Sorption of triclosan onto activated carbon, kaolinite and montmorillonite: Effects of pH, ionic strength, and humic acid

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ABSTRACT

Sorption of triclosan on three sorbents, viz., activated carbon, kaolinite and montmorillonite was studied as a function of pH, ionic strength and humic acid (HA) concentration through controlled batch experiments. Triclosan sorption was found to be higher in the acidic pH range, as varying pH showed significant influence on the surface charge of the sorbents and degree of ionization of the sorbate. Sorption capacity of the sorbents increased with an increase in the ionic strength of solution. At low pH (pH 3), the overall increase in triclosan sorption was 1.2, ~4 and 3.5 times, respectively for activated carbon, kaolinite and montmorillonite when ionic strength was increased from 1×10^{-3} to 5×10^{-1} M. Triclosan sorption onto activated carbon decreased from 31.4 to $10.6\,\mathrm{mg\,g^{-1}}$ by increasing the HA concentration to $200\,\mathrm{mg\,C\,L^{-1}}$. However, during sorption onto kaolinite and montmorillonite, the effect of HA was very complex probably due to (i) hydrophobicity ($\log K_{ow} = 4.76$) of triclosan; and (ii) complexation of HA with triclosan. Though triclosan sorption onto activated carbon is higher, the potential of kaolinite and montmorillonite in controlling the transport of triclosan in subsurface environment can still be appreciable.

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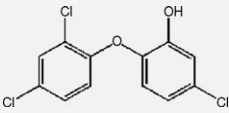
1. Introduction

Triclosan [5-chloro-2-(2,4-dichlorophenoxy)-phenol], a very common antimicrobial agent, is currently used in a large number of consumer products such as toothpastes, detergents, shampoos, body washes, deodorants, lotions, and dish washing liquids. It is also used as an additive in plastic, polymer and textile manufacturing industry. The inherent antibacterial property of triclosan makes them a more effective agent for use in household sponges, kitchen chopping boards, socks and inner garments. The incorporation of triclosan in this vast array of products results in its discharge to wastewater treatment plants (WWTPs) and then into surface waters [1,2]. Triclosan is reported to be highly toxic to certain aquatic organisms, particularly to green algae [3,4]. Besides, triclosan has been shown to indirectly affect aquatic organisms by blocking enzyme carrying proteins, leading to concerns of the possible build-up of bacterial resistance in these organisms [5]. Another concern of the presence of triclosan in surface water is the formation of more toxic by-products such as dioxin and endocrine disruptor chemicals [6]. Not only the presence in sewage treatment plants and surface waters but also the presence in human milk has raised serious concern over the use of triclosan in the last few years [7].

Triclosan is a chlorinated phenoxyphenol with a pK_a of 8.14. Although a hydroxyl functional group is present, it is still relatively hydrophobic with $\log K_{ow}$ of 4.76 [8] (Table 1). Due to its extreme hydrophobic nature, triclosan remains highly hydrophobic even in an ionized state [5]. This relatively high octanol–water partition coefficient of the compound can lead to sorption to particles [9]. Besides, sorption to solid phase is accounted for most of the removal of triclosan in a typical activated sludge treatment process [10]. With a pK_a value of 8.14, triclosan exists in both neutral and ionized (negatively charged) forms with a distribution ratio of 50:50 in most of the wastewater treatment plants operating generally at pH ~8.0 [5]. However, being partially ionized, a considerable proportion of triclosan still remains in the aqueous phase of treated wastewater effluent due to its incomplete removal in WWTPs [10]. Thus, it appears imperative to explore, develop, and improve technologies for the elimination of such compounds to ensure a sustainable healthy natural environment.

Adsorption process has the advantages of applicability at very low concentrations, suitable for both batch and continuous processes, ease of operation, little sludge generation, possibility of regeneration and reuse, and low capital cost [11]. Adsorption using activated carbon is ideal for removing small molecular organic compounds due to high surface area, and the combination of well developed pore structure and surface functional group properties. Moreover, the presence of surface functional groups such as carboxyl, phenolic, lactone, lactol and quinone and their concentration levels play an important role in the adsorption capacity and

Table 1
Physicochemical properties of triclosan[a].

CAS No.	Molecular structure	MW	Log K_{ow}	pK_a	Water solubility (mg L^{-1})
3380-34-5		289.5	4.76	8.14	10

[a] Data from Trenholm et al. [9].

removal mechanism of the sorbates [12,13]. Activated carbons can play an important role in the removal of contaminants arising from pharmaceutical industry, which is based on the assumption that these contaminants are similar to pesticides in both physicochemical properties and concentration levels present in water bodies [14]. In spite of this fact, due to the relatively high cost of activated carbons and difficulty in their regeneration, there have been attempts to utilize low cost, naturally occurring sorbents that do not represent an economic cornerstone for the large scale implementation to remove trace organic and inorganic contaminants from water and wastewater. The wide usefulness of clays lies in the presence of a variety of surface and structural properties such as high cation exchange capacity, low point of zero charge (PZC), high specific surface area and high chemical and mechanical stability, which enable them to adsorb different kind of molecules, specially organic and inorganic cations [15,16]. Kaolinite is a 1:1 phyllosilicate consisting of stacked pairs of tetrahedral silica and octahedral alumina sheets. On the contrary, montmorillonite is a 2:1 phyllosilicate having a layered structure consisting of a sandwich of one octahedral alumina sheet between two tetrahedral silica sheets and negative charge is developed on montmorillonite permanently due to isomorphous substitution [17].

