Adsorption and Degradation of Triclosan and Triclocarban in Soils and Biosolids-Amended Soils

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Triclosan and triclocarban are antibacterial agents that are widely used in numerous personal care products. Limited information is available on their environmental behavior in soils and soils land applied with wastewaters and biosolids. In this study, laboratory experiments were performed to investigate their adsorption and degradation in soils. Both antibacterial agents adsorbed strongly to the sandy loam and silty clay soils with and without addition of biosolids, with distribution coefficients (K_d) ranging from 178 to 264 L kg^{-1} for triclosan and from 763 to 1187 L kg^{-1} for triclocarban. Sorption of triclosan decreased with increase in soil pH from 4 to 8, whereas triclocarban sorption showed no effect within the tested pH range. Competitive sorption was observed when triclosan and triclocarban coexisted, but the cosolute effect was concentration dependent. Biosolids amendment increased the sorption of triclosan and triclocarban, likely due to the addition of soil organic matter, but displayed no significant effect on degradation.

KEYWORDS: Antibacterial; personal care products; sorption; degradation; soil; biosolids amendment

INTRODUCTION

The extensive use of the antibacterial agents triclosan (5-chloro-2-(2,4-dichlorophenoxy)phenol) and triclocarban (3-(4-chlorophenyl)-1-(3,4-dichlorphenyl)urea) in household and personal care products has received increasing attention because of their potential to promote resistant bacteria (*1−3*) and their adverse effects on aquatic organisms (*4−7*). Studies on the occurrence and fate during wastewater treatment indicate that the removal efficiency of triclosan and triclocarban can be substantial from the aqueous phase when an activated sludge process is used (~90%) but is dependent on treatment techniques (*8−10*). Nevertheless, trace level residues are still detected in effluents, and considerable amounts of triclosan and triclocarban are sequestered and accumulated in biosolids (*11−13*). In surface water, triclosan is one of the most frequently detected organic contaminants found in U.S. streams (*14*), and triclocarban is reported to be a cocontaminant in both wastewater and surface water (*15*). In addition, triclosan has been shown to biotransform into methyltriclosan (*16*) and phototransform into 2,8-dichlorodibenzo-*p*-dioxin (*17, 18*), both of which are more hydrophobic and persistent than the parent compound (*16, 18*). Recently, triclosan and triclocarban were found to accumulate in algae, snails, and earthworms (*19, 20*).

Land application of reclaimed wastewater and biosolids is a common agricultural practice in the United States and worldwide (*21*). Their physicochemical properties (**Table 1**) indicate that both compounds are weak acids and are hydrophobic; thus, pH and soil organic matter can affect their environmental behavior in soils. Land application of biosolids can alter the chemical and physical properties of the soil, with an increase in soil−water retention and organic matter content being reported in both short-term and long-term experiments (*23−25*). The change in soil properties can consequently affect the interaction with chemicals. In this work, the effects of biosolids amendment on adsorption and degradation characteristics for triclosan and triclocarban in a sandy loam and a silty clay soil are investigated.

MATERIALS AND METHODS

Chemicals. Triclosan (≥97%), triclocarban (≥99%), and sodium azide (≥99.5%) were purchased from Sigma-Aldrich (St. Louis, MO). Ammonium acetate, anhydrous calcium chloride, sodium hydroxide, sulfuric acid, methanol, acetonitrile, and acetone were ACS certified and obtained from Fisher Chemicals (Fair Lawn, NJ). Deionized water (>18.0 MΩ-cm) was supplied by a Barnstead Nanopure system (Dubuque, IA).

Soils and Biosolids. A silt clay soil (BC) and a sandy loam soil (DL) were collected from the top layer (0−20 cm) in two fields with no biosolids application history in Lucas county, northwestern Ohio. The soil samples were air-dried, gently disaggregated, and sieved to a particle size of ≤2 mm. Biosolids were aerobically digested sludge generated at a local wastewater treatment plant (Oregon, OH), which uses activated sludge treatment techniques. Samples were taken directly from a field at the time of biosolids application. Biosolids contained 37.2 g L^{-1} total suspended solids and 20.5 g L^{-1} volatile suspended solids and had a pH of 7.9. Biosolids were mixed with BC and DL soils at a ratio of 1:2 (v/w) to simulate biosolids land applied soils, which was equivalent to an application rate of about 40 dry tons per hectare, assuming that biosolids are mixed with the top 15 cm of soil with a bulk density of 1.4 g cm^{-2}. This rate represents a high commercially viable application rate (*25*) and is comparable to the application rate used locally. The biosolids-amended soils (BCB and DLB) were air-dried and passed through a 2 mm sieve. All soil samples were stored in plastic bags in dark conditions at room temperature (23 ± 3 °C) for less than a month before use. Soil pH was measured in 0.01 M CaCl$_2$ at a ratio of 1:2 (w/v) and texture was

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determined using the pipet method according to U.S. Department of Agriculture (USDA) methods (*26*). Water-holding capacity (WHC) was determined gravimetrically (*27*). Soil organic matter (SOM) content was measured by loss on ignition at 450 °C for 4 h. Total organic carbon (TOC) content and cation exchange capacity (CEC) were determined by Spectrum Analytic Inc. (Washington Court House, OH). Properties of tested soils are listed in **Table 2**.

