Phosphate has a differential influence on arsenate adsorption by soils with different properties

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ABSTRACT

The adverse effect of the application of phosphorus (P) on arsenic (As) adsorption by soils can result in increasing mobility and availability of As. However, in different soils, P might influence As adsorption differently. In this study, the arsenate [$As^{(V)}$] adsorption capacities of six soils with different properties and the effects of P application were studied. The results indicated that the adsorbed $As^{(V)}$ contents all increased as a function of the $As^{(V)}$ content in equilibrium. When analysed using the Langmuir and Freundlich equations, the maximum $As^{(V)}$ adsorption capacity of 0.72 mg/g was found for an Alumi-Plinthic Acrisol, while the minimum capacity of 0.09 mg/g was observed for an Epigleyic Cambisol. The adverse effects of P application on $As^{(V)}$ adsorption by the six soils were observed to be variable. When the P/As molar ratio in a culture experiment was increased from 0 to 10, the maximal and minimal decreases in the $As^{(V)}$ adsorption capacity of 0.086 and 0.014 mg/g were found in the Alumi-Plinthic Acrisol and Epigleyic Cambisol, respectively. P was relatively more effective in competing for adsorption sites with $As^{(V)}$ in the Alumi-Plinthic Acrisol and Luvi-Endogleyic Phaeozem due to their higher A/P values (decrease in adsorbed As/added P) of 1.143 and 1.135, respectively. These results will help decrease the environmental risk of some As-contaminated agricultural soils through the controlled application of P.

Keywords: isothermal equation; adsorption capacity; P/As molar ratio; As availability

Arsenic (As) adsorption is an important means of fixation and plays a major role in controlling the mobility and availability of this element (Yolcubal and Akyol 2008). In agricultural soils in particular, the considerable adsorption of As by soils is conducive to decreasing As availability and reducing As accumulation in crops. Numerous studies are focused on As adsorption by individual soil minerals, such as on kaolinite, montmorillonite, illite (Saada et al. 2003), iron oxides (Taylor et al. 2009) and aluminium oxides (Beaulieu and Savage 2005). Comparatively less information is available regarding As adsorption in intact soils, although a number of studies have been conducted on this topic (Violante and Pigna 2002, Sahu et al. 2011). Individual soil components cannot truly model soils, and the adsorption of As is best studied using actual soils as adsorbents (Jiang et al. 2005a). Furthermore, phosphorus (P) can compete with As for adsorption sites in soils and correspondingly increase the mobility and availability of As because of their similar chemical behaviours (Smith et al. 2002). Studies have been carried out to explore the interaction between As and P adsorption on clay minerals, Fe/Al oxides/hydroxides, or intact soils (Liu et al. 2001, Violante and Pigna 2002, Pigna et al. 2009). However, the adverse effects of P application on As adsorption might vary in different soils. Therefore, further investigation is needed to identify the types of soil in which As adsorption is more strongly affected by the application of P and those in which it is only mildly affected.

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Thus, six soils with different properties were collected from five provinces in China. The primary objectives of this study were to (i) quantify the $As^{(V)}$ adsorption capacities of these soils using the Langmuir and Freundlich equations; (ii) assess the effects of P application on $As^{(V)}$ adsorption by these soils; (iii) determine the soil in which $As^{(V)}$ adsorption was strongly affected and those in which it is only mildly affected by P treatment. This work will shed light on how to decrease the environmental risk of some As-contaminated agricultural soils in future by controlling the application of P.

