might become physisorbed as  $P_{dHCl}$ . Therefore, the loess-based substrata could be significantly lower in  $P_{CHCl}$  and  $P_{AR}$  and higher in  $P_{dHCl}$  than those formed from limestone. This lateral variation in the P contents is unlikely to be explained by P inputs via fertilization. The high content of  $P_{dHCl}$  throughout profile 11 might instead be due to the replacement of P anions by  $OH^-$  (Amberger, 1996; Binner et al., 2015). This could only affect the whole profile when the material is deeply moistened. Perhaps the silty, compacted texture of the loess loam fosters these processes by holding large amounts of immobile water. This would enable long-term contact between the soil matrix and solution, and thus intensify reactions. Also, we assume that profile 11 was laterally enriched with dissolved P from higher up the slope (e.g., by translocated fertilizer). Still, it is unlikely that this process results in P graphs as vertically homogeneous as in profile 11 and the middle slope sites (e.g., profile 10; Fig. 3).

Regarding the vertical P distribution, the highest  $P_{dHCl}$  values occur in the plough layers. This applies for all the agricultural sites and could be explained by fertilization and biogenic P mobilization in the rhizosphere (Barber, 1995; Kruse et al., 2015). Furthermore, organic residues and the applied liquid biogas manure can chelate Fe, Al, and Ca ions on the surfaces of oxides, thus preventing chemisorption of P in less easily soluble forms (Oburger et al., 2011; Binner et al., 2015; Kruse et al., 2015). Still, we also found high  $P_{CHCl}$  and  $P_{AR}$  contents in the plough layers, what might partly be due to organic residues from harvest. Furthermore, the  $P_{CHCl}$  and  $P_{AR}$  contents rise with  $P_{dHCl}$ , because the P fractions determined here are cumulative.

#### 4.3 Middle slope

We interpret the P patterns of the middle slope sites analogously to profile 11 (see above). Their high  $P_{dHCl}$  contents could be due to fertilizer input. Yet, it seems unlikely that this would affect the whole profiles, but mainly the topsoils

(Fig. 3). Also, only small amounts of P fertilizer were applied here and no such strong enrichment is to be expected. It is possible that the high  $P_{dHCl}$  amounts, especially those in the subsoils, result from lateral translocation of dissolved P from the topslope. Additionally, the replacement of P anions by  $OH^-$  and mobilization of P during the reduction of Fe-oxyhydroxides is to be expected (Reddy et al., 2005; Syers et al., 2008). These processes are probably favored by the silty soil texture (Tab. 1) and climate, causing relatively long spans with immobile water in the profiles.  $P_{CHCl}$  and  $P_{AR}$  are instead less common on the middle slope. The loess-based substrata are poor in Ca and carbonate and could thus sorb less P and precipitate less Ca-P-minerals. Hence, the high  $P_{AR}$  content in profile 10 (Fig. 3) seems to represent local specificities, e.g., pores coated with calcite and thus sorbing and precipitating more P (Blume et al., 2010).

The vertical homogeneity of the P graphs on the middle slope (Fig. 3) indicates that land use and fertilization are not major influences for the spatial P distribution here. P inputs would probably cause enrichment of the topsoils compared to the subsoils. The rather homogeneous P graphs could instead result from the loess loam being originally very similar, as texture implies (Tab. 1). Still, the other soil characteristics change with depth and indicate that chemical conditions vary as well. It also seems unlikely that agricultural vegetation would cause no variation in the P graphs. Finally, there are pedogenetic differences among the horizons of the Cambisols and Luvisols, e.g., in clay content. Thus, vertical variation in the P levels is to be expected, although the substrata are homogeneous in texture. As such, the slight vertical variation in the P contents on the middle slope is interpreted as secondary. Silty soils poor in clay and high in adsorbed Ca develop a net of fine pores which can hold high amounts of immobile water (Müller, 1997; Blum, 2012). This favors reactions between the soil matrix and solution. Also, in such a net of immobile water, widespread ionic diffusion between concentration gradients could be fostered, thus equalizing the P distribution (Wang and Chu, 2015). This seems to affect the middle slope profiles completely, as is reported common for loess-based soils (Lieberoth, 1982).