Sorption Experiment. Sorption of triclosan and triclocarban was studied using a batch equilibrium approach following the Organization for Economic Co-operation and Development (OECD) Guideline Test 106 (*28*). In a preliminary experiment, sorption kinetics were investigated by determining the liquid phase concentration at 2, 4, 8, 24, 48, 72, and 120 h. After 48 h, concentration varied by < 5%; thus, 48 h was chosen as the apparent equilibrium time for the sorption experiments. In a spiked control experiment, sorption on test vessel surfaces was observed for both triclosan and triclocarban (see Supporting Information for details); therefore, both aqueous phase and sorbed phase concentrations were analyzed as recommended in the OECD Guidelines (*28*).

In the sorption experiment, soil samples (1 g, dry weight) were mixed with 40 mL of 0.01 M $CaCl_2$ solution containing 0.1% NaN_3 (w/v) in 50 mL glass centrifuge tubes, sealed with Teflon-lined screw caps, and agitated for 24 h using a reciprocal shaker. NaN_3 was used to inhibit the activity of microbes. In the preliminary experiment, no significant change in concentration of soil solution was observed after 48 h, suggesting that no degradation occurred in the presence of NaN_3. The soil slurry was then spiked with 0.04 mL of standard solution prepared in acetone (final acetone content = 0.1%) to achieve concentrations of 0.1, 0.4, 0.8, 1.4, and 2.0 mg L^{-1}. After agitation for 48 h, samples were withdrawn and centrifuged at 1500*g* for 20 min. Aliquots of 0.5 mL of supernatant were transferred into 2 mL amber glass vials and diluted with 0.5 mL of methanol to reduce sorption to the vials.

The remaining supernatant was decanted, and soil samples were then freeze-dried and extracted using accelerated solvent extraction (ASE) with a Dionex ASE 200 system. Briefly, freeze-dried samples were mixed with Ottawa sand and transferred into 11 mL extraction cells. The cells were sealed at the bottom with glass fiber filters, filled with Ottawa sand to the top, and tightened with screw caps. The prepared cells were extracted with methanol using the following conditions: preheat, 0 min; static, 5 min; flush, 60%; cycles, 2; purge, 120 s; oven temperature, 80 °C; pressure, 1500 psi. The extracted samples were collected in 60 mL glass vials. Final volume was about 20 mL. Aliquots of 1 mL extracted samples were transferred into amber glass vials. Extraction recoveries were > 80% for

both compounds with < 10% relative standard deviation (see Supporting Information).

Two specific experiments at a single initial concentration of 0.4 mg L^{-1} were performed to investigate the effects of pH and cosolute. The pH values of soil slurries were adjusted by using 5% (v/v) sulfuric acid and/or 1 M sodium hydroxide to achieve pH values ranging from 4 to 8. Soils were equilibrated overnight, the pH was checked, and the process was repeated to achieve targeted pH values. Then tubes were spiked, agitated for 48 h, and sampled. The addition of standard solution showed no impact on the pH of soil solution. In cosolute experiments, the initial concentration of one solute was fixed and the concentration of cosolute varied from 0.1 to 2.0 mg L^{-1}.
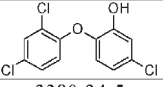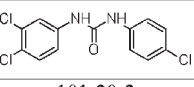
All sorption experiments were performed in duplicate. Blank samples without spiking were also included. Prepared samples were analyzed using liquid chromatography–tandem mass spectrometry (LC-MS/MS).

Degradation Experiment. The degradation of triclosan and triclocarban was investigated under aerobic conditions. Five gram aliquots of soil were weighed into 20 mL glass scintillation vials. The soil moisture was adjusted to 15% using deionized water, which corresponded to 28, 28, 33, and 32% WHC for BC, BCB, DL, and DLB, respectively. After a 5 day acclimation period, each sample was spiked with 0.1 mL of 0.1 mg mL^{-1} triclosan or triclocarban standard solution prepared in acetone, providing a theoretical concentration of 2 mg kg^{-1}. This concentration is toward the high end of concentration that can be expected in soils (*20*). The vials were put under a ventilation hood for 1 h to allow acetone to evaporate, and then soil samples were mixed manually with a stainless steel spatula. Mixed samples were covered with aluminum foil and incubated in the dark at room temperature. The soil moisture was monitored every other day by weighing and was adjusted with deionized water to compensate for any water loss. Samples were withdrawn at specified times (0, 5, 10, 20, 30, 40, and 60 days) and used to determine the residue of each compound. Collected samples were freeze-dried, and 2 g subsamples were extracted using ASE as previously described and subsequently analyzed using LC-MS/MS.