MATERIAL AND METHODS

Experimental soils. Six agricultural soils with different properties were collected from five provinces in China. A Luvi-Endogleyic Phaeozem (PHel) developed from the clay sediments was sampled from the Heilongjiang Province. An Epigleyic Cambisol (CMe) developed from the alluvial material was sampled from the Beijing City. A Rhodi-Calcaric Cambisol (CMcr) developed from purplish sandy shale of the Penglaizhen group was sampled from the Sichuan Province. A Calcari-Leptic Cambisol (CMlc) developed from the limestone was sampled from the Guizhou Province. An Alumic Acrisol (ACa) developed from granite and an Alumi-Plinthic Acrisol (ACpa) developed from quaternary red clay were both sampled from the Hunan Province. Each surface soil sample was obtained through the collection of 12–15 subsamples from a 20 × 20 m square. All soil samples were ground and passed through a 0.15 mm mesh sieve after being air-dried and were subsequently used to determine soil properties.

Soil properties. The soil organic carbon (C_{org}) content was determined using the potassium dichromate heating method (Nelson and Sommers 1982). Total P was determined after extracting with NaOH (Smith and Bain 1982). Available P was extracted using sodium bicarbonate (Olsen et al. 1954). The P content in the extracts was determined via the ammonium molybdate-ascorbic acid method (Murphy and Riley 1962). Cation exchange capacity (CEC) was determined by the method of Rhoades (1982). Total As was digested with HNO_3-H_2O_2 (2:1 v:v) (Fritzsche et al. 2006) and measured using hydride generation atomic fluorescence spectrometry (HG-AFS 9120, Jitian Instrument Co., Beijing, China; detection limit < 0.02 µg/L As). Available As was extracted with 0.5 mol/L $NaHCO_3$ and measured using HG-AFS. The Fe_{CD} and Al_{CD} in soil samples extracted with citrate-dithionite (CD) were determined according to the Manning and Goldberg's procedure (1997). The soil pH was measured at a 1:2.5 soil:water ratio using a pH electrode. The mineral composition of each soil sample was estimated using X-ray diffraction analysis (D/Max-RC, Rigaku, Japan).

$As^{(V)}$ isothermal adsorption. A soil sample weighing 1.0000 g was transferred to a 50 mL polypropylene centrifuge tube containing 20 mL $As^{(V)}$ solution. The examined concentrations of $As^{(V)}$ ranged from 1 mg/L to 100 mg/L. The pH of the $As^{(V)}$ solution was adjusted to 5.0 using NaOH or HCl. The soil suspensions were shaken for 24 h (25 ± 1°C, 180 rpm) and then centrifuged for 5 min (8000 × g) to obtain the supernatant. The total As concentration in the supernatant was determined via HG-AFS. The amount of $As^{(V)}$ adsorbed on the soils was calculated as the difference between the initial and the final As concentration in the supernatants. Sodium arsenate tribasic ($Na_3AsO_4 \cdot 12\ H_2O$) purchased from the Sigma Chemical Co. (St. Louis, USA) was used as the source of $As^{(V)}$. Three replicates were performed for each treatment. Throughout the cultivation period, no change in $As^{(V)}$ speciation was found in randomly selected samples subjected to analysis of the chemical speciation of As using the methodology of Zeng et al. (2011).

The Langmuir and Freundlich equations were used to fit the adsorption data and to further explore the differences in the maximum $As^{(V)}$ adsorption capacities of the different soils. The Langmuir one-surface (1); Langmuir two-surface (2), and Freundlich (3) equations are given as follows:

$$q = KMc/(Kc + 1) \qquad (1)$$

$$q = K_1M_1c/(K_1c + 1) + K_2M_2c/(K_2c + 1) \qquad (2)$$

$$q = bc^{1/n} \qquad (3)$$

Where: q and c – adsorbed $As^{(V)}$ (mg/kg) and the equilibrium concentration of $As^{(V)}$ (mg/L), respectively. In (1), M – maximum capacity of $As^{(V)}$ adsorption (mg/kg); K – adsorption equilibrium constant related to the bonding energy (L/mg). In (2), M_1 and K_1 – adsorption maximum (mg/kg) and adsorption equilibrium constant (L/mg) for the low-energy surface, respectively. M_2 and K_2 – adsorption maximum (mg/kg) and adsorption equilibrium constant (L/mg) for the high-energy surface, respectively. The total adsorption capacity is the sum of M_1 and M_2. In (3), b is the adsorption constant related to the maximum capacity of $As^{(V)}$ adsorption, and n is the adsorption constant related to the bonding energy.