#### 4.4 Toeslope

The toeslope has the highest average P contents without the occurrence of any geological or land use change. Therefore, the loess-loam-derived soils probably have been enriched with P. This might, at least in part, be due to fertilizer P that could have been translocated down the slope in surface runoff and have finally seeped into the toeslope profiles. Furthermore, the high P contents could directly result from the colluvial slope sediments, but coming from the middle slope it seems unlikely for them to be so high in P originally. Also, the colluvial sediments are not much higher in humus and clay and, thus, could not significantly sorb more P than the middle slope sites (Tab. 1). However, the stagnic toeslope sites contain carbonate, are high in EC and have significantly higher pH than the middle slope soils. Regarding the soil conditions of the other reference profiles, this could hardly be an original feature of the colluvial sediments. Hence, the carbonate probably precipitated secondarily because high concentrations of

$\text{Ca}^{2+}$  and  $\text{HCO}_3^-$  leached down the slope. The high EC also implies the presence of many ions in the soil solution. Thus, because of pH and carbonate  $\text{OH}^-$  and  $\text{HCO}_3^-$  might cause intensive P mobilization (Binner et al., 2015; Priezel et al., 2016) and, thus, high amounts of  $\text{P}_{\text{dHCl}}$ .

Additionally, lateral input of dissolved P from the middle slope seems possible. Thus, the immobilized water on the middle slope probably is not completely immobile but might move slowly down the hill (Müller, 1997) transporting solutes to the toeslope. This also applies for Ca, which is on average 107% higher in the toeslope sites than on the middle slope (data not shown). Hence, like on the plateau and topslope, the toeslope has favorable conditions for the sorption of P to carbonate and for the precipitation of Ca-P-minerals. As a result, more  $\text{P}_{\text{cHCl}}$  and  $\text{P}_{\text{AR}}$  were detected on the toeslope than on the middle slope.

Still,  $\text{P}_{\text{dHCl}}$  and  $\text{P}_{\text{AR}}$  increase most, probably due to climate and the stagnic characteristics of the toeslope soils (Fig. 3). Like on the middle slope, the silty substrata can hold high amounts of immobile water (Blume et al., 2010; Glaesner et al., 2011b). This favors intensive P mobilization and, after drying, the formation of  $\text{P}_{\text{dHCl}}$ . During the wet phases the soil solution might often become oversaturated with P anions which precipitate with  $\text{Ca}^{2+}$  laterally leached down the slope (Barber, 1995; Reddy et al., 2005; Özgül et al., 2012). Thus, wet conditions would foster the formation of  $\text{P}_{\text{AR}}$ . Therefore, the stagnic environment seems to strongly facilitate rising  $\text{P}_{\text{dHCl}}$  and  $\text{P}_{\text{AR}}$  levels. Instead,  $\text{P}_{\text{cHCl}}$  would probably rather be favored by longer dry phases and decrease with P mobilization. Yet, drying would also raise pH, thus lessening P chemisorption to the mainly negatively charged soil minerals (Barber, 1995; Binner et al., 2015; Priezel et al., 2016). Hence, wet and dry conditions probably do not favor  $\text{P}_{\text{cHCl}}$  formation in these soils.

However, the strong moistening of the profiles veils P-related specificities of substrata and soil horizons. It also seems to influence vertical P distribution. Like on the middle slope,  $\text{P}_{\text{dHCl}}$  and  $\text{P}_{\text{cHCl}}$  are very homogeneously dispersed below the plough layers on the toeslope. Again, we interpret that to be a result of diffusive P migration in wet phases, equalizing deviation between P concentrations of different locales (Reddy et al., 2005; Glaesner et al., 2011a). These processes are known to bridge short distances though ( $0.13 \text{ mm d}^{-1}$ ; Marschner, 1995, after Syers et al., 2008). Yet, we assume that P concentrations of larger soil sections could become vertically and laterally equalized, when (1) there is much immobile water, (2) the concentration of P ions in the solution is high, (3) the tortuosity of the pore spaces is low (Glaesner et al., 2011c; Rakotoson et al., 2016; Margenot et al., 2017), and (4) when moistening does not occur over too short a duration. These conditions seem to be fulfilled in the toeslope soils during longer phases in the year.