There have been a few published reports mentioning the sorptive interactions between triclosan, kaolinite and montmorillonite. The objective of this research was therefore to quantify the triclosan sorption capacity of kaolinite and montmorillonite and compare the result with that of activated carbon. The effects of pH, ionic strength and HA on the sorption phenomenon were studied and the transport and retention mechanisms in the subsurface environment were assessed. Furthermore, sorption isotherms of triclosan were obtained under a wide range of initial pH.

2. Materials and method

2.1. Sorbents and their characterization

The charcoal-based activated carbon in granular form (DC Chemical Co. Ltd., Seoul, Korea) was washed and rinsed with distilled water to remove fines and impurities, dried at 105 °C overnight and stored in a desiccator for further use. Kaolinite (in the form of Kaolin) was obtained from Junsei Chemical Co., Japan and montmorillonite (in the form of Bentonite) was obtained from Yakuri Pure Chemicals Co., Japan. Particle size analysis of activated carbon was performed by sieve analysis, as per ASTM standard specification, and those of kaolinite and montmorillonite were evaluated using a laser particle size analyzer (Malvern, Mastersizer 2000). A Brunauer–Emmett–Teller (BET) surface analyzer (SA 3100, Beckman Coulter) was used to measure nitrogen adsorption at 77.3 K and the surface area of the sorbents (Table 2) was calculated from the isotherms using the BET equation.

2.2. Chemicals and sample preparation

Triclosan (≥97%) was obtained from Fluka (Steinheim, Germany). Triclosan stock solutions were prepared by dissolving 100 mg of triclosan in 100 ml of HPLC grade methanol (SK Chemical,

Ulsan, Korea), stored at 4 °C and used within a month of preparation. The sample solutions for all sorption tests were prepared by spiking the stock solution with 20% methanol (prepared with ultra-pure water) to achieve the desired concentration levels. HA stock solution was prepared by dissolving commercial HA in the form of sodium (HA, CAS: 68131-04-4, Aldrich, St. Louis, MO, USA) in ultra-pure water and stirred for about 24 h. Then the solution was filtered through a 1.2 μm GF/C filter (Whatman®, England) to remove remaining particulate material [19]. The total organic carbon (TOC) content of the solution was measured with a TOC analyzer (TOC-V model, Shimazdu, Kyoto, Japan) and the HA concentrations are expressed as milligrams of organic carbon per liter of water. The solution was stored in the dark at 4 °C until used. The sample solutions containing HA were produced through diluting HA stock solution with ultrapure water to reach organic carbon content of 0–200 mg C L^{-1}. Pure water was served as the dissolved organic carbon (DOC)-free control. NaCl (DC Chemical Co. Ltd., Seoul, Korea) was used to make solutions of various ionic strengths. All chemicals were used without further purification.

2.3. Batch sorption experiments

Batch sorption experiments were conducted in a series of 100 mL glass bottles containing 50 mL of 60 mg L^{-1} triclosan solution. Predetermined concentrations of sorbents (1 g L^{-1}), were added to these bottles, fitted with Teflon-lined rubber septa and sealed air-tight with aluminum caps. All the experiments were performed in the dark (to avoid possible photodegradation of triclosan) in a thermostat controlled shaker (iNtRON Biotechnology Co. Ltd., Korea), maintained at 25 °C and 200 rpm. Based on the results from preliminary kinetic studies, stirring time of 6 and 24 h was selected as equilibrium time for activated carbon and clays, respectively. On completion of the sorption experiments with activated carbon, samples were filtered through 1.2 μm GF/C filters to separate the activated carbon particles from the aqueous phase. During the experiments with clay minerals, the separation of the liquid from the solid phase was achieved by centrifugation at 5000 rpm for 10 min, followed by filtration of the supernatant using 1.2 μm GF/C filters [19]. The triclosan concentration in the filtrate was measured by a high performance liquid chromatography (HPLC) (Ultimate® 3000, Dionex, Sunnyville, CA, USA).

The amount of sorbed triclosan was calculated from the difference between the amount of triclosan initially added to the system and that remained in the solution after equilibration by using the following equation:

$$q_e = \frac{(C_0 - C_e)V}{M} \tag{1}$$

Table 2
Properties of sorbents.

Property	Activated carbon	Kaolinite	Montmorillonite
Average particle size	1.5 mm	6 μm	60 μm
BET surface area (m^2 g^{-1})	738.8	2.31	34.28
PZC[a]	6.88	4.13	2.15

[a] PZC was determined by the method mentioned in Faria et al. [18].

where q_e is the sorption capacity $(mg\,g^{-1})$, C_0 and C_e are the initial and equilibrium concentrations in the solution $(mg\,L^{-1})$, respectively, V is the volume of the solution (L) and M is the mass of sorbents (g).