Effects of phosphate on $As^{(V)}$ adsorption. A soil sample weighting 1.0000 g was transferred to a

Table 1. Major properties of six experimental soils

Soil types	C_{org}	total P	available P	total As	available As	Fe_{CD}	Al_{CD}	pH (H_2O)	CEC (cmol/kg)
	(g/kg)		(mg/kg)			(g/kg)			
PHel	44.68	0.91	42.59	7.18	0.14	10.59	0.76	7.62	25.4
CMe	17.52	1.33	31.03	6.56	0.06	10.10	0.19	7.95	15.8
CMcr	7.65	0.84	4.99	7.64	0.06	13.14	0.61	7.89	20.5
ACa	2.67	0.40	1.05	14.09	0.01	7.59	0.56	5.62	13.5
ACpa	12.08	0.66	15.46	18.71	0.03	32.63	1.06	4.60	12.4
CMlc	11.86	0.70	22.13	6.95	0.02	24.78	0.79	7.69	19.6

C_{org} – soil organic carbon; Fe_{CD} – citrate-dithionite extractable Fe; Al_{CD} – citrate-dithionite extractable Al; PHel – Luvi-Endogleyic Phaeozem; CMe – Epigleyic Cambisol; CMcr – Rhodi-Calcaric Cambisol; ACa – Alumic Acrisol; ACpa – Alumi-Plinthic Acrisol; CMlc – Calcari-Leptic Cambisol

50 mL centrifuge tube containing 20 mL of a mixed solution of phosphate and $As^{(V)}$. In the solution, the molar concentration of $As^{(V)}$ was maintained at 0.1 mmol/L, while the P/As molar ratio was brought to 0, 0.1, 0.5, 1, 5, or 10 by varying the P concentration. The content of $As^{(V)}$ adsorbed by the soils was determined using the aforementioned method. Phosphate was produced from $NaH_2PO_4·2H_2O$ (Beijing Chemical Works, Beijing, China). Three replicates were run for each treatment. No change in $As^{(V)}$ speciation was found using the method described above.

Quality assurance and quality control. A certified standard water-sample of As (GBWZ50004-88, Institute for Environmental Reference Materials, Ministry of Environmental Protection, Beijing, China), elemental spikes, and blanks were incorporated in batches of samples as quality control measures. The recovery (%) of As in the elemental analysis was up to 94% in this case. As standards were prepared using As stock solutions (GBW08611, Chinese Metrology Institute of Science and Technology, Beijing, China). After compulsively through the origin, the linear equation was obtained and the correlation coefficients all exceeded 0.9990. All glassware used in the experiment was cleaned by soaking in 10% HNO_3 for 12 h and then rinsed with ultra-pure water from a Milli-Q water purification system (Molsheim, France). All reagents were of analytical grade.

Data analysis. Fitting of adsorption isotherms and calculation of the parameters for equations (1); (2), and (3) were undertaken via nonlinear regression using Origin 8.0 (Northampton, USA). The significance analysis was performed on the obtained equations and parameters using *F*-test at the 0.01 level.

RESULTS AND DISCUSSION

Soil properties. Great differences in soil properties were found (Table 1). Among the six soils, the pH level in ACpa and ACa were the lowest, at 4.60 and 5.62, respectively. Both the Fe_{CD} and Al_{CD} contents were the highest in ACpa, at 32.63 and 1.06 g/kg, respectively. The CEC was the highest in PHel, at 25.4 cmol/kg. Based on the mineral