Analytical Method. Instrumental analysis was performed using a Varian 1200 L LC-MS/MS. Separation was performed on a Phenomenex Luna C8 (*2*) column (100 × 4.6 mm, 3 μm) with SecurityGuard guard column (4 × 2.0 mm). The MS/MS detection was conducted in negative ionization mode under selected reaction monitoring (SRM). The transition was from *m/z* 286.7 to 35.1 for triclosan and from *m/z* 312.7 to 159.5 for triclocarban. External calibration curves were used for quantification, and the regression coefficients (R^2) of the curves were > 0.99. Matrix effects were also evaluated and found to be not significant. Detailed methodology is provided in the Supporting Information.

Data Analysis. The sorption data were fit using a linear sorption model, $C_s = K_d \times C_w$ and Freundlich sorption model, $C_s = K_f \times C_w^n$, where K_d (L kg^{-1}) and K_f (μg^{1-n} L^n kg^{-1}) are sorption coefficients, C_s (mg kg^{-1}) is the solid phase concentration, C_w (mg L^{-1}) is the liquid phase concentration, and n is the linearity constant. Organic carbon normalized sorption coefficient K_{oc} (L kg^{-1}) was calculated using the following equation: $K_{oc} = K_d/f_{oc}$, where f_{oc} (%) is the TOC content. All blank samples were analyzed, and only triclocarban (2.1 ng g^{-1}) was detected in biosolids-amended soils, but not in the unamended soils. Thus, for sorption in biosolids-amended soils, concentrations were adjusted using blank samples. The degradation kinetics were described using a first-order reaction model: $C_t = C_0 \times e^{-kt}$, where C_0 (mg kg^{-1}) is the initial concentration, C_t (mg kg^{-1}) is the concentration at time t (days), and k (day^{-1}) is the rate constant. The half-life $t_{1/2}$ (days) was calculated as

Table 1. Structures and Selected Properties of Triclosan and Triclocarban[a]

	triclosan	triclocarban
CAS number	3380-34-5	101-20-2
molecular weight	289.5 g mol^{-1}	315.6 g mol^{-1}
melting point	54-57 °C	255-256 °C
solubility (water)	≤1 mg L^{-1}	<0.05 mg L^{-1}
solubility (acetone)	very soluble	40 g L^{-1}
log P (octanol/water)	4.8	4.9
pK_a	7.9	12.7

[a] Physicochemical properties are from ref (*22*).

Table 2. Properties of Tested Soils

soil	texture	soil pH	slurry pH[c]	WHC (%)	SOM (%)	TOC (%)	CEC (cmol kg^{-1})	particle size (%)		
								sand	silt	clay
BC	silty clay	4.7	5.9	53.1	4.59	1.61	16.6	4.8	47.6	46.9
BCB[a]	silty clay	6.2	6.3	53.2	5.58	1.66	18.0	5.5	47.6	46.9
DL	sandy loam	4.1	5.8	45.7	4.24	1.56	9.5	80.4	8.4	11.2
DLB[b]	sandy loam	5.9	6.6	46.1	5.17	1.57	8.1	81.3	5.4	13.3

[a] Soil BC amended with biosolids. [b] Soil DL amended with biosolids. [c] Soil solution pH for the sorption experiment.

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$t_{1/2} = \ln(2)/k$. Isotherm parameters were estimated using SPSS v15.0 software (Chicago, IL).

