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Enhanced chlorophenol sorption of soils by rice-straw-ash amendment

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ABSTRACT

Rice-straw burning is a common post-harvest practice on rice paddy land, which results in the accumulation of rice-straw ash (RSA) in paddy soil. Because the occurrence of RSA in soil may affect the fate and transport of contaminants, this study investigated the sorption of 3-chlorophenol (3-CP) on RSA and RSA amended soils to evaluate the sorptive properties of RSA in soils. The results showed that the sorption of 3-CP to RSA proceeds through a surface reaction rather than through partitioning and that the neutral form of 3-CP is preferentially sorbed to the surface when compared to the deprotonated anionic form of 3-CP. The addition of RSA to the soils enhanced the overall 3-CP sorption, indicating that RSA amendment may be applied to retard the movement of 3-CP in contaminated soils. As the RSA content in the soils was increased from 0% to 2%, the Langmuir sorption maximum of the soils increased from 18–80 to 256–274 mg kg^{-1}. Thus, RSA contributed more to the total sorption of the soils than other major components in the soils. Nonetheless, the 3-CP sorption of the soils containing RSA was less than the combination of pure RSA and the soils, thereby indicating that the 3-CP sorption of RSA was suppressed. This may be attributed to the competition of organic matter or other soil components for the surface binding sites of RSA.

1. Introduction

The sorption of contaminants is one of the most important processes in determining the fate of contaminants in soils. Extensive studies have focused on the sorptive properties of major constituents of soils, including both minerals and organic matter, for contaminants to elucidate their roles in determining the mobility and bioavailability of contaminants in soils. Recent studies have suggested that the ash arising from burning crop residues may also be a potential sorbent for organic [1–6] and inorganic contaminants [7,8] in agricultural soils. The high sorptivity of crop residue ashes has been attributed to the content of black carbon, which is formed from the incomplete combustion of crop residues. Therefore, without considering the contribution of black carbon to the total sorption capacity of agricultural soils for contaminants, the contributions of soil minerals and organic matter may be significantly overestimated.

Rice production plays a prominent role in world food supply. After the grain is harvested, the rice-straw by-product is left in the field and burned since this land cleaning practice is inexpensive with respect to alternative forms of waste management [9–11]. The burned rice straw is often incorporated into soils and provides several agronomic benefits such as reducing weed problems and rice diseases, and increasing soil fertility [9,12–14]. Incorporation of rice-straw ash (RSA) into soils also results in the accumulation of RSA, which is expected to play a significant role in the sorption of contaminants. For example, previous studies have demonstrated that RSA and other burned crop residues are effective sorbents for herbicides, such as 2-methyl-4-chlorophenoxyacetic acid [3], atrazine [4], and diuron [5]. Thus, the contribution of RSA to the total sorption of a contaminant needs to be considered when modeling the fate and transport of the contaminant in paddy lands.

In the present study, the effects of amending RSA to soils on the sorption of 3-chlorophenol (3-CP) were investigated. Chlorophenols (CPs) are a group of chlorinated phenolic compounds that have been widely used as biocides in agriculture, industry and public health [15–17]. Their broad application has resulted in widespread distribution of these highly toxic substances in the natural environment. Previous studies showed that the transport of CPs in soils is predominantly determined by their sorption and desorption on the organic matter in soil [18–23]. However, in rice paddy soil on which rice-straw burning is a common post-harvest practice, the contribution of soil organic matter to the CP sorption can be overestimated if that of RSA is not included. It is therefore necessary to understand the effects of RSA amendment on the sorption of CPs in soils in order to understand the fate and transport of CPs in soils containing RSA. Thus, this work aimed to investigate the potential

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role of RSA in the immobilization of the model CP, 3-CP, in soils. Particularly, the 3-CP sorption of the RSA amended soils were compared with the sum of pure RSA and the soils to show the effects of other soil components on the 3-CP sorption of RSA in soils. 3-CP was selected as the model sorbate because its pK_a (i.e., 9.0 [24]) is higher than the common soil pH range (about pHs 4–8), meaning that the dominant form of 3-CP will be the protonated neutral form. Since the chemical structures of CPs contain one OH group and one phenyl ring, their structures are the suitable models for studying sorption/desorption processes of small polar organic contaminants in soils containing burned crop residues. The results of this work may be extended to explain the effects of RSA or other burned crop residues on the mobility of other small polar organic contaminants in agricultural soils.

