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Adsorption and desorption characteristics of diphenylarsenicals in two contrasting soils

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Abstract

Diphenylarsinic acid (DPAA) is formed during the leakage of aromatic arsenic chemical weapons in soils, is persistent in nature, and results in arsenic contamination in the field. The adsorption and desorption characteristics of DPAA were investigated in two typical Chinese soils, an Acrisol (a variable-charge soil) and a Phaeozem (a constant-charge soil), their thermodynamics and some of the factors influencing them (i.e., initial pH value, ionic strength and phosphate) using the batch method in order to understand the environmental fate of DPAA in soils. The results indicate that the Acrisol had a stronger adsorption capacity for DPAA than the Phaeozem. Soil DPAA adsorption was a spontaneous and endothermic process and the amount of DPAA adsorbed was affected significantly by variation in soil pH and phosphate. In contrast, soil organic matter (OM) and ionic strength had no significant effect on adsorption. This suggests that DPAA adsorption may be due to specific adsorption on soil mineral surfaces. Therefore, monitoring the fate of DPAA in soils is recommended in areas contaminated by leakage from chemical weapons.

Key words: diphenylarsinic acid; adsorption and desorption; chemical weapons; residual soil contamination.

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Introduction

Aromatic arsenicals (AAs) were once used for the manufacture of chemical weapons such as Clark I (diphenylchloroarsine) and Clark II (diphenylcyanoarsine) which were employed as vomiting and sneezing agents during both World Wars (Arao et al., 2009). After the Second World War chemical weapons were buried and dumped in several parts of China (Wada et al., 2006), Japan (Hanaoka et al., 2005; Ishizaki et al., 2005), Germany (Hempel et al., 2009), and Belgium (Bausinger and Preuss, 2005). The AAs can be modified by hydrolysis and oxidation, leading to the formation of diphenylarsinic acid (DPAA) which is persistent and results in arsenic contamination of soils (Nakamiya et al., 2007; Harada et al., 2010). In 2002 there was an arsenic poisoning incident in Kamisu, Japan when local people drank DPAA-contaminated well water (Ishizaki et al., 2005). Several inhabitants exhibited unusual clinical symptoms of the central nervous system (Ishii et al., 2004). DPAA was then detected as the predominant species of arsenic in the groundwater in which the maximum concentration was found to be 15 mg/L (Arao et al., 2009). The DPAA may have originated from illegal dumping of DPAA itself (Kinoshita et al., 2008).

Studies on the behavior of DPAA in soils are urgently required in order to fully understand its potential to exert adverse environmental effects. Arao et al. (2009) showed that DPAA can be taken up by rice and was readily transferred from the straw to the grains in a pot experiment in which rice was grown soil amended with different forms of aromatic arsenicals. These results indicate that DPAA has some ability to transfer in the soil and can be readily absorbed by crops. Maejima et al. (2011) investigated the mobility of DPAA and its derivatives in two typical Japanese soils (a Fluvisol and an Andosol) and found that the mechanism of adsorption of DPAA was mainly due to ligand exchange reactions between variable-charge minerals and the arsenate group. AAs also appeared to have the potential to contaminate the groundwater by leaching.

Soils are complex systems and the fate of pollutants can exhibit different behaviors in different typical soils. There were numerous battlefields in China during the Second World War and chemical weapons were used extensively. Many locations such as Haerbaling area in

Dunhua city and Huang Huzi Mountain in Nanjing city have become contaminated through the leakage of buried chemical weapons, most of which contain large amounts of arsenicals (Tu, 2011). However, little is known about the environmental fate of DPAA in Chinese soils (Deng and Evans, 1997). Therefore, in the present study two typical Chinese soils, a Phaeozem and an Acrisol, were selected because they are widely subject to AA leakage and they have contrasting physicochemical properties. Phaeozems are high in organic matter and are considered to be constant-charge soils. Acrisols, in contrast, contain little organic matter but have large amounts of minerals containing iron and aluminum oxides and are often referred to as variable-charge soils (Yu, 1997). Thus, our main objective was to investigate the adsorption and desorption characteristics onto these two typical Chinese soil types which often occur in areas contaminated with AAs. The effect of the soil components, the initial soil pH, ionic strength and phosphate concentration on the adsorption of DPAA onto the soils were investigated to better understand the sorption mechanism and the pathways of AA movement in soils. It was hoped that the results would help to assess the potential fate of DPAA in contaminated soils.