The effect of pH on triclosan sorption was also investigated in batch mode. The solutions were first prepared by diluting the stock solution using 20% methanol and the pH values (varying from 3 to 10) were adjusted using a few drops of 0.1 M HCl or NaOH. Triclosan concentration, sorbent dosage and the solution temperature were fixed at $60\,mg\,L^{-1}$, $1\,g\,L^{-1}$, and $25\,^{\circ}C$, respectively. For ionic strength and HA effect studies, under similar experimental conditions, NaCl and HA concentrations were varied between 0.001 and 0.5 M and $0–200\,mg\,C\,L^{-1}$, respectively. Equilibrium isotherm experiments were undertaken at $25\,^{\circ}C$ over the triclosan concentration range of $10–60\,mg\,L^{-1}$ and sorbent dosage of $1\,g\,L^{-1}$ over a pH range of 3–10. All the experiments were carried out in duplicate and the mean values were reported. The relative errors of the data were within 5%.

2.4. Chemical analysis

An HPLC system (Ultimate® 3000) (Dionex, Sunnyville, CA, USA) equipped with a Acclaim® C-18 column ($4.6\,mm \times 250\,mm$, $5\,\mu m$) and an UV–vis detector was used to measure the triclosan concentrations at a detection wavelength of 220 nm. The mobile phase used for elution was deionized water (30%) and acetonitrile (70%), which was delivered at $1\,mL\,min^{-1}$ through the column. A sample injection volume of $50\,\mu L$ was used. The column temperature was maintained at $25\,^{\circ}C$. For quantification purposes a calibration plot was performed within the range of experimental concentrations used (coefficient of determination (R^2) greater than 0.99).

3. Results and discussion

3.1. Effect of pH on triclosan sorption

The experimental results showed that the sorption capacity of activated carbon was $\sim 30\,mg\,g^{-1}$ between pH 3 and 6 (Fig. 1a). With an increase in the pH the sorption capacity gradually decreased, reaching a value of $8.8\,mg\,g^{-1}$ at pH 10. In terms of removal efficiency, it was $\sim 60\%$ between pH 3 and 6 and decreased to $\sim 15\%$ at pH 10. Fig. 1b and c shows that the sorption capacities of kaolinite and montmorillonite were 6.4 and $19.3\,mg\,g^{-1}$, respectively at pH 3. As pH was increased to 10, sorption capacities of these clay minerals decreased dramatically to 1.5 and $\sim 3\,mg\,g^{-1}$, respectively. In terms of removal efficiencies, they were 10.5 and 32.3%, respectively for kaolinite and montmorillonite at pH 3 and subsequently decreased to 2.5 and $\sim 5\%$ at pH 10.

Changes in pH affected the dissociation of the triclosan molecule. According to the dissociation constant of triclosan ($pK_a = 8.14$), more than 50% of the triclosan is expected to be deprotonated at $pH > pK_a$. Generally, increase in solution pH results in partially or fully deprotonated surface functional groups and thus a loss of positive charge and/or build up of negative charge [20]. When sorption occurs above pH 9 ($pH > pK_a$), the anionic form of triclosan is dominant in solution [5] and the overall surface charge on the activated carbon is negative when the solution pH is greater than pH_{pzc} (6.88). In this case, the sorption capacity is reduced due to electrostatic repulsion between the deprotonated triclosan and the negatively charged activated carbon surface. On the other hand, at acidic pH, the net charge on activated carbon surface is positive and triclosan is mainly non-dissociated. Thus, repulsive electrostatic interactions are minimized and sorption is enhanced. With respect to the clay minerals, as the surface functional groups are deprotonated at $pH > 5$ as indicated by the pH_{pzc} of kaolinite (4.13),
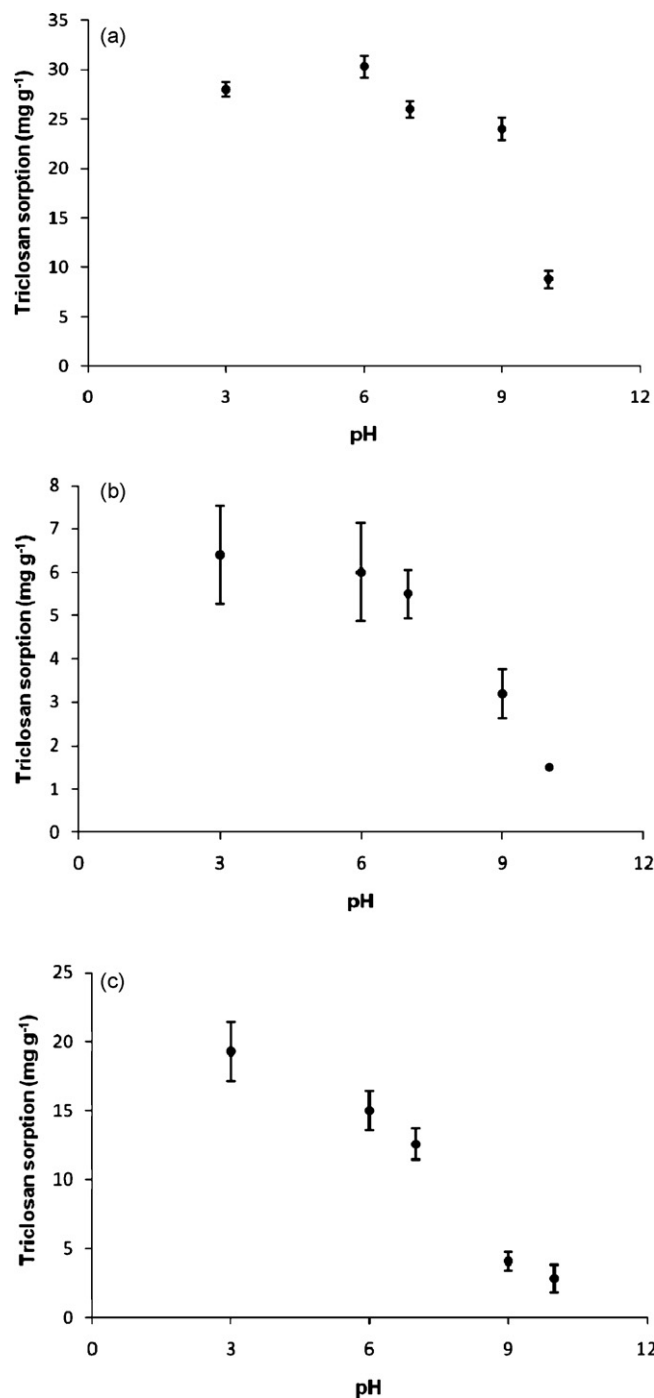