Table 2. Mineral composition (%) of six experimental soils

Soil types	Kaolinite	Montmorillonite	Illite	Chlorite	Quartz	Dolomite	Calcite	Feldspar	Amphibole	Iron ore
PHel	nd	nd	5	5	60	nd	10	15	5	nd
CMe	nd	nd	10	10	40	5	5	20	10	nd
CMcr	nd	nd	10	5	60	5	10	10	nd	nd
ACa	nd	nd	10	5	80	nd	nd	nd	nd	5
ACpa	40	nd	20	nd	20	nd	nd	20	nd	nd
CMlc	nd	nd	15	5	50	5	5	20	nd	nd

nd – not detected; PHel – Luvi-Endogleyic Phaeozem; CMe – Epigleyic Cambisol; CMcr – Rhodi-Calcaric Cambisol; ACa – Alumic Acrisol; ACpa – Alumi-Plinthic Acrisol; CMlc – Calcari-Leptic Cambisol

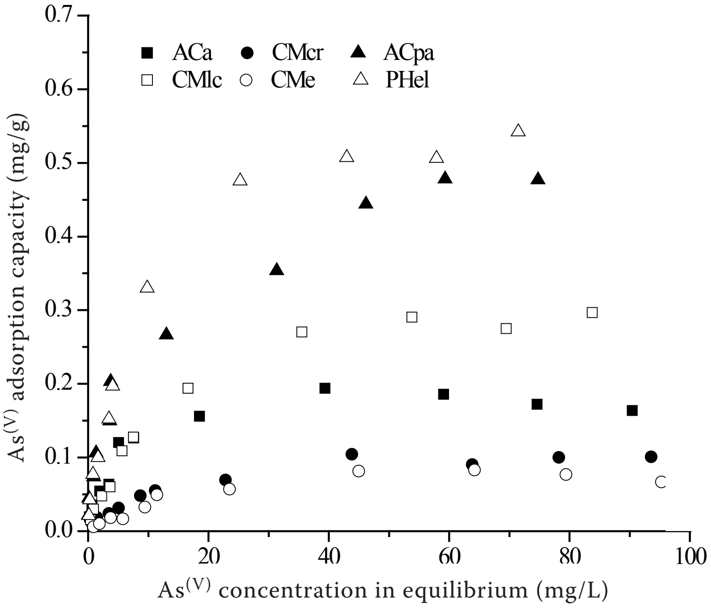

Figure 1. $As^{(V)}$ adsorption capacities in six soils with different properties as a function of the $As^{(V)}$ concentration in equilibrium. PHel – Luvi-Endogleyic Phaeozem; CMe – Epigleyic Cambisol; CMcr – Rhodi-Calcaric Cambisol; CMlc – Calcari-Leptic Cambisol; ACa – Alumic Acrisol; ACpa – Alumi-Plinthic Acrisol

composition analysis (Table 2), quartz represented the majority of the content in all of soils, except in ACa. Iron oxides/hydroxides were only detected in ACpa, while ACa exhibited a large percentage of kaolinite.

$As^{(V)}$ adsorption on soils and comparison of the adsorption models. Different $As^{(V)}$ adsorption capacities were obtained for the six soils (Figure 1). With an increase in the $As^{(V)}$ concentration in equilibrium, the $As^{(V)}$ adsorption capacities of the soils all showed an upward trend. During the equilibrium process, PHel showed the highest $As^{(V)}$ adsorption capacity, while CMe presented the lowest. Both the Langmuir one-surface and Freundlich equations showed a good fit for the $As^{(V)}$ adsorption isotherms (Table 3). In the Langmuir one-surface equation, the highest value of M, of 0.60 mg/g, was found for PHel. CMe presented the lowest absorption capacity of 0.09 mg/g. Similar results were observed using the Freundlich equation. The adsorption constant, *b*, of PHel was the highest, at 0.11, whereas it is the lowest for CMe, at 0.01. Comparing these two equations, the Langmuir one-surface equation-fitted $As^{(V)}$ adsorption might be more accurate than that of the Freundlich equation because of the higher correlation coefficient, with the exception of the value obtained in ACpa. Therefore, it could be inferred that in most of the experimental soils, there are a finite number of binding sites that are homogeneously distributed over the surface of the soils, showing the same affinity and no interaction between adsorbed $As^{(V)}$ (Aryal et al. 2011).

