

## Is the Atmosphere an Important Source of Phosphorus in Northern Poland?

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### Abstract

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In this paper we determined the phosphorus (P) concentration in precipitation, and calculated the annual P deposition rate at three study sites located in northern Poland. We observed that the mean annual volume-weighted concentration of mineral phosphorus ( $P_{\min}$ ) in wet deposition varies from site to site. The estimated annual wet deposition rate of  $P_{\min}$  in the study region amounted from 0.1 to 0.39 kg/ha. The concentration and deposition rate of  $P_{\min}$  in the southern part of the study region was significantly lower than that in the northern and central part. We detected that the  $P_{\min}$  deposition was strongly positively correlated with the fraction of arable land in the study site vicinity, and also positively correlated with the mineral P fertilizer consumption in the area. The mean annual volume-weighted concentration of  $P_{\min}$  in wet deposition varied from 0.015 mg/l in the area of the lowest arable land percentage and the lowest mineral fertilizer consumption, to 0.046 mg/l in the area where the arable lands comprise 84% of agricultural land, and mineral P fertilizer consumption exceeded 33 kg/ha. In the P soil surface balance the atmospheric wet deposition represents 2.1–5.6% of annual total inputs of total P. The level of atmospheric P input varies widely by catchment and is related to land use (fertilizing, intensity and type of crop production).

**Keywords:** non-point source; nutrient; pollution; precipitation

Atmospheric deposition has been mainly recognized as an important nitrogen source, accordingly many existing reports deal with nitrogen atmospheric supply (SHEN *et al.* 2013; CUI *et al.* 2014). On the other hand, data on atmospheric phosphorus (P) deposition rates are quite scarce, and the understanding of P deposition is poorer than for many other substances (VET *et al.* 2014). In many studies P has been ignored as a minor precipitation constituent. Many nutrient budgets have been calculated on the basis of a few scattered literature values for P in rainfall, and definite measurements on its regional or continental variability are lacking (MORALES *et al.* 2001). Despite the fact that the atmospheric deposition has often been considered an insignificant source of P if compared e.g. to agricultural runoff or wastewaters, recent studies have suggested that it may represent its significant source (ANDERSON & DOWNIN 2006; ZHAI *et al.* 2009). ZHANG *et al.* (2011) reported that the atmospheric deposition of total phospho-

rus (TP) in Shanghai reached 0.31 kg/ha/year, and P concentration in rainfall amounted 0.03 mg/l. KOPACEK *et al.* (1997) (Czech Republic) also deciphered the annual TP deposition between 0.12 and 0.31 kg/ha. BERGAMETTI *et al.* (1992) found a higher deposition rate (about 0.4 kg P/ha) in the island of Corsica (north-western Mediterranean) and DAMMAGEN *et al.* (1994) reported TP annual bulk deposition of 0.68 kg/ha in south Germany grassland ecosystems. As shown by WINTER *et al.* (2002), atmospheric deposition was the largest source in annual TP budget of Lake Simcoe (Canada), and amounted 0.56 kg/ha.

In view of the above, our objectives were: (i) to determine the P concentration in rainfall and to calculate the annual P deposition in northern Poland; (ii) to determine the effect of land use on the inorganic P concentrations in precipitation; and (iii) to assess the relative importance of P atmospheric deposition in the P balance.