2. Materials and methods

2.1. Preparation of 3-CP solution

A stock solution of 1 g L^{-1} 3-CP was prepared by dissolving 0.1 g of 3-CP in 100 mL 0.01 M $CaCl_2$ solution. Several drops of 200 mg L^{-1} $HgCl_2$ solution were added into the 3-CP solution to inhibit microbial growth. This stock solution was used to prepare all the 3-CP solutions for the subsequent sorption experiments.

2.2. Rice-straw ash and soil samples

Rice straw was collected in the summer of 2007 from the experimental field in the Agricultural Research Institute at Taichung County, Taiwan. To simulate the field burning, rice straw was burned at 300 °C with an air supply for 10 min. The resulting ash (i.e., RSA) was collected, ground, passed through a 20-mesh sieve and stored in a plastic bottle. The RSA had a specific surface area of 5 m^2 g^{-1} and a point of zero charge (PZC) at pH 5.5, measured using Micrometritics ASAP 2000 surface area analyzer and the pH drift method [25], respectively.

Three soil samples were collected from Tartu mesa near Taichung city (soil 1), Agricultural Research Institute (soil 2) and the Agricultural Station of National Chung Hsing University (soil 3) in Taiwan. These soils were selected because of no recent records of crop or vegetation burning and it was therefore presumed to have minimal black carbon content. The soils were air-dried, ground, passed through a 2-mm sieve and stored in plastic bottles. For these soil samples, soil pH was determined in a slurry with a soil/water ratio of 1:1 [26]. Organic carbon was measured using the Walkley–Black method [27]. Soil texture was determined using the pipette method [28]. The cation exchange capacity (CEC) was measured by the ammonium acetate (pH 7.0) method [29]. The basic properties of these soils are listed in Table 1.

2.3. Sorption experiments

3-CP sorption by RSA was carried out using the batch method. 0.05 g of RSA was added into 25 mL of 0.01 M $CaCl_2$ solution in 50-mL brown glass centrifuge tubes and the tubes were then agitated for 1 h to homogenize the suspensions. Various amounts of the 3-CP stock solution were added into the samples to have the initial 3-CP concentrations of 3, 5, 10, 15, 20, 25, 30, 40 and 50 mg L^{-1}. To investigate the effect of pH on 3-CP sorption, the sorption experiment was conducted at pHs 4, 6, 8, and 10. The pH of the samples was adjusted frequently to maintain a constant value during the experiment. After the reaction was equilibrated at 25 °C for 48 h, the samples were filtered with 0.2-μm (pore size) PTFE syringe filters, which were pre-treated with isopropanol to become hydrophilic. The concentrations of 3-CP in the filtrates were determined using a high-performance liquid chromatograph (HPLC) equipped with a UV–vis detector (wavelength set to 274 nm) and a C-18 column. The mobile phase was a mixture of 65% methanol and 35% water at a flow rate of 1 mL min^{-1}. The amount of 3-CP sorbed by RSA was calculated as the difference between the initial and remaining concentrations of 3-CP and the data from triplicate samples were averaged.