The Langmuir two-surface equation was also tested, but its fitting to the adsorption isotherm data was unsuccessful, except for ACpa and

Table 3. The fitting of $As^{(V)}$ adsorption by six soils with different properties with Langmuir one-surface and Freundlich equations

Soil types	Langmuir one-surface			Freundlich		
	M (mg/kg)	K (L/mg)	R^2	*b*	*n*	R^2
PHel	0.60	0.12	0.99	0.11	2.59	0.95
CMe	0.09	0.07	0.93	0.01	2.52	0.84
CMcr	0.12	0.08	0.97	0.02	2.65	0.93
ACa	0.19	0.24	0.95	0.06	3.87	0.83
ACpa	0.51	0.13	0.96	0.10	2.65	0.98
CMlc	0.35	0.08	0.99	0.05	2.38	0.94

Obtained equations and parameters all showed significance at the level of 0.01 via *F*-test; PHel – Luvi-Endogleyic Phaeozem; CMe – Epigleyic Cambisol; CMcr – Rhodi-Calcaric Cambisol; CMlc – Calcari-Leptic Cambisol; ACa – Alumic Acrisol; ACpa – Alumi-Plinthic Acrisol

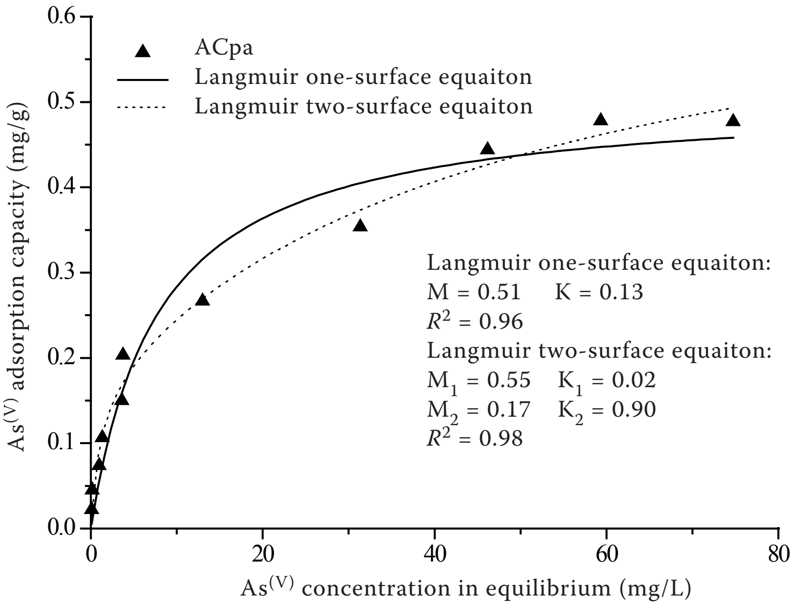

Figure 2. Comparison of the Langmuir one-surface and Langmuir two-surface equations for fitting to the adsorption of As$^{(V)}$ by ACpa. Obtained equations and parameters all showed significance at the level of 0.01 via *F*-test; ACpa – Alumi-Plinthic Acrisol

CMe. However, the correlation coefficient of the Langmuir two-surface equation fitting As$^{(V)}$ adsorption on CMe was lower than that of the Langmuir one-surface equation. Therefore, the corresponding result is not shown here. Compared to the Langmuir one-surface equation, the Langmuir two-surface equation-fitted As$^{(V)}$ adsorption on ACpa was more accurate due to the higher correlation coefficient (R^2 = 0.98) (Figure 2). The maximum absorption capacity of 0.72 mg/g (M_1 + M_2) for ACpa predicted using the Langmuir two-surface equation was obviously higher than that obtained using the Langmuir one-surface equation (0.51 mg/g). Thus, it was interesting to find that ACpa other than PHel showed the maximum As$^{(V)}$ adsorption capacity among all of the examined soils. Furthermore, the M_1 and K_1 values for the lower-energy surface were 0.55 mg/g and 0.02 L/mg, respectively, while the M_2 and K_2 values for the higher-energy surface were 0.17 mg/g and 0.90 L/mg. The As$^{(V)}$ adsorption on ACpa occurred predominantly on the low-energy surface.