Fig. 1. Effect of solution pH on triclosan sorption onto (a) activated carbon, (b) kaolinite, and (c) montmorillonite ($C_0 = 60\,mg\,L^{-1}$, sorbent dose = $1\,g\,L^{-1}$). Error bars indicate one standard deviation.

and at $pH > 3$ as indicated by the pH_{pzc} of montmorillonite (2.15), an enhanced electrostatic repulsion between negatively charged triclosan and the clay surfaces is responsible for the significant reduction in triclosan sorption. Thus, as expected, triclosan sorption capacities of these clay minerals decreased when solution pH was increased from strongly acidic to weakly alkaline conditions. Based on these results, pH_{pzc} seems to be an excellent marker of triclosan removal from aqueous solution.

Furthermore, it is evident from these results that the extent of triclosan sorption onto kaolinite and montmorillonite is far less than that onto the activated carbon, which can be attributed

primarily to their lower surface areas in comparison to the activated carbon (Table 2). The sorption edges indicated a more favorable interaction of the fully protonated triclosan molecule with montmorillonite than kaolinite. Greater triclosan sorption to montmorillonite than to kaolinite was observed in all pH values. On a mass basis, triclosan sorption capacities of montmorillonite were 2–3 times higher than kaolinite. This could be attributed to the higher surface area ($34.28\,m^2\,g^{-1}$) and organic matter content (7.8%) of montmorillonite compared to those of kaolinite ($2.31\,m^2\,g^{-1}$ and 3.1%, respectively). The differences in magnitude of triclosan interactions with these two clays indicate that clay type must be considered when estimating triclosan interactions with soil clay components.

3.2. Effect of ionic strength on triclosan sorption

As shown in Fig. 2a, triclosan sorption onto activated carbon was almost constant up to ionic strength of 1×10^{-1} M and, there was an increase in sorption from 37.54 to 44.88 mg/g when ionic strength was increased from 1×10^{-1} to 5×10^{-1} M at pH 3. Overall, the net increase in triclosan sorption was not that significant (only 1.2 times) with the ionic strength increase from 1×10^{-3} to 5×10^{-1} M. From the investigation of the influence of ionic strength on triclosan sorption onto kaolinite and montmorillonite, it was observed that there is almost no change in the sorption of triclosan on to kaolinite and montmorillonite up to an ionic strength of 5×10^{-2} M (Fig. 2b and c). However, with the increase in ionic strength from 5×10^{-2} to 5×10^{-1} M, there was a significant increase in the sorption of triclosan on to kaolinite (from 5 to 24.66 mg g^{-1}) and montmorillonite (from 10.6 to 30.2 mg g^{-1}) at pH 3. Overall increase in sorption was ~4 times and 3.5 times, respectively for kaolinite and montmorillonite with increasing ionic strength from 1×10^{-3} to 5×10^{-1} M.