1 Materials and methods

1.1 Chemicals

The diphenylarsenicals ($C_{12}H_{11}AsO_4$, 294.13) standard (purity 97%) were purchased from Wako Company, Osaka, Japan. The logarithm *n*-octanol/water partition coefficient (Log K_{ow}) of DPAA is 2.80, and the pKa is 5.0 (Nakamiya et al., 2007). The acetonitrile used in quantitative analysis was of high performance liquid chromatography (HPLC) grade. Other chemicals were of analytical grade and all solutions were prepared using 18.2 M Ω Milli-Q water (Millipore Corporation, Billerica, MA).

1.2 Soil samples

Two soil types, a Phaeozem and an Acrisol, were collected from the plough layer (top 15 cm) of fields in Hailun county (Heilongjiang province) and Yujiang County (Jiangxi province), respectively. The soil samples were air-dried and passed through a 0.25 mm sieve prior to use.

Selected physico-chemical properties of the two soils are presented in **Table 1**.

Table 1 Selected properties of the soils tested

Soil	pH ^a	OM ^b	CEC ^c	Free Fe ₂ O ₃ ^d	Total P ^e	Particle size distribution (%) ^f		
		(g/kg)	(cmol/kg)	(g/kg)	(g/kg)	Clay	Silt	Sand
Acrisol	4.6	7.5	11.8	60.9	1.19	55.4	41.4	3.2
Phaeozem	6.2	70.3	40.1	14.3	1.08	17.7	45.3	36.9

^a Ratio of soil:water was 1:10 (V/V); ^b dichromate method (Lu, 2000); ^c CEC (cation exchange capacity), ammonium acetate method (Lu, 2000); ^d DCB method (Lu, 2000); ^e aqua regia digestion; ^f the soil particle component was analyzed by Laser particle size analyzer (LS230, Beckman Coulter, USA).

1.3 Adsorption/desorption experiments

The adsorption experiments were carried out according to the batch equilibrium method. A series of DPAA solution (including 2, 4, 12, 16, 20 mg/L) were prepared in 0.01 mol/L NaNO₃ as the background solution containing 100 mg L⁻¹ NaN₃ as an anti-bacterial agent. The 2.000 g of soil was placed in a glass centrifuge tube (80 mL) with a Teflon-lined screw cap, and the whole tube was weighed as M₁. After adding 20 mL DPAA solution at different concentrations, the pH of the mixture was adjusted according to the soil sample pH using NaHCO₃ solution. Previous work had shown that equilibrium can be reached in 72 hr (data not published), The tube was shaken at 200 r/min for 72 hr at (25 ± 1)°C in the dark. After centrifugation at 1500 r/min for 15 min, the supernatant was filtered through a 0.22 μm membrane filter.

The desorption experiments were conducted immediately following the adsorption experiments. The whole tube was weighed as M₂ to calculate the solution remaining in the soil after adsorption, then 20 mL of 0.01 mol L⁻¹ NaNO₃ was added as the desorption solution and the tube was shaken again at 200 r/min for 72 hr at (25 ± 1)°C in the dark. At the end point the samples were collected using the same procedure described above. All experiments were conducted in triplicate.

The amount of DPAA adsorbed on the soil sample (Q_{ads} , mg/kg), was calculated from the

difference between the initial DPAA concentration (C_i , mg/L) and the equilibrium concentration (C_{eq} , mg/L) in the supernatant as follows:

$$Q_{ads} = \frac{(C_i - C_{eq}) \times V}{M} \quad (1)$$

where, V is the volume (20 mL) and M (g, dry weight basis) is the mass of the soil sample.

The amount of DPAA desorbed from the soil sample (Q_{des} , mg/kg), was calculated according to Eq. (2):

$$Q_{des} = \frac{(V_d C_d - V_r C_{eq})}{M} \quad (2)$$

Where V_d is the volume (20 mL) and C_d is the equilibrium concentration of desorption in the supernatant. V_r is the volume of adsorption equilibrium solution remaining in the soil and is equal to $M_2 - M_1$. C_{eq} is the equilibrium concentration of adsorption. M is the mass of the soil sample (g, dry weight basis).