However,  $\text{P}_{\text{AR}}$  is less homogeneously distributed (e.g., profile 3; Fig. 3). The precipitation reactions forming Ca-P-minerals strongly depend on the inputs of Ca and P ions (Barber, 1995; Reddy et al., 2005). These could happen in different depths according to where the ions are leached from. Further-

more,  $\text{P}_{\text{AR}}$  precipitation depends on the degree of soil solution oversaturation with P and Ca ions (Alvarez et al., 2004; Reddy et al., 2005). This again is strongly influenced by temperature, evaporation and the drying of soils. Thus, probably the plough layer in profile 3 is relatively low in  $\text{P}_{\text{AR}}$  (Fig. 3) because it dries faster. This would concentrate the soil solution in the deeper profile sections, increasing the ion concentrations there and fostering precipitation of  $\text{P}_{\text{AR}}$ . Instead, the low  $\text{P}_{\text{AR}}$  contents in > 90 cm of depth in profile 3 (Fig. 3) imply the loss of P ions. This could possibly happen when the groundwater table oscillates according to changes in the water table of the nearby brook (Fig. 1). Thus, precipitation would be lessened.

#### 4.5 Spatial P variation

As soil P originally comes from bedrock (Smeck, 1985; Kruse et al., 2015), bedrock and substratum are often expected to shape the spatial P distribution most (Blume et al., 2010; Fu et al., 2010; Pecoroni et al., 2014). In our study bedrock/substrata by their geogenic loading draw a general frame of P levels to be expected in a certain area. Still, no significant substratum-based trends were identified. Also, the vertical and lateral variation within the spatial P distribution do not generally seem to be due to the occurrence of certain bedrocks/substrata. Our P contents had on average high CV from the mean P levels in single soil horizons and substratum layers (Fig. 4). This shows that spatial P variation within soil sections is common, even if they developed from the same material or were subject to the same pedogenetic trend. Hence, bedrock/substratum or the kind of soil horizon do not generally shape the spatial P distribution most.

Land use can significantly veil the geological P legacy of soils. Still, it can hardly explain the strong lateral and vertical variation of the spatial P distribution on the examined agricultural field generally receiving low P fertilizer inputs. An anthropogenic P legacy from former times cannot be excluded, but is not reflected by the P data. Thus, the detected variation in the spatial P distribution could be interpreted as the result of P translocation (hypothesis 1). This P might to some degree stem from anthropogenic inputs (e.g., fertilizer in leachate).

A large part of P translocation is assumed to happen underground (hypothesis 2). Above-ground processes, like erosion and surface-runoff, could not primarily explain the high P contents of the toeslope in our study, although this interpretation is commonly found in literature (Moore et al., 1993; Halbfäß and Grunewald, 2003; Pecoroni et al., 2014; Bol et al., 2016). On the investigated toeslope, not only the topsoils are P enriched, but the entire profiles (Fig. 2). This might at least in part have been driven by prior superficial translocation (e.g., surface runoff), but only if it finally seeped into the profile so that P thereafter migrated underground. Also, the colluvial slope sediments (e.g., humus, texture) are not much different from the soils on the middle slope (Tab. 1), but are much higher in P. Thus, colluvial slope sediments are not necessarily P-enriched.

Consequently, the high P contents on the toeslope could result from underground translocation. Such processes are often deemed negligible in literature (Godlinski et al., 2008;



Andersson et al., 2015; Bol et al., 2016; Stewart et al., 2017), except for preferential flow (Glaesner et al., 2011c; Ulén et al., 2013; Sharpley et al., 2014). Little is known about how it affects the spatial P distribution, because preferential flow was often investigated with lysimeters (Godlinski et al., 2008; Andersson et al., 2015; Bergström et al., 2015). Generally, one would expect macro-pore flux to dislocate solutes and particles vertically along the preferential flow paths (Blume et al., 2010; Glaesner et al., 2011a; Sharpley et al., 2014). Thus, it is a very local phenomenon. It is improbable that all our soil cores accidentally and completely cut through preferential flow paths. Hence, macro-pore flux could not systematically explain the observed P distribution in the Mihla transect, what confirms our hypothesis 2.