Theoretically, sorbent particles and triclosan molecules are both surrounded by an electric double layer due to electrostatic interactions [21]. Based on the Gouy–Chapman theory of the diffuse double layer, the thickness of the double layer is compressed by an increase in the ionic strength of the solution [22]. At higher ionic strength, the sorption of triclosan will be high owing to the partial neutralization of the positive charge on the sorbent surface and a consequent compression of the electrical double layer by the chloride ion. The chloride ion can also enhance the sorption of triclosan ion by pairing their charges, and hence reducing the repulsion between the triclosan molecules sorbed on the surface. This initiates the sorbent to sorb more positive triclosan ions. A similar phenomenon was observed on the sorption of an herbicide onto activated carbon [23]. The increased sorption of triclosan onto clay minerals with increase in ionic strength could also be due to the decrease in solubility of triclosan in salt solution, which is characterized as 'salting out' effect [24].

Irrespective of the increase in ionic strength, triclosan sorption onto all the sorbents was almost constant at pH 9. However, at pH 3, sorption of triclosan onto these sorbents increased significantly at higher ionic strengths. This indicates that at pH 3, the effect of pH was predominant compared to the effect of ionic strength. Despite the increase in ionic strength at pH 9, the constancy in sorption capacity could be due to the fact that at pH 9 the anionic form of triclosan (88%) is dominant in solution and the overall surface charge on all the sorbents is negative. This led to very high electrostatic repulsion between the deprotonated triclosan and the negatively charged sorbent surfaces, and was most likely prevailed over the effect of ionic strength at this pH.

3.3. Effect of HA on triclosan sorption

To examine the effect of natural organic matter (NOM) on the triclosan sorption, HA was selected as the model NOM, since
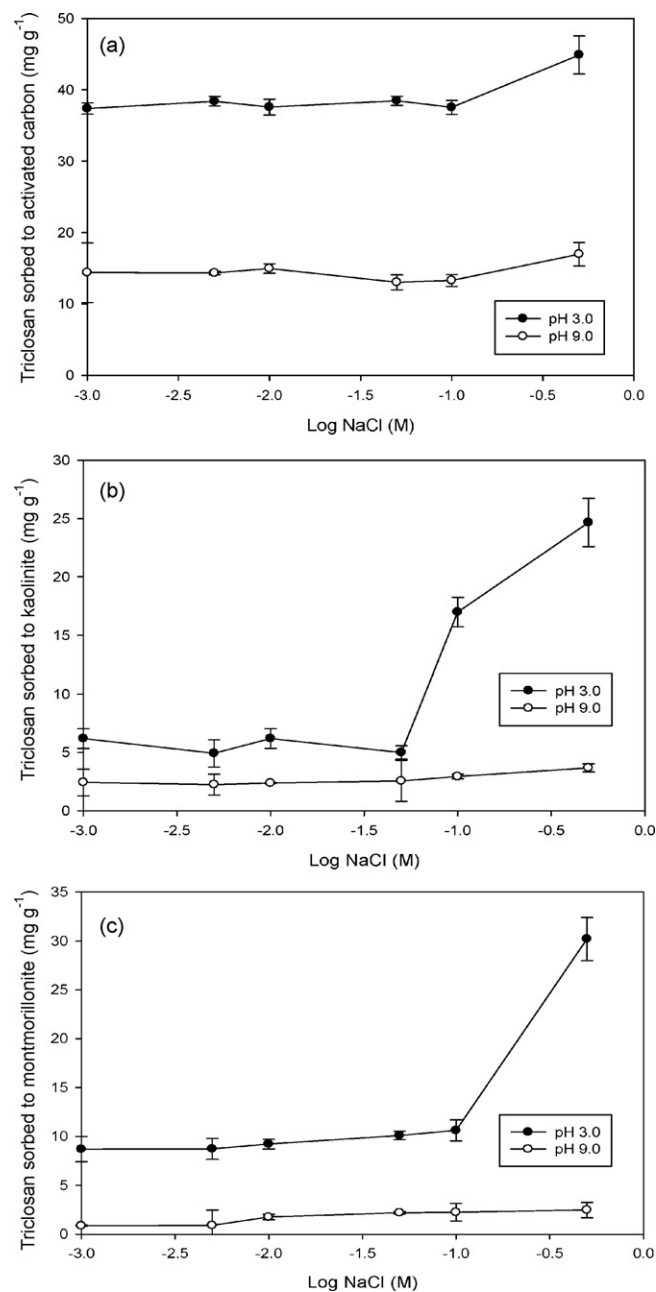

Fig. 2. Effect of ionic strength on triclosan sorption onto (a) activated carbon, (b) kaolinite, and (c) montmorillonite ($C_0 = 60\,mg\,L^{-1}$, sorbent dose = 1 g L^{-1}, pH 3 and 9). Error bars indicate one standard deviation.