1.4 Effect of temperature on the adsorption of DPAA in soils

A series of DPAA solutions (2, 4, 8, 12, 16, and 20 mg/L) were prepared as described above. The 2.000 g of soil was weighed in a glass centrifuge tube, 20 ml of DPAA solution of different concentrations was added, the pH was adjusted as required, and the tube was placed on a shaker in the dark at 200 r/min for 72 hr at $(25 \pm 1)^\circ\text{C}$ and $(45 \pm 1)^\circ\text{C}$, respectively. Then the supernatants were collected as described above.

1.5 Effects of initial pH, ionic strength and phosphate on adsorption of DPAA in soils

The effects of initial pH, ionic strength and phosphate on the adsorption of DPAA in the soils were examined by varying the pH, the concentration of the background solution and the content of phosphate in the DPAA solution, respectively. Experiment 1 examined the effect of the pH value. The 2.000 g of soil was weighed in a glass centrifuge tube and 20 mL of DPAA (20 mg/L) solution was added. The pH of the soil suspension was adjusted with HCl or NaOH and the pH treatments were pH 3.0, 4.0 5.0, 6.0, 7.0 and 8.0. Then the tube was placed in a shaker in the dark at 200 r/min for 72 hr at $(25 \pm 1)^\circ\text{C}$. Experiment 2 investigated the effect of

ionic strength. The 20 mg L⁻¹ DPAA solutions were prepared in 0.01 and 0.1 mol/L NaNO₃ as the background solution. Then the adsorption tests were followed as described above. Experiment 3 studied the effect of phosphate. NaH₂PO₄ was added to the 20 mg/L DPAA solution to give a P concentration of 1.0 mg/L in the solution (approximate to the total content of P in the two soils). A zero-P NaH₂PO₄ control was also set up. Then 2.000 g of soil was added to the solution and the adsorption test was carried out by the procedure described above.

1.6 Analytical procedure

In other studies measurements based on GC-MS, LC-ICP-MS or LC-LC-MS have been used to determine the concentration of aromatic arsenicals (Haas and Krippendorf, 1997; Wada et al., 2006). However, a method for determining DPAA concentrations in filtered solutions has been developed using HPLC (Shimadzu, Kyoto, Japan). The column used was a Shimadzu VP-ODS reversed phase C18 column (150 mm × 4.6 mm, 5 μm). The sample injection volume was 10 μL and the mobile phase was 19% acetonitrile and 81% 0.02 mol/L KH₂PO₄ solution. The flow rate was 1 mL/min and the detector wavelength was 220 nm using SPD-m20A (Shimadzu, Kyoto, Japan).

2 Results

2.1 Adsorption isotherms of DPAA in the two soils

The adsorption isotherms of DPAA onto the surfaces of the Acrisol and Phaeozem are shown in **Fig. 1**. The amount of DPAA adsorbed on both soils increased with increasing concentration of DPAA in the starting solution and the data obtained from the batch adsorption experiments were well described by both the Freundlich model and the Henry model, all the parameters of which are presented in **Table 2**.

Table 2 Adsorption coefficients of DPAA in two different soils

	Henry Model	Freundlich Model
Soil	$Q_{\text{ads}} = K_D C_{\text{eq}}$	$Q_{\text{ads}} = K_f C_{\text{eq}}^n$

	K_D ((mg/kg)/(mg/L))	R^2	K_f ((mg/kg)/(mg/L) ⁿ)	n	R^2
Acrisol	20.10(±1.07)	0.992	31.69(±0.84)	0.79(±0.04)	0.999
Phaeozem	1.50(±0.08)	0.990	2.29(±0.38)	0.87(±0.06)	0.999

The Freundlich model can be expressed as:

$$Q_{\text{ads}} = K_f C_{\text{eq}}^n \quad (3)$$

where K_f , the adsorption coefficient, and n , the nonlinearity factor, are the constants that give estimates of the adsorption capacity and intensity, respectively. As shown in **Fig. 1**, more DPAA was adsorbed on the Acrisol than the Phaeozem. In this study the data were also regressed to the linear Henry model to estimate the soil/water distribution coefficient (K_D) and the organic carbon adsorption coefficient K_{oc} , which represent the extent of partition of organic compounds to soils. K_{oc} can be expressed as $K_{oc} = K_D / f_{oc}$, where f_{oc} is the soil organic carbon content. Normalization of the K_D values results in great variation in K_{oc} values among soils. The K_{oc} value in the Acrisol is 6.01 L/g and 0.04 L/g in the Phaeozem. Because the K_{oc} can reflect the mobility of organic chemicals in soil, it can be concluded that DPAA was less mobile in the Acrisol than in the Phaeozem.