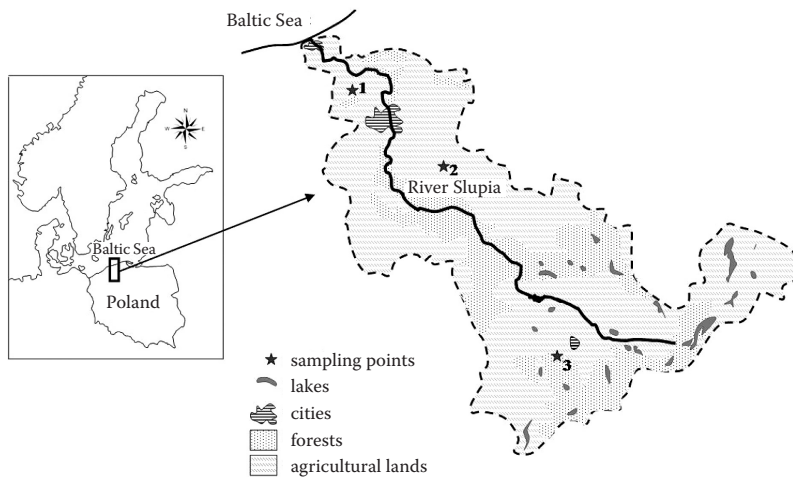


Figure 1. Location of measurement points

## MATERIAL AND METHODS

Measurements were carried out from 2007 to 2008 at three sites located in the catchment area of the River Slupia (northern Poland) (Figure 1). This region is less industrialized, and the main sources of air pollution are associated with agricultural activity (crop and animal production) and municipal services (transportation routes, household heating, low emissions from cities, etc.). In the study region, agricultural land constitutes about 65% of the catchment area. Arable land dominates (about 75% of agricultural land) and forests constitute about 35% of the area. The measurement points were distributed across the study region from the North – site 1 (54°30'09"N, 16°58'10"E) to the South – site 3 (54°05'07"N, 17°26'02"E). Site 2 (54°22'46"N, 17°08'49"E) was in the central part of the region. Description of the study catchment is presented in Table 1.

**Sampling and analysis.** At each of the three measurement sites one sampling point was used. Samples of precipitation waters (rain, snow) were collected using the Hellmann gauge and/or a polyethylene container, located in open space 1.5 m above ground to avoid small insects, debris, litter or other contaminants from the ground. Nutrient wet deposition was assessed, and rainfall samples were taken manually. Sampling

was performed from September 1<sup>st</sup>, 2007 to December 31<sup>st</sup>, 2008 at sites 1 and 2, and from January 1<sup>st</sup>, 2007 to December 31<sup>st</sup>, 2008 at site 3 (Table 2). The period of sample collection was 24 h. Rainfall volume (in mm) was monitored every day, whereas chemical analyses were made when the daily precipitation was sufficiently high (at least above 3 mm/day).

The concentration of inorganic phosphorus ( $P_{\min}$ ) in unfiltered rainwater samples was measured spectrophotometrically (SP-830 plus; Metertech, Taipei, Taiwan) at 690 nm, after reaction with ammonium molybdate, according to the standard method published by HERMANOWICZ (1976).

For each site the monthly (annual) volume-weighted mean  $P_{\min}$  concentrations were calculated. The monthly wet deposition (in kg/ha/month) of  $P_{\min}$  was calculated as follows:

$$D_{P_{\min i}} = 0.01 \times v_{mi} \times c_{P_{\min i}} \quad (1)$$

where:

$D_{P_{\min i}}$  – inorganic P deposition in month  $i$

$v_{mi}$  – total rainfall in month  $i$

$c_{P_{\min i}}$  – volume-weighted mean inorganic P concentration (mg/l) in month  $i$

0.01 – unit conversion factor

Table 1. Characteristics of the study catchment (source: National Agricultural Census 2010 (GUS 2010))

Site	Population density (person/km <sup>2</sup> )	Dominant soil texture	Production	Livestock density* (LU/ha)	Mineral P fertilizer consumption (kg P/ha)	Arable lands (%)
Site 1	53	clay, clay loamy	cereals, oilseeds	0.24	33.3	82
Site 2	32	loamy sand, sand	cereals, oilseeds, potatoes	0.31	7.1	69
Site 3	35	peat, loamy sand	grass, pastures, cereals	0.18	2.3	30

\*animal unit (LU) per ha (1 LU refers to 1.2 horse, one cow, three pigs, and 250 heads of poultry)

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Table 2. Annual data on rainfall depth, number and rainfall type for each of the sampling points

Site	Year	Sample collection period (h)	Annual rainfall volume (mm)	Days with precipitation	
				rain	snow
Site 1	2008	24	799.5	161	21
Site 2	2008	24	837.3	145	33
Site 3	2007	24	850.4	173	45
	2008	24	680.0	170	49

Annual deposition of  $P_{\min}$  (in kg/ha/year) was calculated by multiplying the annually volume-weighted mean  $P_{\min}$  concentration by the annual rainfall depth.