RESULTS AND DISCUSSION

Sorption Isotherm. Isotherms for triclosan and triclocarban in tested soils are presented in **Figure 1**, and estimated parameters are listed in **Table 3**. The sorption isotherms can be well described using both linear and Freundlich models ($R^2 > 0.92$), although the estimated n value using the Freundlich model indicates a nonlinear sorption behavior for triclosan in the DLB soil and for triclocarban in BCB, DL, and DLB soils. In all tested soils, triclocarban had a stronger sorption than triclosan ($p < 0.05$, one-tailed t test), likely due to the weaker hydrophobicity of triclosan. For both compounds, K_d and K_{oc} were significantly
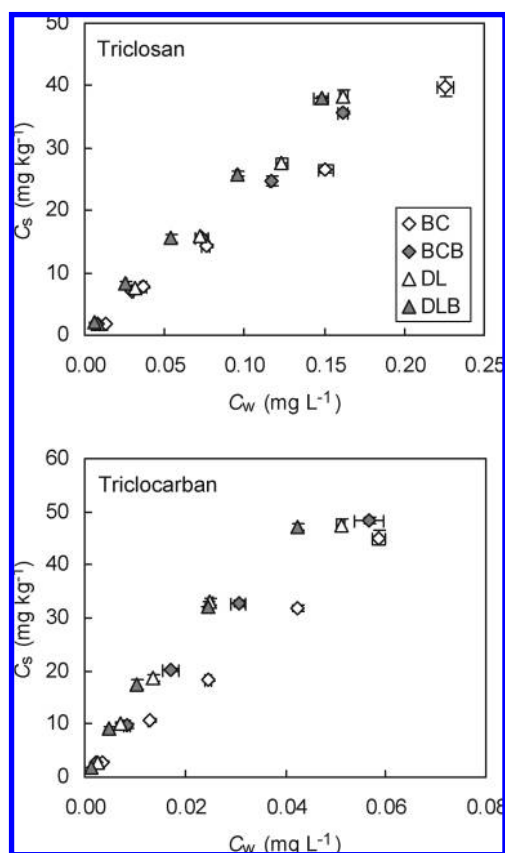

Figure 1. Sorption isotherm for triclosan and triclocarban in unamended and amended soils. (Each point and error bar represents the mean and standard error of two replicates.)

higher in the DL soil compared to the BC soil ($p < 0.05$). Noting that BC soil had slightly higher SOM, TOC, and CEC, the stronger sorption in the DL soil might be attributed to the difference in soil organic matter. Previously, the sorption of organic contaminant was found to be affected by the quality of soil. Xing found that sorption of naphthalene increased with increasing aromaticity and decreasing polarity of SOM (*29*). Chefetz et al. demonstrated the importance of aliphatic structures in natural organic matter on the sorption of pyrene (*30*). In addition, decreased sorption was also observed when organic matter was associated with minerals (*31*). The high clay content in BC soil might hinder the interaction between the solute and SOM.

Biosolids Effect on Sorption. Biosolids amendment by land application alters the properties of soil. Here, increasing soil pH, SOM, TOC, and CEC were observed as a result of biosolids amendment (**Table 2**). Biosolids-amended soils showed a significantly higher sorption for both triclosan and triclocarban ($p < 0.05$); however, no significant difference was observed for triclocarban in BC and BCB soils ($p = 0.12$). The increased sorption in the amended soil can be primarily attributed to the increase in SOM. Amendment also affects the sorption of triclosan by changing soil pH. Here, both soil pH and soil solution pH increased, resulting in a reduction in sorption (see following section). However, an increase of sorption here suggests that increase of SOM due to biosolids amendment is more important in determining the overall sorption. Previously, the amount of organic carbon in soils was found to increase linearly with biosolids amendment (*23*). Long-term and repeated biosolids land application showed that the effect of biosolids on soil pH and organic matter can last for many years (*32−34*). Thus, the biosolids effect on sorption is not transient and can persist for a long time.

pH Effect on Sorption. The change of soil solution pH can affect the speciation and consequently the sorption of these two compounds. By increasing the pH of soil solution within the environmentally relevant range of 4−8, a decrease in sorption was observed for triclosan, whereas the sorption of triclocarban was nearly unchanged (**Figure 2**). The trends followed the expected pH−pK_a relationship. At any pH, the fraction of the neutral (α^0) form can be calculated as $\alpha^0 = 1/(1 + 10^{pH-pKa})$ and the fraction of the anionic (α^-) form can be calculated as $\alpha^- = 1 - \alpha^0$. As the pH of the soil solution increased from 4 to 8, the amount of triclosan existing in the neutral form decreased from 100 to 39%, whereas almost 100% of the triclocarban existed in the neutral form even at pH 8, due to a high pK_a value. To examine the contribution of individual species to overall sorption for triclosan, the single-point sorption coefficient of neutral (K_d^0) and anionic (K_d^-) was estimated by fitting the data using the following

Table 3. Estimated Linear, OC Normalized, and Freundlich Model Sorption Parameters for Triclosan and Triclocarban

	K_d (L kg^{-1})	R^2	K_{oc} (L kg^{-1})	K_f (μg^{1-n} L^n kg^{-1})	n	R^2
			Triclosan			
BC	178 (173−183)[a]	0.99	11397	235 (155−314)	0.947 (0.881−1.012)	0.99
BCB	217 (210−224)	0.99	13847	216 (108−323)	1.002 (0.899−1.104)	0.99
DL	231 (223−278)	0.99	14348	200 (106−295)	1.029 (0.933−1.125)	0.99
DLB	264 (251−277)	0.98	15892	483 (321−646)	0.873 (0.802−0.944)	0.99
			Triclocarban			
BC	763 (734−792)	0.99	48865	799 (457−1140)	0.988 (0.877−1.099)	0.99
BCB	917 (819−1015)	0.94	58656	2294 (1342−3245)	0.758 (0.647−0.869)	0.98
DL	1029 (899−1159)	0.92	64037	2624 (1381−3867)	0.745 (0.614−0.876)	0.97
DLB	1187 (1073−1300)	0.95	71687	2939 (2429−3449)	0.744 (0.692−0.792)	0.99

[a] 95% confidence interval.