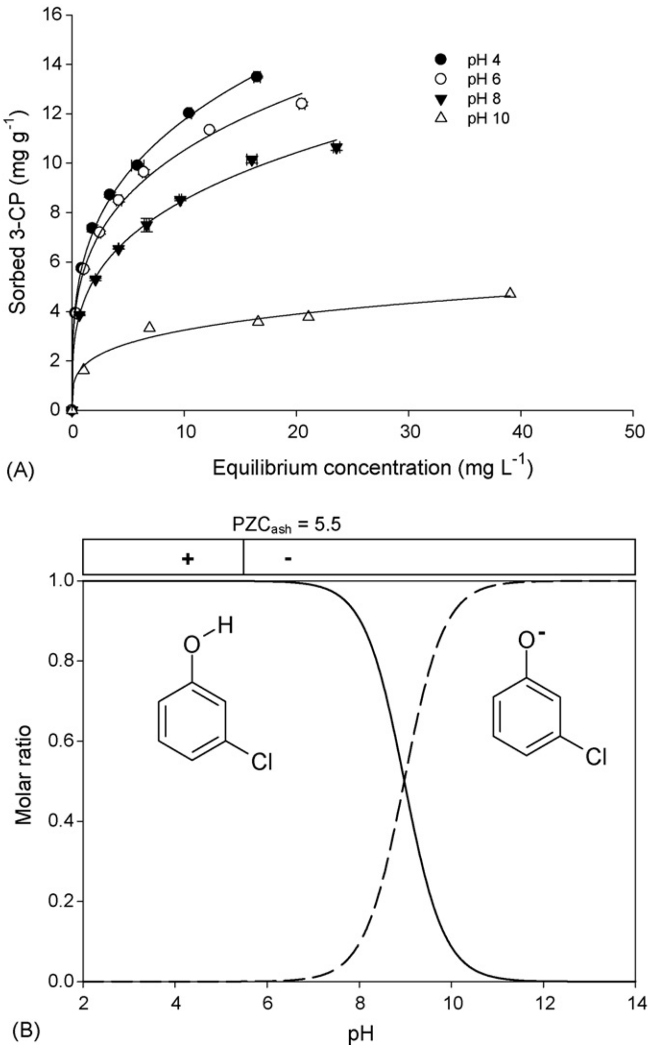

Fig. 1. (A) 3-CP sorption of rice-straw ash at pHs 4, 6, 8 and 10 and (B) the distributions of neutral and anionic 3-CP and the charge properties of rice-straw ash as a function of pH.

In the other set of experiments, the influence of RSA amendment on the 3-CP sorption of soils was studied with procedures similar to those described in the RSA system but, instead of RSA, 5 g of soil with a RSA content of 0%, 0.5%, 1% or 2% was used as the sorbent. The soils were suspended into 25 mL of 0.01 M $CaCl_2$ solution. The suspensions were equilibrated for 24 h while the pHs of the suspensions were maintained at 6. Subsequently, various amounts of 3-CP solutions were added into the soil suspensions and the samples were equilibrated at 25 °C for 48 h. The remained concentrations of 3-CP after sorption reaction were determined using HPLC.

3. Results and discussion

Fig. 1A shows the sorption isotherms of 3-CP onto RSA at various pHs at 25 °C. The L-shaped isotherms suggest that limited sites on RSA are available for 3-CP and that the mechanism of the interaction between RSA and 3-CP is surface sorption rather than partition. All the sorption isotherms at various pHs could be fit (R^2 > 0.97) with

Table 1
Physical and chemical properties of the soil samples.

Soil sample	pH	Organic matter (%)	Cation exchange capacity (cmol$_c$ kg^{-1})	Clay (%)	Silt (%)	Sand (%)	Soil texture
Soil 1	4.9	1.2	5.8	35	37	28	Clay loam
Soil 2	6.4	3.7	10.0	15	28	57	Sandy loam
Soil 3	7.6	1.9	6.9	27	53	20	Silt loam

Table 2
Langmuir parameters for 3-CP sorption on rice-straw ash and soils.

	Sample						
	Ash				Soil 1	Soil 2	Soil 3
	pH 4	pH 6	pH 8	pH 10	pH 6	pH 6	pH 6
S_m (mg g^{-1})	14	13	11	5	0.018	0.052	0.089
K_L (L mg^{-1})	0.69	0.64	0.47	0.25	0.29	0.10	0.02
r^2	0.977	0.992	0.981	0.978	0.932	0.980	0.998

the Langmuir model and the Langmuir sorption maximum (S_m) and energetic K_L constant were determined for each isotherm (Table 2). As the pH was increased from 4 to 8, the Langmuir maximum sorption decreased from 14 to 11 mg g^{-1}. A further increase in the pH dramatically reduced the Langmuir maximum sorption to 5 mg g^{-1}. Both the S_m and K_L values of 3-CP sorption on RSA decreased as the pH was increased (Table 2), which indicated a decreasing affinity of RSA for 3-CP with increasing pH.