To estimate the ratio of atmospheric P deposition to TP inputs to the soil, the officially recommended OECD soil surface balance method was used. By this method the difference between the total quantity of P entering the soil and the quantity of P leaving the soil annually is determined. The P balance (in kg P/ha) was calculated for each site using the following formula:

$$F_M + (LU \times 14.9) + (2 \times A) + O = C + SD \quad (2)$$

where:

$F_M$  – mineral P fertilizers consumption

$LU$  – livestock density (in LU/ha)

14.9 – standard content of P in manure of one livestock unit (in kg P/LU)

$(LU \times 14.9)$  – natural P fertilizers consumption

$A$  – annual deposition of  $P_{\min}$

2 – coefficient based on the assumption that  $P_{\min}$  wet deposition constitutes 50% of TP wet deposition (KOPACEK *et al.* 1997; MORALES *et al.* 2001; ANDERSON & DOWNING 2006; JAROSIEWICZ & WITEK 2014)

$(2 \times A)$  – annual atmospheric deposition of TP (in kg/ha)

$O$  – other inputs, e.g. P in seeds and planting material

$C$  – P removed with marked and non-marked (grass) crops of each crop type (in kg P/ha)

$SD$  – phosphorus surplus (S) or deficiency (D)

The P soil surface balance was calculated for the smallest administrative units existing in Poland i.e. for gmina Słupsk (site 1), gmina Dębica Kaszubska (site 2), and gmina Bytów (site 3).

Statistical analyses were conducted in Statistica Ver. 10 software. To relate rainfall volume (in mm) to  $P_{\min}$  concentration (in mg/l), Spearman's rank-order correlation was used. To identify significant differences for the measurement sites, nonparametric variance (ANOVA followed by Kruskal-Wallis test) was applied. If differences were detected, the median of each data set was compared using the Dunn's *post-hoc* test adjusted *P*-values.

## RESULTS

**P concentration in wet deposition.** A total of 145 rainfall water samples collected from sites 1 and 2 during the period September 2007–December 2008 and from site 3 during the period January 2007–December 2008 were chemically analyzed. The analytical results and statistical description are listed in Table 3.

The  $P_{\min}$  concentration in precipitation ranged from undetectable to almost 0.15 mg/l. At site 1, the P concentrations in collected precipitation samples were generally less than 0.08 mg/l, and about 70% of the phenomenon was between 0.025–0.1 mg/l. At site 2, the  $P_{\min}$  concentrations ranged from 0 to 0.112 mg/l, however, for over 40% of the measured precipitation events it remained between 0.025 to

Table 3. Inorganic phosphorus ( $P_{\min}$ ) concentration in rainfall (mg/l) and annual wet deposition rate of  $P_{\min}$  and total phosphorus (TP) (kg/ha) at the statistical description of the three sampling sites

Site	$n$	Min/max	$P_{\min}$ annual volume-weighted mean concentration	Annual deposition rate	
				$P_{\min}$	TP*
Site 1	37	0/0.135	0.048	0.39	0.78
Site 2	47	0/0.112	0.040	0.36	0.72
Site 3	61	0/0.146	0.015	0.10	0.20

\*based on the total/inorganic phosphorus ratio (2:1) in wet deposition

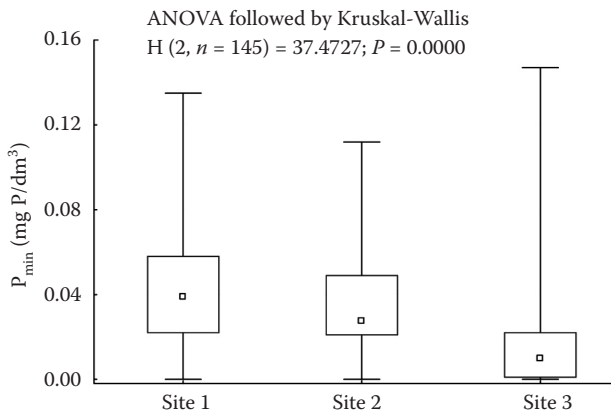


Figure 2. ANOVA analysis of inorganic phosphorus ( $P_{\min}$ ) concentration with the sampling sites; median (25–75%) (box) and minimum–maximum values (whisker)

*post-hoc* followed by Dunn

	site 1	site 2	site 3
site 1	–	ns	***
site 2	ns	–	***
site 3	***	***	–

\*\*\* $P < 0.0001$ ; ns – not significant

0.05 mg/l. The concentration of  $P_{\min}$  at site 3, situated most southerly, it was significantly lower (Figure 2) than at sites 1 and 2 ( $P = 0.0000$ ), and in 80% of the

measured rainfall phenomenon it amounted less than 0.025 mg/l. In the present study, the concentration of  $P_{\min}$  was not statistically significantly correlated with