Fig. 1 Adsorption isotherms of DPAA in the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

2.2 Desorption characteristics of DPAA from the soils

Figure 2 shows the relationships between the desorption capacity and the adsorption capacity of DPAA in the soils. The amount of DPAA desorbed from the soils increased with increasing amount of DPAA adsorbed, and the data were well regressed by linearity. The slope reflects the desorption yield in soil and a higher slope value means a lower holding capacity. According to the parameters, the slope of the Acrisol (0.23) is less than that of the

Phaeozem (0.66), suggesting that the Acrisol can hold more DPAA than the Phaeozem.



Fig. 2 Relationship between the adsorption and desorption of DPAA in the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

2.3 Thermodynamics of DPAA in soils

The effect of temperature on the adsorption of DPAA onto the soils is shown in **Fig. 3**. The data obtained were also fitted by the Freundlich and Henry models (**Table 3**). At low equilibrium concentration no effect of temperature on the adsorption capacity of DPAA in the two soils was evident. The adsorption capacity was only slightly enhanced as the equilibrium concentration increased. In the Acrisol when the temperature reached 45°C the K_f value in the Freundlich model increased from 31.69 to 35.21. The same phenomenon was also found in the Phaeozem, with the K_f value increasing from 2.29 to 3.19. The insignificant temperature effect indicates that the adsorption of DPAA in soils is a spontaneous process with a low activation energy (Tiwari et al., 1999). The nonlinearity parameter n also showed no significant variation and changed only from 0.7956 to 0.7907. It may be that n , reflecting the degree of abnormality of soil particles, is independent of the temperature.

In order to fully understand the mechanism of DPAA adsorption on the soils, some thermodynamic parameters, standard Gibbs free energy ΔG^0 , enthalpy variation ΔH^0 , entropy variation ΔS^0 , presented in **Table 4**, were calculated by the following equations (Krishna et al., 2000): $\Delta G^0 = -RT \ln K_c$ and $\Delta G^0 = \Delta H^0 - \Delta S^0 T$, where K_c , the equilibrium constant, is equal to the K_D in the Henry model.

Table 3 Adsorption coefficients of DPAA in two different soils at 318 K

Freundlich Model	
Soil	$Q_{\text{ads}} = K_f C_{\text{eq}}^n$

	K_f ((mg/kg)/(mg/L) ⁿ)	n	R^2
Acrisol	31.69(±2.46)	0.79(±0.04)	0.995
Phaeozem	2.29(±0.76)	0.87(±0.09)	0.979

Table 4 Isotherm parameters and thermodynamic parameters of DPAA in different temperature

Soil	Temperature (K)	K_c	$-\Delta G^0$ (kJ/mol)	ΔH^0 (kJ/mol)	ΔS^0 (J·K/mol)
Acrisol	298	20.10	7.44	5.08	42
	318	22.80	8.27		
Phaeozem	298	1.50	0.99	8.84	33
	318	1.87	1.65		

The negative values of ΔG^0 at all temperatures in the two soils indicate the feasibility of the process and the spontaneous nature of the DPAA adsorption onto soils. A decrease in the value of ΔG^0 with increasing temperature suggests more adsorption of DPAA at higher temperatures. However, this temperature effect was not obvious as stated above. The positive values of ΔH^0 , 5.08 kJ/mol for the Acrisol and 8.84 kJ/mol for the Phaeozem, imply that the adsorption of DPAA is also an endothermic process, and because of the weaker attractive forces the physisorption mechanism may play a role in this adsorption system (Tiwari et al., 1999). The positive values of ΔS^0 in the two soils reveal that the degrees of freedom increased at the solid-liquid interface during the sorption of DPAA onto the soils.



Fig. 3 Effect of temperature on DPAA adsorption on the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

2.4 Effect of pH value on DPAA adsorption

The adsorption of DPAA in both soils decreased with increasing pH in 0.01 mol/L NaNO₃ background solution (**Fig. 4**). In the Acrisol the pH value had no effect on DPAA adsorption until the pH value increased from 3.0 to 6.0 and then adsorption decreased markedly in high pH conditions. When the pH value was 8.0, the amount of DPAA adsorption reached 54.8 mg/L, accounting for only 40% of the normal adsorption capacity. In the Phaeozem the adsorption capacity for DPAA was also enhanced substantially in low pH conditions. When the pH value was 3.0 the amount of DPAA adsorbed increased to 125.3 mg/L, which was almost as high as the adsorption capacity of DPAA in the Acrisol. As the pH value increased, the adsorption capacity decreased. When the pH value changed from 5.0 to 7.0, DPAA adsorption decreased very slightly. At the end the amount adsorbed was only 12.0 mg/L.