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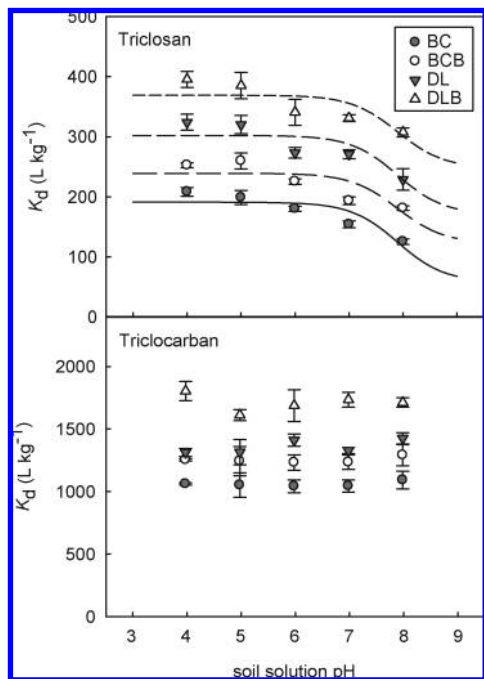*J. Agric. Food Chem.,* Vol. 57, No. 11, 2009   **4903**

Figure 2. Effect of soil solution pH on the sorption of triclosan and triclocarban. (Each point and error bar represents the mean and standard error of two replicates.)

Table 4. Sorption Coefficients for Neutral and Anionic Forms of Triclosan

soil	K_d^0 (L kg^{-1})	K_d^- (L kg^{-1})	R^2
BC	191 (176−206)[a]	58 (4−112)	0.763
BCB	239 (219−258)	122 (55−189)	0.613
DL	302 (280−325)	170 (95−244)	0.623
DLB	369 (343−394)	247 (158−337)	0.490

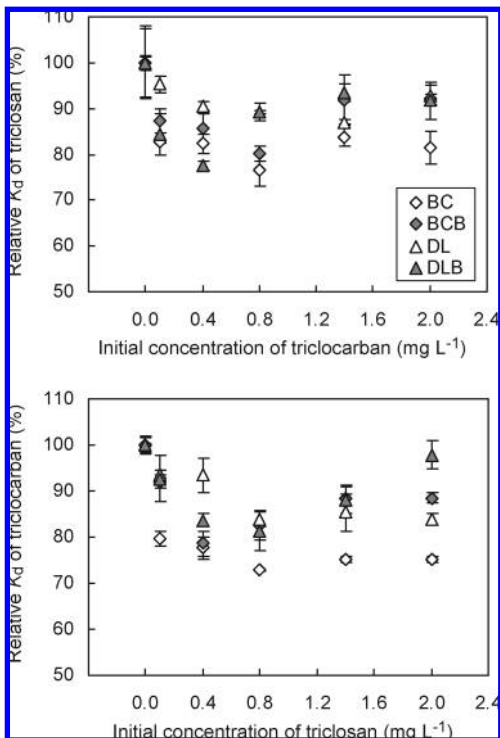[a] 95% confidence interval in parentheses.

Figure 3. Competitive sorption of triclosan and triclocarban in unamended and amended soils. (Each point and error bar represents the mean and standard error of two replicates.)
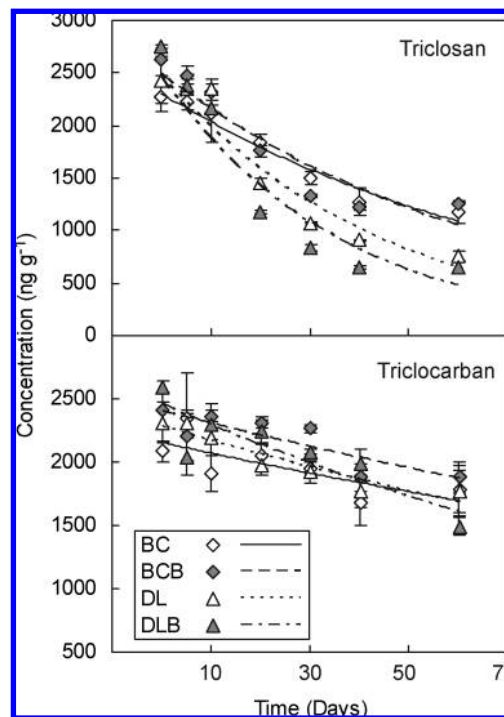

Figure 4. Degradation of triclosan and triclocarban in unamended and amended soils under aerobic condition. (Each point and error bar represents the mean and standard error of two replicates.)