It was found that an increase in pH led to a decrease in the sorption of 3-CP by RSA (Fig. 1A). A significant decrease occurred when the pH was increased to a value above the pK_a (9.0) of 3-CP [24]. 3-CP is a weak acid with a pK_a of 9.0 so its neutral and anionic forms prevail at pH <9 and pH >9, respectively (Fig. 1B). On the other hand, the PZC of RSA was determined to be 5.5 meaning that the surface of the sorbent has a net positive charge below its PZC and net negative charge above its PZC (Fig. 1B). Since both the sorbent and sorbate have functional groups that are subject to protonation and deprotonation, the negative effect of increasing pH on 3-CP sorption apparently arose from increasing electrostatic repulsion between sorbate and sorbent. Specifically, when the pH was increased from 8 to 10, the anionic form of 3-CP is mainly present due to deprotonation of the phenolic group of CP. The concomitant build-up of net negative charge on the surface of RSA thus led to an enhanced electrostatic repulsion between 3-CP and the RSA surface, which was responsible for the dramatic reduction in CP sorption from pHs 8 to 10 (Fig. 1A). Meanwhile, the solubility of the anionic form of 3-CP is higher than that of its neutral counterpart. When solution pH was increased, the increasing solubility of 3-CP due to the increasing distribution of its anionic form (Fig. 1B) also decreased the 3-CP sorption on RSA. Comparatively, the neutral form of 3-CP was preferentially sorbed by RSA. Burning rice straw resulted in the formation of carbonaceous materials with aromatic moieties in RSA. The sorption of 3-CP was therefore presumed to occur primarily on the surface of the carbon fraction of RSA, as those of inorganic constituents are expected to be highly hydrophilic and are therefore unlikely to contribute to a considerable extent of 3-CP sorption [30–33]. For the same reason, CPs are predominantly sorbed to organic matter in soils and the contribution of inorganic minerals to the overall CP sorption is insignificant [18–23].

Sorption isotherms of 3-CP at pH 6 for the soils containing various amounts of RSA are presented in Fig. 2. With or without RSA amendment, all the isotherms exhibited the characteristics of the Langmuir sorption isotherm. The Langmuir sorption maximum of soils 1, 2 and 3 were determined to be 18, 52 and 89 mg kg^{-1}, respectively, which were two to three orders of magnitude smaller than that of RSA (i.e., 13 mg g^{-1} or 13,000 mg kg^{-1}) (Table 2). Thus, 3-CP sorptions of the soils were low, thereby indicating a potentially high mobility of 3-CP in these soils. As illustrated by the isotherms of soils containing RSA, increasing RSA content significantly increased the 3-CP sorptions of these soils. For example, the Langmuir sorption maximum for soil 1 increased from 18 to 82, 160 and 274 mg kg^{-1}, as the RSA content was increased to 0.5%, 1% and 2%, respectively (Table 3). The carbon content in RSA was determined to be 26% using elemental analysis. When the RSA content was 0.5%, the corresponding carbon content was 0.13%, which is one order of magnitude smaller than the organic matter content of these soils (Table 1). Thus, the sorption contribution of the carbon fraction in RSA was very high, assuming the negligible contribution from the inorganic fraction of RSA [30–33]. These results showed that the presence of a small amount of black carbon in soils, resulting from the amendment of RSA, can dominate the sorption of 3-CP in soils.

Table 3
Langmuir sorption maximum of 3-CP for soils amended with various ash contents.

Ash content (%)	Langmuir sorption maximum, S_m (mg kg^{-1})		
	Soil 1	Soil 2	Soil 3
0	18	52	89
0.5	82	82	113
1.0	160	156	151
2.0	274	294	256

To model the 3-CP sorption of the soils, a Freundlich model on the basis of combined soil and RSA sorptions was proposed:

$$C_s = f_{soil}K_{F,soil}C_W^{n_{soil}} + f_{ash}K_{F,ash}C_W^{n_{ash}}$$

where C_s and C_W are the sorbate concentrations in soil and aqueous solution, respectively; f_{soil} and f_{ash} are the mass fractions of soil and RSA, respectively; $K_{F,soil}$ and $K_{F,ash}$ are the Freundlich distribution coefficients for soil and RSA, respectively; and n_{soil} and n_{ash} are the Freundlich exponents of soil and RSA, respectively. The values of the Freundlich parameters, K_F and n, determined for the soils and RSA are listed in Table 4.

With the above Freundlich model, one basic assumption is that the sorptions of 3-CP by soil and RSA in a soil sample containing

Table 4
Freundlich parameters for 3-CP sorption on rice-straw ash and soils at pH 6.

Sample	n	K_F[a] (mg^{1-n} L^n kg^{-1})	r^2
Ash	0.259	5814	0.969
Soil 1	0.395	4.05	0.960
Soil 2	0.515	5.78	0.997
Soil 3	0.644	3.88	0.995

[a] $K_F = C_S/C_W^n$, where C_s (mg kg^{-1}) and C_W (mg L^{-1}) are the sorbate concentrations in sorbent and aqueous solution, respectively.