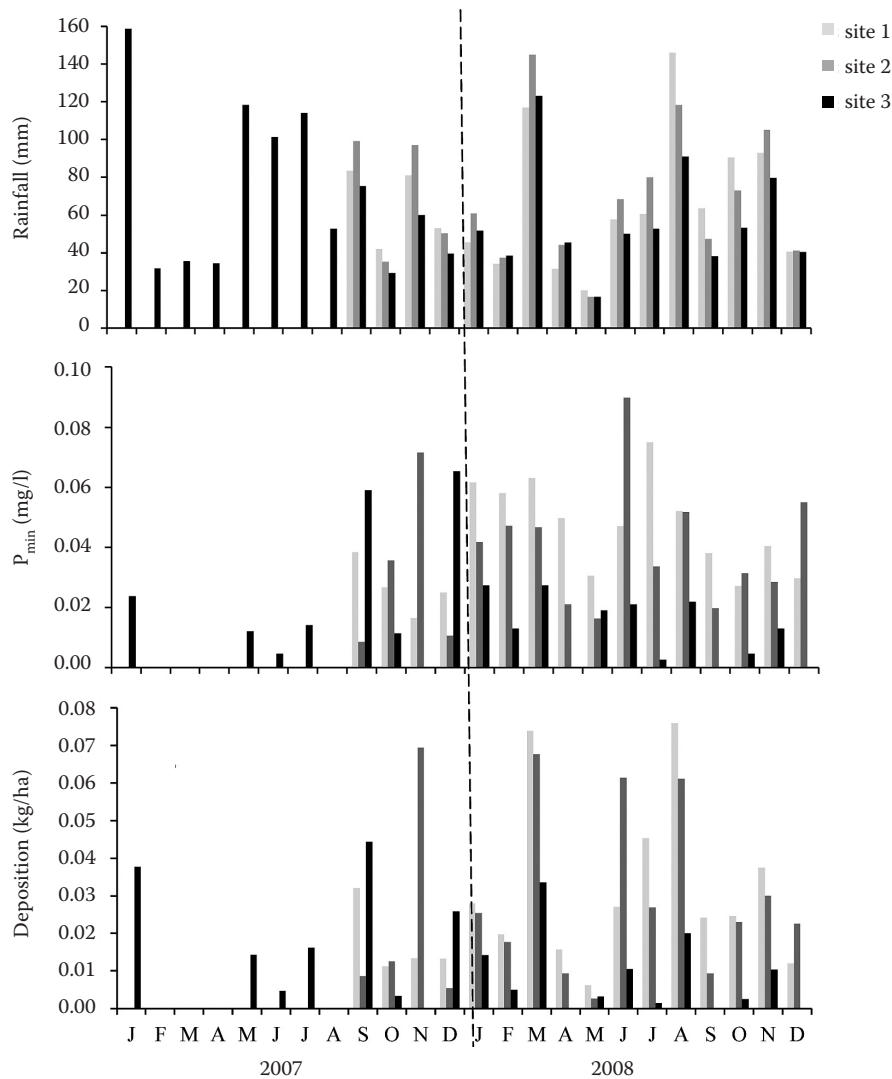


Figure 3. Monthly variations of rainfall (mm), volume-weighted inorganic phosphorus ( $P_{\min}$ ) concentrations (mg/l), and P deposition rate (kg/ha) at three measured sites

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the daily rainfall depth ( $P < 0.05$ ), and only at site 2 a weak positive correlation (statistical significance  $P < 0.1$ ) was noted.

There were obvious differences of monthly volume-weighted  $P_{\min}$  concentrations between the study sites, but no clearly distinguishable seasonal concentration changes were observed (Figure 3). Monthly volume-weighted  $P_{\min}$  concentrations ranged 0.016–0.063 mg/l at site 1, 0.008–0.09 mg/l at site 2, and 0–0.065 mg/l at site 3. Mean annual volume-weighted  $P_{\min}$  concentration amounted 0.048 and 0.040 mg/l for site 1 and 2, respectively, and 0.015 mg/l for site 3.