Table 5. Degradation Rate and Half-Life of Triclosan and Triclocarban in Tested Soils

soil	triclosan			triclocarban		
	k	R^2	$t_{1/2}$	k	R^2	$t_{1/2}$
BC	0.012 (0.010−0.014)[a]	0.956	58	0.003 (0.001−0.006)	0.506	231
BCB	0.017 (0.013−0.021)	0.912	41	0.004 (0.002−0.006)	0.709	139
DL	0.022 (0.016−0.029)	0.923	32	0.005 (0.004−0.007)	0.901	173
DLB	0.034 (0.026−0.042)	0.957	20	0.008 (0.005−0.012)	0.663	87

[a] 95% confidence interval.

equation: $K_d = K_d^0 \alpha^0 + K_d^- \alpha^-$ (*35*), and the results are provided in **Table 4**. For all tested soils, neutral species had higher K_d^0 than K_d^-, suggesting that the hydrophobic interaction of un-ionized species is more important to overall sorption. However, sorption of the anionic form was also considerably strong, as indicated by K_d^-. Thus, the increase in pH can only slightly decrease sorption of triclosan in soils. For triclocarban, sorption was preliminarily attributed to the neutral species, whereas that of the anionic form could not be examined within the tested pH range. With such a high pK_a value, sorption of triclocarban in typical soils is unlikely to be affected by pH.

Cosolute Effect on Sorption. Because triclosan and triclocarban can be present in the environment at the same time (*15*), their coexistence in soils can affect their mutual sorption behavior. In the cosolute experiments, decreased sorption was observed when the cosolute existed at low concentration, whereas sorption was less affected or remained unchanged as concentrations were increased (**Figure 3**). This trend was true for both compounds. The sorption decrease at low concentration is likely due to the competition for available sites. However, as the concentration of cosolute increases, large amounts of sorbed solute may cause swelling and disordering of the SOM, resulting in an increase of sorption sites (*36*).

Degradation in Soils. In soil, triclosan and triclocarban were found to be degradable biologically only under aerobic conditions but persistent under anaerobic conditions (*37*). Here, a degradation experiment was performed under aerobic conditions with the purpose of understanding the effects of biosolids amendment on the degradation of these two compounds. The concentrations of triclosan and triclocarban during the experiment are presented in **Figure 4**, and estimated parameters using a first-order kinetic model are listed in **Table 5**. The estimated half-life ($t_{1/2}$, days) ranges from 20 to 58 days for triclosan and from 87 to 231 days for triclocarban. The results correspond well with the data from Ying et al. (*37*), who reported half-lives of 18 days for triclosan and 108 days for triclocarban in a loam soil. Triclocarban was degraded to a lesser extent compared to triclosan in all treatments and may be attributed to the sorptive strength limiting the availability to soil microorganisms.

Degradation was significantly faster in the DL soil compared to the BC soil for triclosan ($p < 0.05$) but not for triclocarban ($p = 0.47$). The different degradation rates in the two soils could be attributed to their differences in texture, microbial activity, and amount of water in the system. Because the same moisture was used during the experiment, in the BC soil, less water might have been available to microbes due to a high clay content and WHC (**Table 2**). In biosolids-amended soils, both compounds appeared to degrade quickly. However, no statistical difference ($p > 0.05$) was found by comparing the slopes of the logarithmically transformed data for each treatment. This indicates that biosolids amendment had no significant effect on the degradation rate.

In conclusion, both triclosan and triclocarban had a strong affinity in the sandy loam and silty clay soils. Increasing pH can slightly decrease the sorption of triclosan, whereas for triclocarban no pH effect was observed within an environmentally relevant range. Competitive sorption was observed when both compounds were present. However, the cosolute effect is concentration dependent. At an environmentally relevant concentration, the coexistence of both compounds will likely reduce the sorption strength. Degradation data suggest that triclosan can persist in soils for several days to months and triclocarban can persist in soils for several months to years. Under field conditions these two compounds can be even more stable, considering the possible anaerobic conditions and drought events. Amendment of biosolids increased the sorptive strength but had no testable effect on degradation. Further research is needed to study the effect of triclosan and triclocarban on soil microorganisms and their potential to be taken up by plants.