Soil properties strongly affect As adsorption and subsequently influence the environmental fate of As (Jiang et al. 2005b, Buschmann et al. 2006, Girouard and Zagury 2009). Jiang et al. (2005a) suggested that the Langmuir two-surface equation describes As$^{(V)}$ adsorption better than the Langmuir one-surface equation for soils with lower contents of OM, DOC, or extractable P, where both high-energy and low-energy surface adsorption sites would be available. However, a similar pattern was not observed in this case. The highest Fe_{CD} and Al_{CD} contents (32.63 and

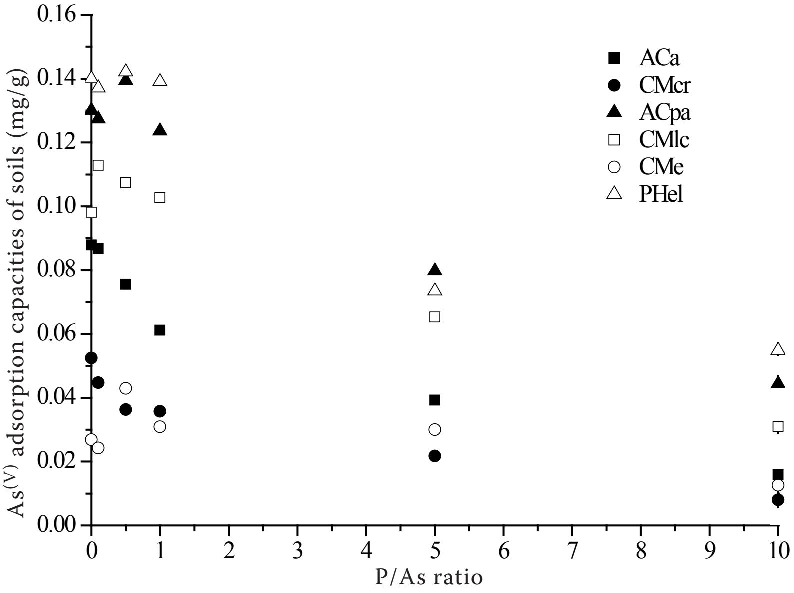

Figure 3. Effect of adding phosphate on As$^{(V)}$ adsorption in six soils with different properties. PHel – Luvi-Endogleyic Phaeozem; CMe – Epigleyic Cambisol; CMcr – Rhodi-Calcaric Cambisol; CMlc – Calcari-Leptic Cambisol; ACa – Alumic Acrisol; ACpa – Alumi-Plinthic Acrisol

Table 4. The decreases in the $As^{(V)}$ adsorption capacity in six soils with different properties when the P/As molar ratio in the solution was increased from 0 to 10

Items	PHel	CMe	CMcr	ACpa	ACa	CMlc
Decrease in the $As^{(V)}$ adsorption capacity (mg/g)	0.085	0.014	0.044	0.086	0.072	0.067
Percent of the initial adsorption capacity (%)	60.8	53.3	84.8	65.9	81.9	68.5
A/P value	1.135	0.191	0.593	1.143	0.959	0.896

Following an increase of the P/As molar ratio from 0 to 10, the decrease in the $As^{(V)}$ adsorption capacity in each soil was determined using the subtraction method; the molar content of P was 1 mmol/L when the P/As ratio reached 10; A/P value-decrease in adsorbed As/added P; PHel – Luvi-Endogleyic Phaeozem; CMe – Epigleyic Cambisol; CMcr – Rhodi-Calcaric Cambisol; CMlc – Calcari-Leptic Cambisol; ACa – Alumic Acrisol; ACpa – Alumi-Plinthic Acrisol