Fig. 4 Effect of pH on DPAA adsorption on the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

2.5 Effect of competing ions (phosphate) and ionic strength on DPAA adsorption

It is known that phosphate adsorption on soil surfaces is similar to that of arsenate, and it is thus a strong competitor to arsenate for adsorption sites (Manning and Goldberg, 1996; Jackson and Miller, 2000; Seaman et al., 2001). Here, the effect of phosphate was also examined and is shown in **Fig. 5a**. When the phosphorus concentration in the background solution was 1 mg/L and approximately equal to the total P in two soils (**Table 1**), DPAA adsorption in the Acrisol was inhibited significantly, accounting for only 30% of the amount adsorbed on the control. However, in the Phaeozem the adsorption capacity did not seem to be affected by adding phosphorus.

The effect of ionic strength on DPAA adsorption is shown in **Fig. 5b**. When the ionic strength of the solution increased to 0.1 mg/L NaNO₃, adsorption of DPAA was unaffected in both soils.

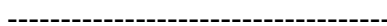


Fig. 5 Effect of ionic strength and phosphorus on DPAA adsorption. Error bars represent the standard deviation of the mean of 3 replicate samples.

3 Discussion

3.1 The mechanism of DPAA adsorption onto the soil

The significant differences in adsorption and desorption capacity for DPAA between the Acrisol and Phaeozem may be due to several factors, including the difference in soil particle size distribution, soil organic matter (OM) content, content of iron/aluminium oxides in clays, pH value, and concentration of competing ions, all of which may influence the adsorption mechanism of DPAA. Ljung et al. (2006) reported that the amount of As absorbed onto the soil surfaces was positively correlated with the percentage of clays. The clay distribution had no significant difference between the Acrisol and Phaeozem (**Table 1**), indicating that particle size is not a major factor affecting the adsorption and desorption processes and the functional groups may play a more important role.

In general, the adsorption mechanisms of organic compounds depend largely on their hydrophobic interactions with soil organic matter (Rutherford et al., 1992; Spark and Swift, 2002; Nguyen et al., 2005), especially in the case of non-ionic organic compounds (Chiou et al., 1984; Ying and Kookana, 2005; Xu et al., 2009). However, adsorption of ionic organic compounds is influenced to a greater extent by ligand exchange. Several lines of evidence in the present study suggest that the adsorption mechanism of DPAA onto soil was controlled by the arsenate functional group rather than phenyl (Mader et al., 1997). When the two soil types are compared the Acrisol has a larger content of free iron oxides but the Phaeozem has a much higher soil organic matter content. As described above, the K_{oc} value of the Acrisol was 6.01 L/g, significantly higher than that of the Phaeozem at 0.04 L/g. During the desorption process the Phaeozem was unable to retain DPAA. Furthermore, higher temperatures did not lead to any significant increase in the DPAA adsorption capacity of either soil. All these results indicate that high soil OM content made little contribution to the adsorption of DPAA,

and the amount of DPAA sorbed on soils appears to be correlated to the content of free iron oxides. This finding is similar to the results of Maejima et al. (2011) who reported that some AAs (including DPAA) were sorbed strongly on high variable charge soil, although the soil also had a high soil OM content. When soil OM was removed, thereby exposing the iron/aluminium oxides, the DPAA adsorption capacity was enhanced and it was concluded that the adsorption of DPAA may be due to ligand exchange.