Supporting Information Available: Instrumental analysis, method performance, quantification, and loss to vessel. This material is available free of charge via the Internet at http://pubs.acs.org.

LITERATURE CITED

(1) Schweizer, H. P. Triclosan: a widely used biocide and its link to antibiotics. *FEMS Microbiol. Lett.* **2001**, *202*, 1–7.

(2) Walsh, S. E.; Maillard, J. Y.; Russell, A. D.; Catrenich, C. E.; Charbonneau, D. L.; Bartolo, R. G. Development of bacterial resistance to several biocides and effects on antibiotic susceptibility. *J. Hosp. Infect.* **2003**, *55*, 98–107.

(3) Levy, S. B. Antibiotic and antiseptic resistance: impact on public health. *Pediatr. Infect. Dis. J.* **2000**, *19*, S120–S122.

(4) Ishibashi, H.; Matsumura, N.; Hirano, M.; Matsuoka, M.; Shiratsuchi, H.; Ishibashi, Y.; Takao, Y.; Arizono, K. Effects of triclosan on the early life stages and reproduction of medaka *Oryzias latipes* and induction of hepatic vitellogenin. *Aquat. Toxicol.* **2004**, *67*, 167–179.

(5) DeLorenzo, M. E.; Fleming, J. Individual and mixture effects of selected pharmaceuticals and personal care products on the marine phytoplankton species *Dunaliella tertiolecta*. *Arch. Environ. Contam. Toxicol.* **2008**, *54*, 203–210.

(6) Veldhoen, N.; Skirrow, R. C.; Osachoff, H.; Wigmore, H.; Clapson, D. J.; Gunderson, M. P.; Van Aggelen, G.; Helbing, C. C. The bactericidal agent triclosan modulates thyroid hormone-associated gene expression and disrupts postembryonic anuran development. *Aquat. Toxicol.* **2006**, *80*, 217–227.

(7) Yang, L.-H.; Ying, G.-G.; Su, H.-C.; Stauber, J. L.; Adams, M. S.; Binet, M. T. Growth-inhibiting effects of 12 antibacterial agents and their mixtures on the freshwater microalga *Pseudokirchneriella subcapitata*. *Environ. Toxicol. Chem.* **2008**, *27*, 1201–1208.

(8) Heidler, J.; Sapkota, A.; Halden, R. U. Partitioning, persistence, and accumulation in digested sludge of the topical antiseptic triclocarban during wastewater treatment. *Environ. Sci. Technol.* **2006**, *40*, 3634–3639.

(9) Thompson, A.; Griffin, P.; Stuetz, R.; Cartmell, E. The fate and removal of triclosan during wastewater treatment. *Water Environ. Res.* **2005**, *77*, 63–67.

(10) McAvoy, D. C.; Schatowitz, B.; Jacob, M.; Hauk, A.; Eckhoff, W. S. Measurement of triclosan in wastewater treatment systems. *Environ. Toxicol. Chem.* **2002**, *21*, 1323–1329.

(11) Heidler, J.; Halden, R. U. Mass balance assessment of triclosan removal during conventional sewage treatment. *Chemosphere* **2007**, *66*, 362–369.

(12) Ying, G.-G.; Kookana, R. S. Triclosan in wastewaters and biosolids from Australian wastewater treatment plants. *Environ. Int.* **2007**, *33*, 199–205.

(13) Bester, K. Triclosan in a sewage treatment process—balances and monitoring data. *Water Res.* **2003**, *37*, 3891–3896.

(14) Kolpin, D. W.; Furlong, E. T.; Meyer, M. T.; Thurman, E. M.; Zaugg, S. D.; Barber, L. B.; Buxton, H. T. Pharmaceuticals, hormones, and other organic wastewater contaminants in U.S. streams, 1999−2000: a national reconnaissance. *Environ. Sci. Technol.* **2002**, *36*, 1202–1211.

(15) Halden, R. U.; Paull, D. H. Co-occurrence of triclocarban and triclosan in U.S. water resources. *Environ. Sci. Technol.* **2005**, *39*, 1420–1426.

(16) Lindström, A.; Buerge, I. J.; Poiger, T.; Bergqvist, P.-A.; Müller M. D.; Buser, H.-R. Occurrence and environmental behavior of the bactericide triclosan and its methyl derivative in surface waters and in wastewater. *Environ. Sci. Technol.* **2002**, *36*, 2322–2329.

(17) Latch, D. E.; Packer, J. L.; Arnold, W. A.; McNeill, K. Photochemical conversion of triclosan to 2,8-dichlorodibenzo-*p*-dioxin in aqueous solution. *J. Photochem. Photobiol. A* **2003**, *158*, 63–66.

(18) Aranami, K.; Readman, J. W. Photolytic degradation of triclosan in freshwater and seawater. *Chemosphere* **2007**, *66*, 1052–1056.