**Atmospheric P deposition rate.** The monthly wet deposition rates of  $P_{\min}$  changed conspicuously throughout the sampling period and sampling sites (Figure 3) and fluctuated in the study region from 0 to about 0.076 kg/ha. The highest depositions of  $P_{\min}$  at the three sampling sites generally occurred when the monthly total precipitation was the highest. In the northern part of the study area (site 1) monthly mean  $P_{\min}$  deposition in 2008 amounted to 0.032 kg/ha (3.2 mg/m<sup>2</sup>). The highest  $P_{\min}$  deposition rates, about 0.076 kg/ha, were observed in March and September (rainfall of 117 mm and 147 mm, respectively), while the lowest in May (0.006 kg/ha), when the monthly precipitation was the lowest (20 mm). In the central part of the region (site 2) the mean monthly  $P_{\min}$  deposition rate in 2008 was about 0.03 kg/ha. The highest P deposition rates, similar as at site 1, were observed in March and September: 0.067 and 0.061 kg/ha, respectively, and additionally in June (0.061 kg/ha) when the volume-weighted P concentration was the highest (about 0.09 mg/l). The lowest deposition rate was in May, the driest month, and amounted 0.006 kg/ha. At site 3 the  $P_{\min}$  deposition rate was about thrice smaller than in the rest of the area. The highest  $P_{\min}$  deposition noted in September 2007 and March 2008 amounted 0.044 and 0.037 kg/ha, respectively. In the remaining months the deposition fluctuated from less than 0.001 to 0.025 kg/ha.

The estimated annual wet deposition rate of  $P_{\min}$ -nutrient in the study region amounted 0.39, 0.36, and 0.10 kg/ha at sites 1, 2, and 3, respectively. Based on the total/inorganic P ratio (2 : 1) in wet deposition, the estimated annual wet deposition of TP varied from 0.2 kg/ha (site 3) to about 0.8 kg/ha (site 1) (Table 3).

## DISCUSSION

Our results of  $P_{\min}$  wet deposition at site 3 (0.1 kg per ha/year) represent one of the lowest values reported in Poland. In most studies wet deposition of  $P_{\min}$  attained

above 0.13 kg/ha/year and generally ranged from about 0.3 to 0.4 kg/ha/year, which corresponds to our  $P_{\min}$  concentrations at sites 1 and 2 (0.39 and 0.36 kg/ha/year, respectively). The lowest mean  $P_{\min}$  concentration in rainfall and the lowest annual deposition rate were those of MICHALSKA (2001), who reported a wet deposition rate of 0.13 kg/ha/year over the Upper Parsęta region (northern Poland). The highest concentrations were those of DURKOWSKI and KORYBUT WORONIECKI (2009) who reported mean annual wet concentration of 0.08 mg/l over the Lake Miedwie catchment basin (north-western Poland), and estimated the annual wet deposition rate of 0.4 kg/ha/year. SAPEK and NAWALANY (2004) measured a similarly high concentration of  $P_{\min}$  (about 0.07 mg/l) over north-eastern Poland, and deposition rates of 0.41 kg/ha/year. Both of these reports related to the agricultural catchment, with predominantly arable land.

**Effect of land use.** The atmospheric P mostly comes from soil-derived dusts and anthropogenic emissions: agricultural activity (fertilizers, animals breeding) and industrial sources (burning, automobile pollution) (BERGAMETTI *et al.* 1992; KOPACEK *et al.* 1997; AHN & JAMES 2001; POLLMAN *et al.* 2002; NYAGA *et al.* 2013). Additionally forest fires may also sporadically contribute to P deposition (BERGAMETTI *et al.* 1992; ZHAI *et al.* 2009). MAHOWALD *et al.* (2008) estimated that dust may be responsible for about 80% of P deposition, other sources for about 20%. They reported that globally about 5% of atmospheric P deposition originates from anthropogenic sources. Locally, the situation may completely differ, and the P deposition stemming from anthropogenic source may grow up to 50%. Additionally, AHN and JAMES (2001) reported, that the high spatial variability of P atmospheric loads suggests that the atmospheric deposition of P mainly depends on local sources. In our study, P deposition

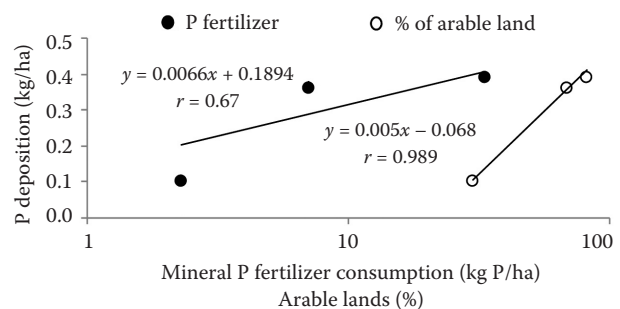


Figure 4. Correlations of annual mineral P fertilizer consumption to annual P deposition, and percentage of arable lands to annual P deposition, at three studied sites