As discussed above, arsenate (V) forms inner-sphere complexes with Fe/Al oxides. This is a strong form of bonding and belongs to the specific adsorption category (Goldberg, 1986; Waltham and Eick, 2002; Luo et al., 2006; Catalano et al., 2007). This charge-dependent adsorption is greatly affected by changes in soil pH (Grossl et al., 1997; Xu et al., 2009). As noted above, the uptake of DPAA decreased with the increasing pH value of both soils. It may be proposed that at lower pH the arsenate groups on the DPAA were in both uncharged species and anionic species (according to the pK_a of DPAA) and also the soil surface released hydroxyl groups and became positively charged, which would facilitate the adsorption of DPAA onto the soil surfaces through both the surface complexation and the van der Waals attraction (Lee and Tiwari, 2012; Tiwari and Lee, 2012). However, at higher pH the surface became negatively charged as well the DPAA being negatively charged and they were repelled by each other so that the sorption of DPAA was hampered. Results also show that phosphate, a known competitor with arsenate in adsorption onto soil particles (Pigna et al., 2006), showed strong inhibition to DPAA adsorption in the Acrisol, suggesting that adsorption of DPAA may be a form of specific adsorption. However, phosphate had little effect on DPAA adsorption in the Phaeozem. It is possible that the high soil OM content of the Phaeozem inhibited the specific adsorption sites and thus adsorption was unaffected by phosphate. The effect of ionic strength in the solution on changing anion partitioning can distinguish between specific and non-specific adsorption (Hayes et al., 1988; Antelo et al., 2005; Xu et al., 2009). Specific adsorption is unaffected by change in ionic strength but non-specific adsorption is likely to be influenced greatly by change in ionic strength because of the competitive adsorption with counter ions. The adsorption capacity of DPAA was not affected by increasing ionic strength in these two soils. Thus, the DPAA adsorption process

was mainly governed by specific adsorption and because of the phenyl groups the bonding characteristics of the arsenate groups with mineral clays was altered drastically, similar to the changes that have been found to occur in other organic arsenicals (Jing et al., 2005; Lafferty and Loeppert, 2005). In addition, because of the low Gibbs free energy in the adsorption process, DPAA adsorption is a physisorption process.

3.2 Mobility of DPAA in soils

Understanding the sorption behavior of DPAA on soils is important to access its mobility in soil systems and potential impacts on the environment. Roxarsone, which also contains a phenyl group and an arsenate group, was once considered to be resistant to degradation and was used in some countries as an additive in poultry feed for prevention of coccidiosis and for growth stimulation (Anderson, 1983). However, recent studies have reported that roxarsone is a major source of total As in soils, sediments and groundwater due to its high mobility which increases further when roxarsone is biologically transformed into inorganic arsenate or arsenite (Arai et al., 2003; Brown et al., 2005). DPAA has also been found to be degraded in soil, although the process is slow (Maejima et al., 2011). In the present study in typical Chinese soils the holding capacity of DPAA was weaker, even in the Acrisol with its high content of iron/aluminum oxides, than inorganic arsenic and other methyl arsenics (Huang et al., 2011), according to the free energy obtained (**Table 4**). Moreover, the soil OM content also reduced the immobilizing capacity. This suggests that in northeast China where most of the chemical weapon leakage accidents have occurred and most of the soils are of the Phaeozem type it is necessary to monitor soil, water and crop DPAA levels in areas contaminated with chemical weapons. In addition, in other areas, arable soils should also be monitored because the application of phosphate to DPAA-contaminated soils for crop production may enhance the mobility of DPAA and result in highly adverse environmental risk to the human food chain.

4 Conclusions

The adsorption and desorption characteristics of DPAA were demonstrated in two typical

but contrasting Chinese soils. DPAA adsorption was found to be strongly associated with soil iron/aluminum oxide content, and can be affected by soil pH and phosphate status. The adsorption mechanism was mainly due to specific adsorption. Because of its poor adsorption capacity in soils due to soil OM and phosphate, DPAA represents a threat to the terrestrial environment and its fate in soils contaminated by chemical weapon leakage fields requires further study and monitoring to minimize harmful effects on the environment and the human population.

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List of Figure Captions

Fig. 1 Adsorption isotherms of DPAA in the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

Fig. 2 Relationship between the adsorption and desorption of DPAA in the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

Fig. 3 Effect of temperature on DPAA adsorption on the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

Fig. 4 Effect of pH on DPAA adsorption on the two soil types. Error bars represent the standard deviation of the mean of 3 replicate samples.

Fig. 5 Effect of ionic strength and phosphorus on DPAA adsorption. Error bars represent the standard deviation of the mean of 3 replicate samples.