NOM constitutes about 70% of HA [25]. As shown in Fig. 3a, triclosan sorption onto activated carbon decreased from 31.4 to 28.1 mg g^{-1}, when HA concentration was increased to 10 mg C L^{-1}. Further increase in HA concentration up to 40 mg C L^{-1} reduced the sorption capacity to 14.87 mg g^{-1}. However, there was negligible effect on the triclosan sorption when the HA concentration was increased from 40 to 200 mg C L^{-1}. Overall, there was a significant decrease in triclosan sorption by 20.82 mg g^{-1} with an increase in HA concentration up to 200 mg C L^{-1}. Pore blockage and direct site competition are considered to be the most common mechanisms that would have affected activated carbon sorption in the presence of NOM. In yet another study pertaining to the effect of NOM on adsorption of agricultural organic chemicals onto activated carbon, pore blockage was shown as the major contributing factor
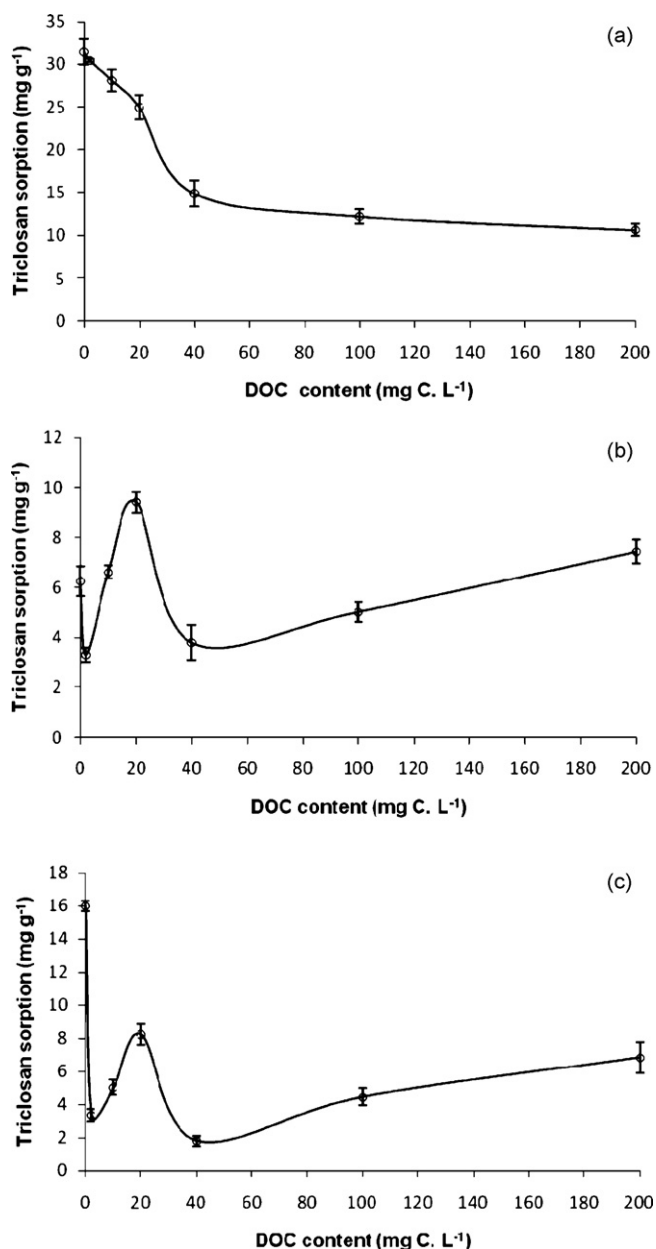
Fig. 3. Effect of humic acid on triclosan sorption onto (a) activated carbon, (b) kaolinite, and (c) montmorillonite (C_0 = 60 mg L^{-1}, sorbent dose = 1 g L^{-1}, pH 6). Error bars indicate one standard deviation.

compared to the competitive adsorption effect between NOM and smaller target molecules [26].

At HA concentration up to 2 mg C L^{-1}, triclosan sorption onto kaolinite decreased from 6.2 to 3.3 mg g^{-1} and onto montmorillonite decreased from 15.7 to 3.3 mg g^{-1} (Fig. 3b and c), presumably due to instantaneous complexation of triclosan with HA in aqueous phase as HA was introduced [27,28]. In the 2–20 mg C L^{-1} HA concentration range, an increase in the triclosan sorption (3.3–9.4 and 3.4–8.3 mg g^{-1}, respectively in kaolinite and montmorillonite) was observed, while higher concentration of HA (40 mg C L^{-1}) resulted in an opposite effect. The increased sorption in the 2–20 mg C L^{-1} HA concentration range is most probably due to (i) sorption of HA-complexed triclosan onto clay minerals and (ii) sorption of free triclosan to HA sorbed to clay surface. Control experiments showed that HA can be sorbed to clay surface regardless of triclosan concentration (data not shown). As previously reported [19], it was clearly evident that the increased amount of HA at clay surface can result

in enhancing the sorption of pharmaceuticals via hydrophobicity. Anew, experiments performed with HA sorbed clays indicated that significant amount of triclosan can be further sorbed (data not shown). On the other hand, the decreased triclosan sorption in the 20–40 mg C L^{-1} HA concentration range may be explained by re-partitioning of triclosan between clay-associated HA and HA in the aqueous phase. With an increase in the HA concentration in aqueous phase, the amount of HA-complexed triclosan also gradually increased, which resulted in the decrease of free triclosan in the aqueous phase. Therefore, more triclosan could be desorbed from clay surface [29].