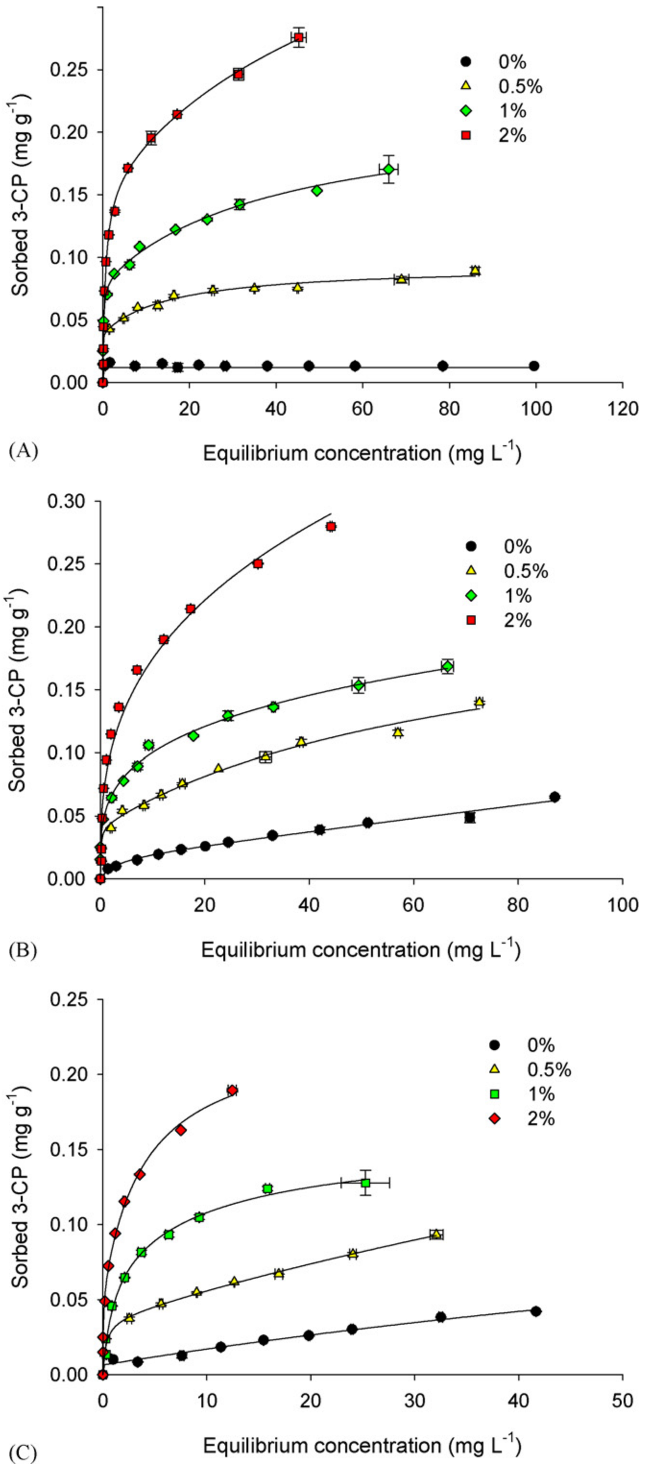

Fig. 2. Sorption isotherms of 3-CP by (A) soil 1, (B) soil 2 and (C) soil 3 amended with various contents of rice-straw ash.