1.06 g/kg) and the lowest pH (4.60) were found in ACpa (Table 1). A large quantity of Fe_{CD} in soils contributes to $As^{(V)}$ adsorption not only on the low-energy surface but also on the high-energy surface (Jiang et al. 2005b). Pigna et al. (2006) suggested that $As^{(V)}$ exhibits a relatively strong affinity for the surfaces of iron and aluminium (hydr) oxides, possibly forming inner-sphere complexes. A lower pH might favour the adsorption of $As^{(V)}$ on soil minerals via ligand exchange reactions or attractive electrostatic interactions (Dixit and Hering 2003). It may therefore be inferred that the sufficient adsorption sites available in ACpa were conducive to the good fitting of the Langmuir two-surface equation.

Effect of adding phosphate on $As^{(V)}$ adsorption on soils. The addition of phosphate strongly influenced $As^{(V)}$ adsorption by the six soils. With an increasing P/As ratio, the $As^{(V)}$ adsorption capacities of the six soils all showed a decreasing trend (Figure 3). This result was consistent with those of Zhang and Selim (2008), who found that the amount of $As^{(V)}$ adsorption decreases significantly when the P concentrations in soil solution increase. However, Sisr et al. (2007) found that the mobility of As was not significantly affected by the presence of P ions in a column experiment. The different experimental methods, soil properties, and added P concentration used in these analyses might be responsible for these differences.

The observed decreases in the $As^{(V)}$ adsorption capacities varied according to different soils (Table 4), when the P/As ratio in the solution was increased from 0 to 10. Among all of the tested soils, the decrease in the $As^{(V)}$ adsorption capacity of ACpa was maximal, at 0.086 mg/g. The minimum decrease of 0.014 mg/g was detected in CMe. Pearson correlation analysis indicated that the decreases in the $As^{(V)}$ adsorption capacities were significantly correlated with the initial $As^{(V)}$ adsorption capacities of all of the soils ($R^2 = 0.92$, $P < 0.01$). This result supported the conclusion that P can compete with $As^{(V)}$ for surface sites in soils and consequently suppress the adsorption of $As^{(V)}$ (Peryea and Kammereck 1997, Liu et al. 2001). It also indicated that addition of P can influence $As^{(V)}$ adsorption in different soils to different extents.

The A/P value (decrease in absorbed As/added P) was used to quantitatively depict the effect of the addition of P on $As^{(V)}$ adsorption by the soils. Among the tested soils, the A/P value of 1.143 for ACpa was the highest, followed by the value of 1.135 obtained for PHel. The lowest A/P value of 0.191 was observed for CMe. It appears to be difficult for P to occupy the $As^{(V)}$ absorption sites in CMe, whereas P competes more effectively for $As^{(V)}$ adsorption sites in ACpa and PHel. Therefore, P application could substantially decrease $As^{(V)}$ adsorption in ACpa and PHel and correspondingly increase the mobility and availability of As. Thus, in some As-contaminated agricultural soils, rationally controlling the application of P might be a feasible means of reducing the environmental risk of As, especially for PHel, which has a relatively higher available P content of 42.59 mg/kg compared with that of the other tested soils.

In conclusion, the examined soils with different properties presented variable $As^{(V)}$ adsorption capacities. Addition of P decreases $As^{(V)}$ adsorption in different soils to different extents and correspondingly makes more As available. In some As-contaminated agricultural soils that are relatively rich in P, reducing P application will not only meet the demands of crop growth but will also be conducive to decreasing the As risk in soils. In contrast, in some As-contaminated agricultural soils that are relatively poor in P, rationally varying the fertilisation method and type of P fertiliser might be more feasible.

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