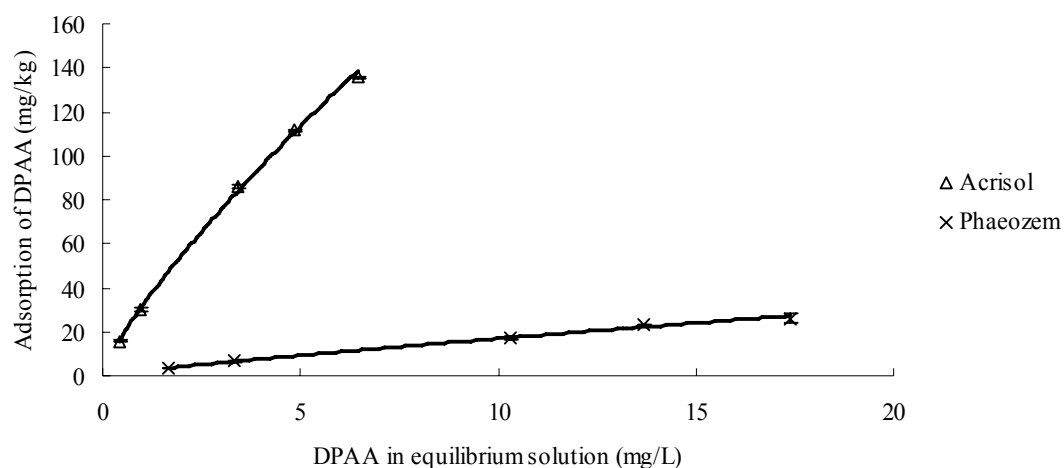


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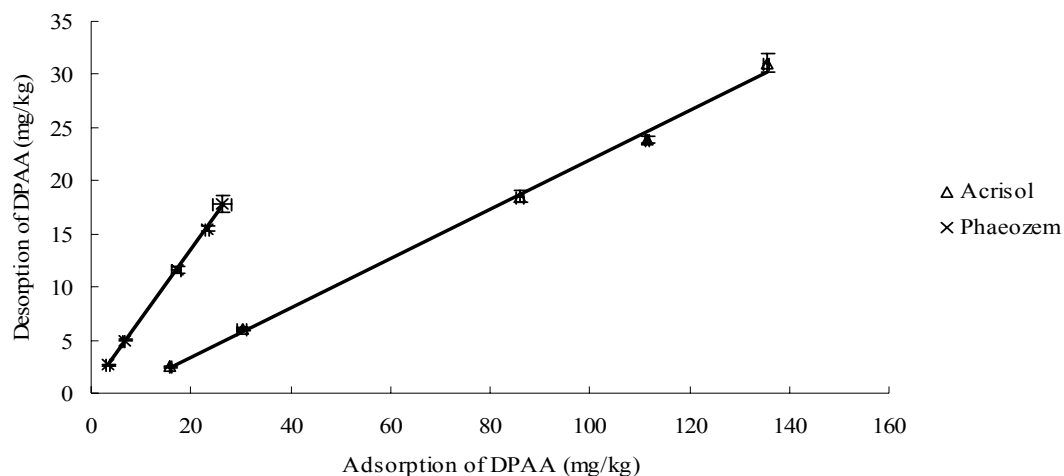


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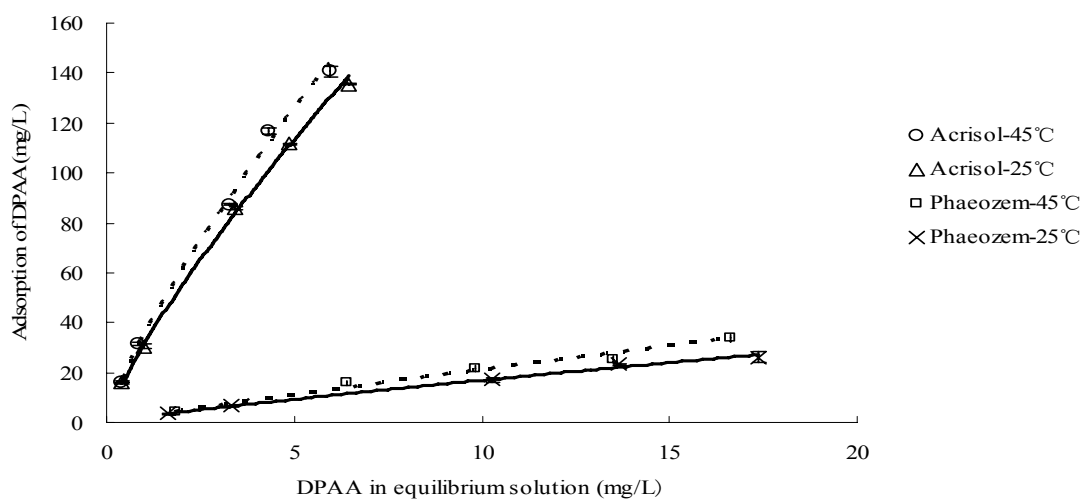