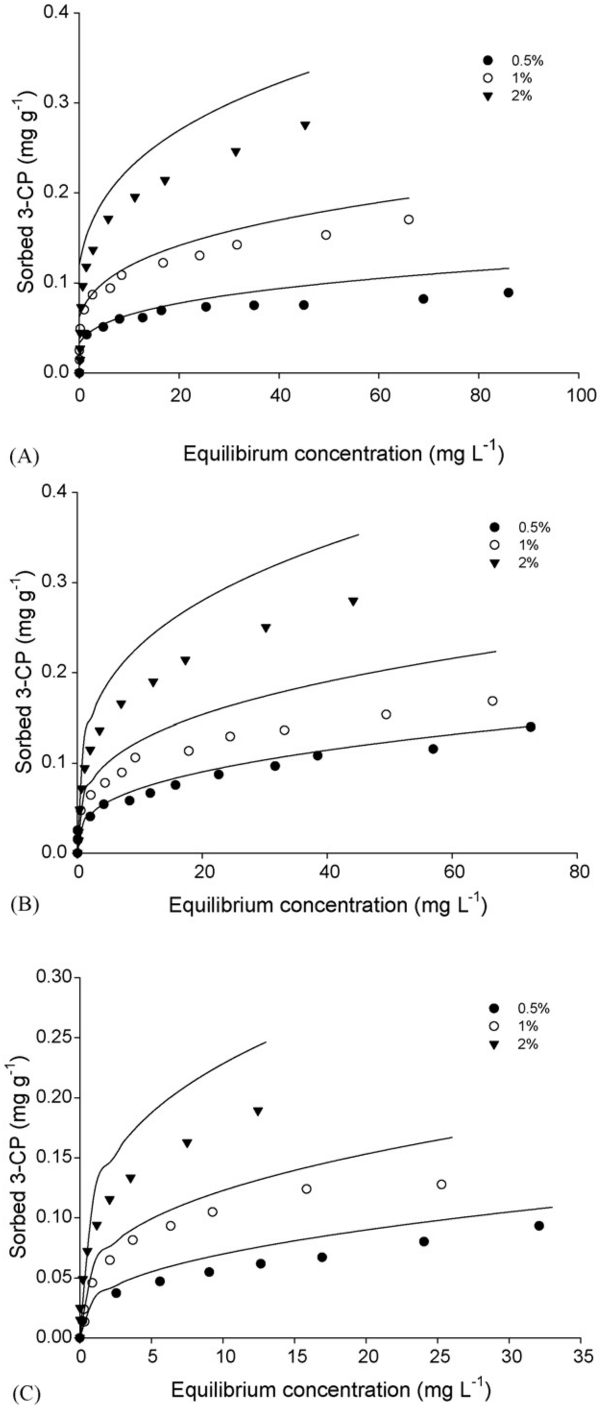

Fig. 3. Comparison of experimental 3-CP sorption of (A) soil 1, (B) soil 2 and (C) soil 3 with various ash contents with the predicted results from a Freundlich model on the basis of the combined sorptions of soil and rice-straw ash.

RSA are independent of each other, and their sum is equal to that of the soil sample containing RSA. Accordingly, the calculated results for the soils containing various amounts of RSA are shown in Fig. 3. When compared with the calculated amounts of sorbed 3-CP, the experimental results were all lower. The deviations indicated that 3-CP sorption of the RSA was suppressed in the soils. Because the mechanism of the interaction between the RSA and 3-CP is surface sorption, which proceeds through the binding of 3-CP to the limited sites on the surface of RSA, the suppression of the 3-CP sorption of RSA in the soils was attributed to competition of organic carbon or other soil components for the limited binding sites on the surface of black carbon in RSA. A similar result, reported in Cornelissen and Gustafsson [34], showed that the presence of organic matter can significantly reduce the sorption of phenanthrene to black carbon. As suggested in Pignatello et al. [35], the interaction of organic matter, including humic and fulvic acids, with the surface of black carbon may block the access of organic compounds to the surface sites and nanopore of black carbon. Thus, to correctly predict the sorption characteristics of CPs in soils containing black carbon derived from burning rice straw or other crop residues, the interaction of black carbon with organic matter needs to be considered. Otherwise, the contribution of black carbon to the overall 3-CP sorption in the soils can be overestimated.

4. Conclusions

The RSA amendment can enhance the overall 3-CP sorption of soils, and thus, may be applied to retard the movement of 3-CP in soils. In soils containing RSA, the contribution of RSA to the total sorption exceeds those of any other components in the soils, including organic matter. The high sorptivity of RSA for 3-CP is likely due to the content of black carbon in RSA. Thus, in soils containing RSA or other burned crop residues, the sorption to black carbon plays a central role in determining the mobility and bioavailability of CPs. Without considering the contribution of black carbon to the total sorption of soils containing burned crop residues, the contribution of organic matter or other components in soils can be significantly overestimated. Nonetheless, 3-CP sorption of RSA was suppressed in soils, which was attributed to competition of the organic matter or other soil components for the limited sites on the surface of black carbon in RSA. Thus, to predict the sorption and mobility of 3-CP or other small polar organic contaminants in agricultural soils, the effects of the black carbon content in the soils and the interaction between black carbon and other soil components need to be put into consideration.

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