(19) Coogan, M. A.; Point, T. W. L. Snail bioaccumulation of triclocarban, triclosan, and methyltriclosan in a north Texas, USA, stream affected by wastewater treatment plant runoff. *Environ. Toxicol. Chem.* **2008**, *27*, 1788–1793.

(20) Kinney, C. A.; Furlong, E. T.; Kolpin, D. W.; Burkhardt, M. R.; Zaugg, S. D.; Werner, S. L.; Bossio, J. P.; Benotti, M. J. Bioaccumulation of pharmaceuticals and other anthropogenic waste indicators in earthworms from agricultural soil amended with biosolid or swine manure. *Environ. Sci. Technol.* **2008**, *42*, 1863–1870.

(21) Chang, A. C.; Pan, G.; Page, A. L.; Asano, T. *Developing Human Health-Related Chemical Guidelines for Reclaimed Water and Sewage Sludge Applications in Agriculture*; World Health Organization: Geneva, Switzerland, 2002.

(22) Loftsson, T.; Össurardóttir, Í. B.; Thorsteinsson, T.; Duan, M.; Másson, M. Cyclodextrin solubilization of the antibacterial agents

Article

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triclosan and triclocarban: effect of ionization and polymers. *J. Inclusion Phenom. Macrocycl. Chem*. **2005**, *52*, 109–117.

(23) Khaleel, R.; Reddy, K. R.; Overcash, M. R. Change in soil physical properties due to organic waste applications: a review. *J. Environ. Qual*. **1981**, *10*, 133–141.

(24) Lindsay, B. J.; Logan, T. J. Field response of soil physical properties to sewage sludge. *J. Environ. Qual*. **1998**, *27*, 534–542.

(25) Moffet, C. A.; Zartman, R. E.; Wester, D. B.; Sosebee, R. E. Surface biosolids application: effects on infiltration, erosion, and soil organic carbon in Chichuahuan desert grasslands and shrublands. *J. Environ. Qual*. **2005**, *34*, 299–311.

(26) USDA. Soil survey laboratory methods manual (version 4.0), 2004.

(27) Wang, W.; Dala, R. C.; Moody, P. W. Evaluation of the microwave irradiation method for measuring soil microbial biomass. *Soil Sci. Soc. Am. J*. **2001**, *65*, 1696–1703.

(28) *OECD Guidelines for the Testing of Chemicals: Test No. 106 Adsorption Desorption Using a Batch Equilibrium Method*; Organization for Economic Cooperation and Development: Paris, France, 2000.

(29) Xing, B. The effect of the quality of soil organic matter on sorption of naphthalene. *Chemosphere* **1997**, *35*, 633–642.

(30) Chefetz, B.; Deshmukh, A. P.; Hatcher, P. G. Pyrene sorption by natural organic matter. *Environ. Sci. Technol*. **2000**, *34*, 2925–2930.

(31) Bonin, J. L.; Simpson, M. J. Variation in phenanthrene sorption coefficents with soil organic matter fractionation: the result of structure or conformation?. *Environ. Sci. Technol*. **2007**, *41*, 153–159.

(32) Mantovi, P.; Baldoni, G.; Toderi, G. Reuse of liquid, dewatered, and composted sewage sludge on agricultural land: effects of long-term application on soil and crop. *Water Res*. **2005**, *39*, 289–296.

(33) Sullivan, T. S.; Stromberger, M. E.; Paschke, M. W.; Ippolito, J. A. Long-term impacts of infrequent biosolids applications on chemical and microbial properties of a semi-arid rangeland soil. *Biol. Fertil. Soils* **2006**, *43*, 258–266.

(34) Walter, I.; Cuevas, G. Chemical fractionation of heavy metals in a soil amended with repeated sewage sludge application. *Sci. Total Environ*. **1999**, *226*, 113–119.

(35) Lee, L. S.; Rao, P. S. C.; Nkedi-Kizza, P.; Delfino, J. J. Influence of solvent and sorbent characteristics on distribution of pentachlorophenol in octanol-water and soil-water systems. *Environ. Sci. Technol*. **1990**, *24*, 654–661.

(36) Weber, W. J.; Kim, S. H.; Johnson, M. D. Distributed reactivity model for sorption by soils and sediments. 15. High-concentration co-contaminant effects on phenanthrene sorption and desorption. *Environ. Sci. Technol*. **2002**, *36*, 3625–3634.

(37) Ying, G.-G.; Yu, X.-Y.; Kookana, R. S. Biological degradation of triclocarban and triclosan in a soil under aerobic and anaerobic conditions and comparison with environmental fate modelling. *Environ. Pollut*. **2007**, *150*, 300–305.