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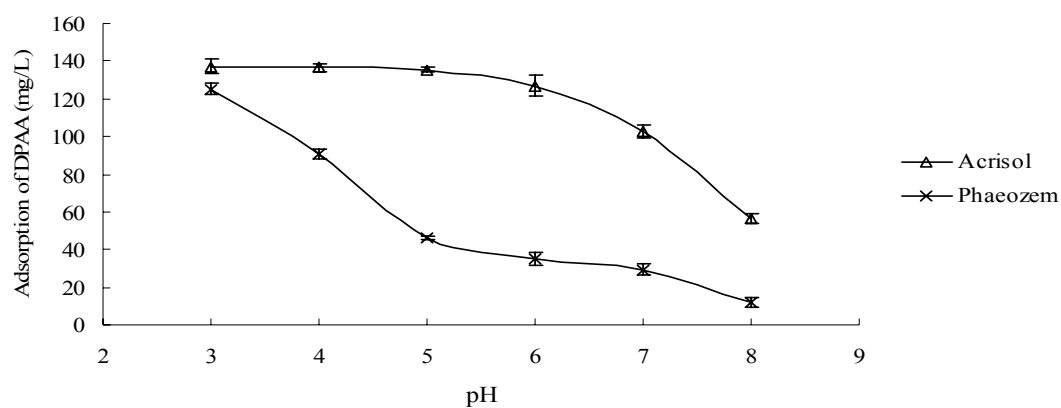


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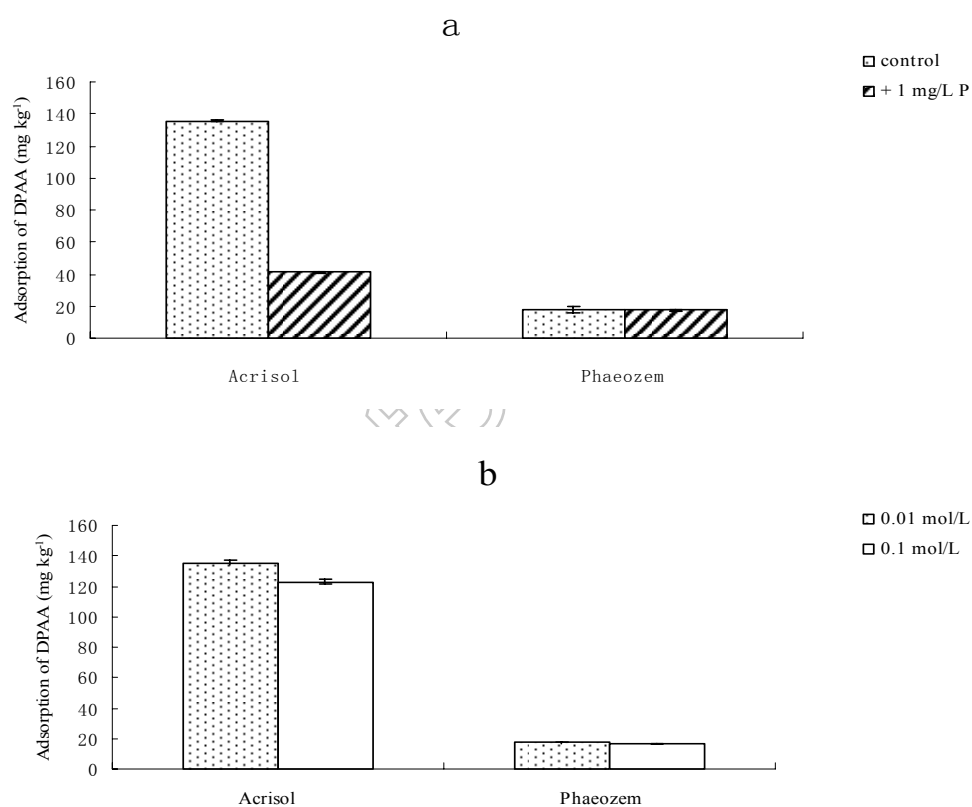


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