Table 4. Phosphorus balance (soil surface balance method) at study sites; balance elements in kg P/ha of agricultural land

Site	P			Other inputs	P removed with crops	Surplus/deficit
	mineral fertilizer	natural fertilizer	atmospheric deposition			
Site 1	33.3	2.7	0.8	0.4	17.6	19.6
Site 2	7.1	4.7	0.7	0.4	14.9	-2.0
Site 3	2.3	3.6	0.2	0.4	10.2	-3.7

was strongly positively correlated with the fraction of arable land (Figure 4,  $r = 0.989$ ). The significantly higher  $P_{\min}$  concentrations and annual deposition rates were observed at sites 1 and 2 located in the immediate vicinity of arable land (82 and 69% of arable land, respectively). About thrice lower  $P_{\min}$  concentration, and about four times smaller deposition rate was observed at the site 3, where the arable land constitutes 39% of agricultural land. Estimated annual P deposition was also positively correlated with the mineral P fertilizer consumption (Figure 4,  $r = 0.67$ ). Similar mean annual  $P_{\min}$  concentration at sites 1 and 2 (0.048 and 0.040 mg/l) despite significantly different mineral fertilizer consumption (about 33 and 7 kg/ha, respectively) indicate that the types of soils and their permeability may affect the P concentration in rainfall. The less fertilized catchment area of site 2 is characterized by relatively light soils (loamy sands), which promote higher P losses. The catchment area of site 1 is formed of clay and clay loam.

Many studies have indicated, that the nutrient deposition rate is related to the degree of agricultural activity (KOPÁČEK *et al.* 1997; MCDOWELL & SHARPLEY 2009; NYAGA *et al.* 2013). LUO *et al.* (2007) reported that in the Taihu Basin (China) agriculture is one of the most important contributors to P deposition. This is supported by the results of MCDOWELL and SHARPLEY (2009) reporting the P deposition rate on woodland being about 5 times smaller than on cropland. According to KOELLIKER *et al.* (2004), sites in New Jersey under a localized application of P fertilizers showed both a high degree of variability and a higher concentration of P in precipitation (7.1–7.6  $\mu\text{g/l}$ ) than a site where no fertilizers were applied (5.8  $\mu\text{g/l}$ ).

**Importance of P atmospheric deposition.** In the P balance (Table 4), the atmospheric wet deposition represents from 2.1 to 5.6% of annual total inputs of TP. Despite the highest annual deposition rate (Table 3), the lowest TP input (in %) with atmospheric deposition was estimated at site 1. A very high annual P fertilizer consumption of 36 kg/ha with a high TP deposition rate might have contributed to P losses from soil, because only less than 50% of the P was taken up by the crops (surplus of 19.6 kg/ha). So a high surplus can stimulate an intensive biological

activity, which can lead to progressing eutrophication of aquatic and terrestrial habitats. Therefore, the atmospheric wet deposition input should be considered in P fertilization recommendations to mitigate unnecessary excessive P inputs at this study site. Totally, about 13 t of P fertilizer can be saved annually over the whole site 1 catchment.

Higher P inputs with atmospheric deposition in the P balance were registered at sites 2 and 3, (5.6 and 3.1% of total inputs, respectively). In contrast to site 1, in these agroecosystems atmospheric deposition partly compensates for the P deficiency (Table 4). This seems to be even more important for local farmers as about 30% of arable lands in the province are poor or very poor in available P (< 4.5 mg/kg).

## CONCLUSION

The annual wet deposition rate of  $P_{\min}$  in the study region amounted from 0.1 to 0.39 kg/ha. The estimated wet deposition of TP varied from 0.2 to about 0.8 kg/ha. The concentration and deposition rate of P in the southern part of the study region was significantly lower than that in the northern and central part.

The P deposition was strongly positively correlated with the fraction of arable land and mineral fertilizer consumption. The significantly higher  $P_{\min}$  concentrations and annual deposition rates were observed at sites located in the immediate vicinity of arable land. Additionally, the types of soils and their permeability may affect P concentration in precipitation.

In the P soil surface balance, the atmospheric wet deposition represents 2.1–5.6% of annual total inputs of TP. The significance of the P atmospheric input varies widely by catchment and is related to land use (fertilizer use, intensity and type of crop production, etc.).

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