Regarding the vertical and lateral variation of the P contents (Fig. 5), it is clear, that  $P_{dHCl}$  is spatially least evenly distributed. Thus, we conclude that underground P translocation mainly affects dissolved P, which probably was weakly adsorbed as  $P_{dHCl}$  while drying the samples for analysis. Our soils directly received dissolved P within liquid manure, which could under certain conditions be easily translocated. With the drying of the soils it would have been sorbed onto particles. Afterwards, dissolved P results from mobilization, which needs water as a medium. Hence, underground P translocation seems to be fostered by P mobilization under moist soil conditions (hypothesis 3). In the Mihla transect this might be a quite common process, in light of the high pH (repulsion on negative mineral surfaces, replacing of P anions by  $OH^-$ ).

Still,  $P_{cHCl}$  and  $P_{AR}$  also show relatively high spatial variation. The P fractions strive for developing equilibria with each other (Barber, 1995; Reddy et al., 2005; Syers et al., 2008). Thus, changes in dissolved P levels affect the other P fractions. Their formation generally lasts longer the more strongly they are bound. Hence, precipitation/dissolution takes more time than sorption/desorption (Holford and Mattingly, 1975; Smeck, 1985; Walpersdorf et al., 2013). Therefore,  $P_{AR}$  in our data is probably more unevenly distributed than  $P_{cHCl}$ . This might be due to the heterogeneous spatial occurrence of conditions where Ca-P-minerals can precipitate. Also,  $P_{AR}$  equilibrates slowly, so spatial heterogeneities last longer. Additionally,  $P_{cHCl}$  is intermediate between  $P_{dHCl}$  and  $P_{AR}$  and thus equilibrates in two directions. Instead, the latter equilibrate in one direction each. Therefore, it seems plausible that  $P_{cHCl}$  is not second in the ranking of heterogeneously spread P fractions.

After all, our methodology is spatially re-dissolved. On this basis, it is tricky to interpret P data in the context of the interplay of mobilization, leaching, secondary enrichment, and uptake by plants and microorganisms. Thus, our data represent a snapshot in time (Leinweber et al., 1999). Conditions and P amounts may vary greatly when investigated at different times. Finally, anthropogenic land management must be taken into account. P inputs and outputs can significantly alter the spatial P distribution. Nevertheless, they do not seem to be of major importance in our investigation due to the low P inputs and the strong vertical and lateral variability of P contents within the examined agricultural field. Still, more information on P management before the 1990s would have been desired for confirming our interpretations.

## 5 Conclusions

The spatial P distribution in a soil transect along a slope of loess–limestone origin was depicted. The data indicate that bedrock/substratum and land use are not the major influences for the vertical and lateral occurrence of P and that the nutrient is not spread homogeneously within soils and landscapes. Especially the toeslope is strongly P-enriched, probably in large part due to underground migration of dissolved P down the hill. Thus, “P fixation” in Ca-P-minerals need not necessarily be the dominant process affecting P in Ca-rich and alkaline soils. Instead, the status of equilibria between P fractions in the continuum of solubility (Syers et al., 2008) seems to determine which process prevails and how much P is plant-available. Therefore, easily soluble P forms are generally most unevenly distributed in our study, implying that the soil moisture regime strongly influences P mobility, chemically and spatially speaking.

High vertical variation of P contents within soil horizons and substratum layers was found. Hence, P research and management based on the examination of mixed samples summarily representing such soil sections seem inappropriate when accurate data is needed. These findings also raise the question of whether the spatial distribution of P is represented adequately in agricultural P management in Germany when determining the amounts of fertilizer to be applied. Currently, there is no adapted mode of soil sampling in this context. Thus, topslopes and toeslopes on the same field are commonly fertilized in the same manner. Instead, toeslope sites are often P-rich and should receive special treatment. This could help prevent ecological issues (e.g., eutrophication), but is also necessary for an effective and resource-efficient P management.

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