Further increase in HA concentration from 40 to 200 mg C L^{-1} increased the sorption of HA-complexed triclosan again to 7.4 and 6.9 mg g^{-1} in kaolinite and montmorillonite, respectively. This increase is due to the fact that HA sorption onto both kaolinite and montmorillonite increased very sharply with an increase in the HA concentration from 40 to 200 mg C L^{-1} as confirmed from additional experiments (data not shown). Thus, the HA-complexed triclosan is sorbed to clay surface as well as the HA associated with clay. As compared to kaolinite, the higher sorptivity of montmorillonite for triclosan may tentatively be attributed to its higher surface area (Table 2) and higher organic matter content.

3.4. Triclosan sorption isotherm at various pH values

Two well known models, namely the Langmuir and Freundlich models, were used to analyze the experimental data. A linearized form of the Langmuir model is [30]:

$$\frac{1}{q_e} = \frac{1}{K_L Q_o}\frac{1}{C_e} + \frac{1}{Q_o}, \tag{2}$$

where Q_o is a constant related to the area occupied by a monolayer of sorbate, reflecting the maximum sorption capacity (mg g^{-1}), C_e is the equilibrium liquid-phase concentration (mg L^{-1}), K_L is a direct measure of the intensity of sorption (L mg^{-1}) and q_e is the amount sorbed at equilibrium (mg g^{-1}).

A linearized form of Freundlich model is [30]:

$$\log q_e = \log K_F + \frac{1}{n}\log C_e, \tag{3}$$

where K_F ((mg/g)(L/mg)$^{1/n}$) and n (dimensionless) are constants incorporating all factors affecting the adsorption process such as adsorption capacity and intensity, respectively. The dependence of the triclosan uptake (q_e) on the equilibrium concentration (C_e) in aqueous solution and the Langmuir and Freundlich isotherm plots of triclosan sorption onto activated carbon, kaolinite and montmorillonite at 25 °C and pH 3 are shown in Figs. 4–6, respectively. The experimental q_e versus C_e data have been fitted to Langmuir and Freundlich models and the isotherm parameters were determined. The plots of experimental values of $1/q_e$ versus $1/C_e$ (Langmuir model), and $\ln q_e$ versus $\ln C_e$ (Freundlich model), at different pHs, were made and the best-fit values of parameters were determined using MS Excel for Windows. The values of the isotherm constants, and the correlation coefficient, R^2 for the best-fit of the equations are listed in Tables 3–5, respectively for activated carbon, kaolinite and montmorillonite. The applicability of the isotherm models was compared on the basis of the values of regression coefficients (R^2) that are regarded as a measure of the quality of fit of experimental data on the isotherm models [31]. The R^2 value near unity indicates an excellent fit to the isotherm equation and near zero indicates a very poor fit. The triclosan sorption experimental data on activated carbon fit better to the Langmuir isotherm model as the regression coefficients of the Langmuir plots are always higher than those obtained for the fitting of the Freundlich isotherm model in all the pHs. Both Langmuir and Freundlich isotherm models are somewhat consistent in indicating the sorption affinity of kaolinite
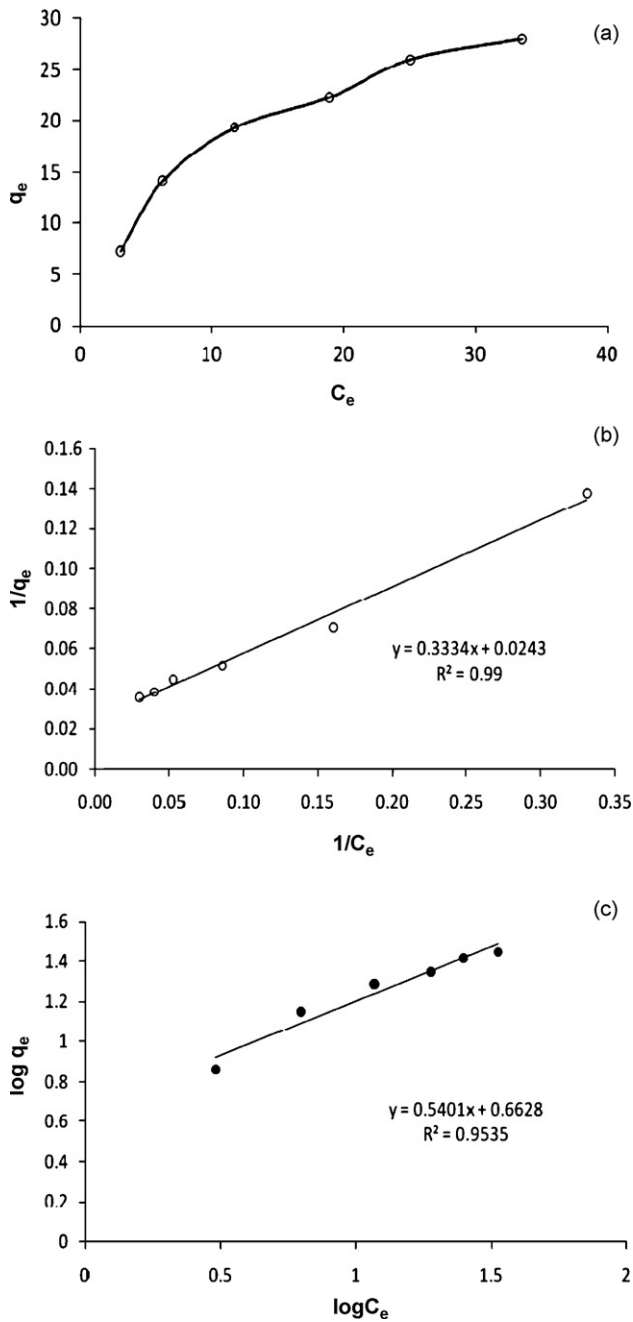
Fig. 4. Plots of C_e versus q_e (a), Langmuir (b) and Freundlich (c) isotherms of triclosan sorption onto activated carbon at pH 3.
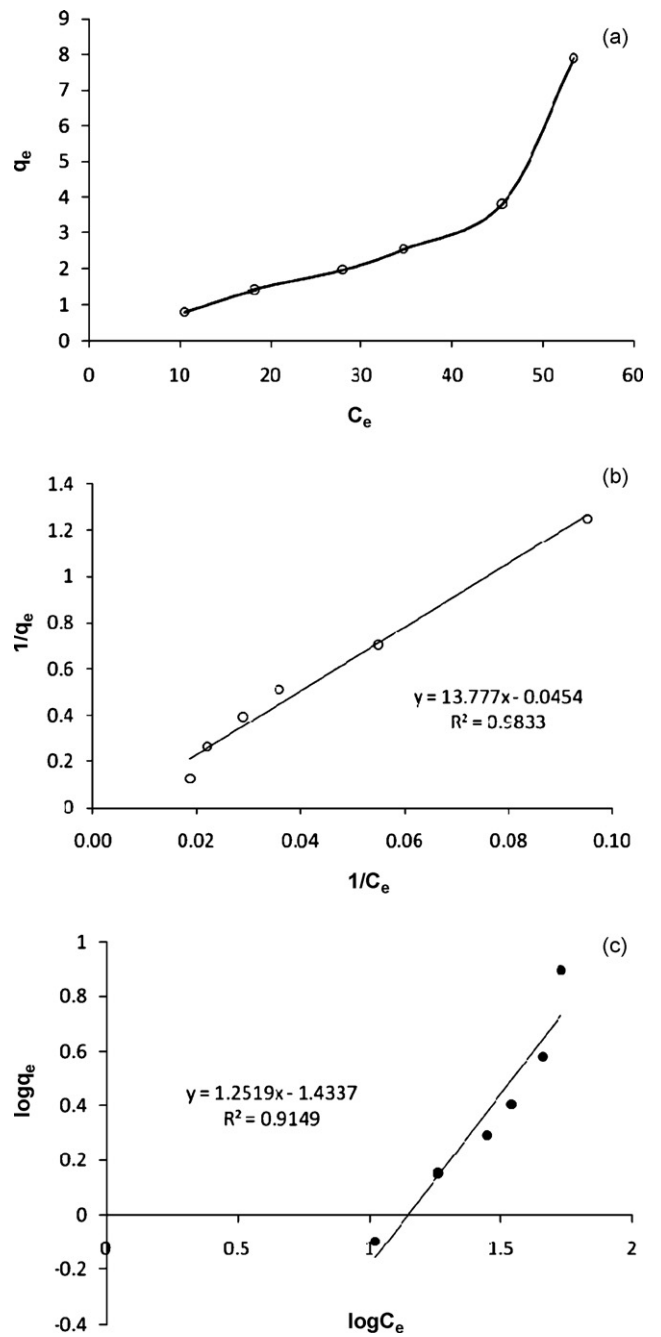

Fig. 5. Plots of C_e versus q_e (a), Langmuir (b) and Freundlich (c) isotherms of triclosan sorption onto kaolinite at pH 3.

Table 3
Calculated isotherm parameters for Langmuir and Freundlich models for sorption of triclosan onto activated carbon at various pHs.

pH	Langmuir isotherm			Freundlich isotherm		
	Q_0 (mg g^{-1})	K_L (L mg^{-1})	R^2	K_F ((mg/g)(L/mg)$^{1/n}$)	n	R^2
3	41.15	0.073	0.99	4.6	1.85	0.95
6	70.42	0.011	0.93	1.13	1.1	0.88
7	67.11	0.023	0.92	3.03	1.61	0.77
9	36.5	0.052	0.97	2.76	1.58	0.93
10	18.5	0.062	0.94	1.68	1.7	0.92

Table 4
Calculated isotherm parameters of Langmuir and Freundlich models for sorption of triclosan onto kaolinite at various pHs.

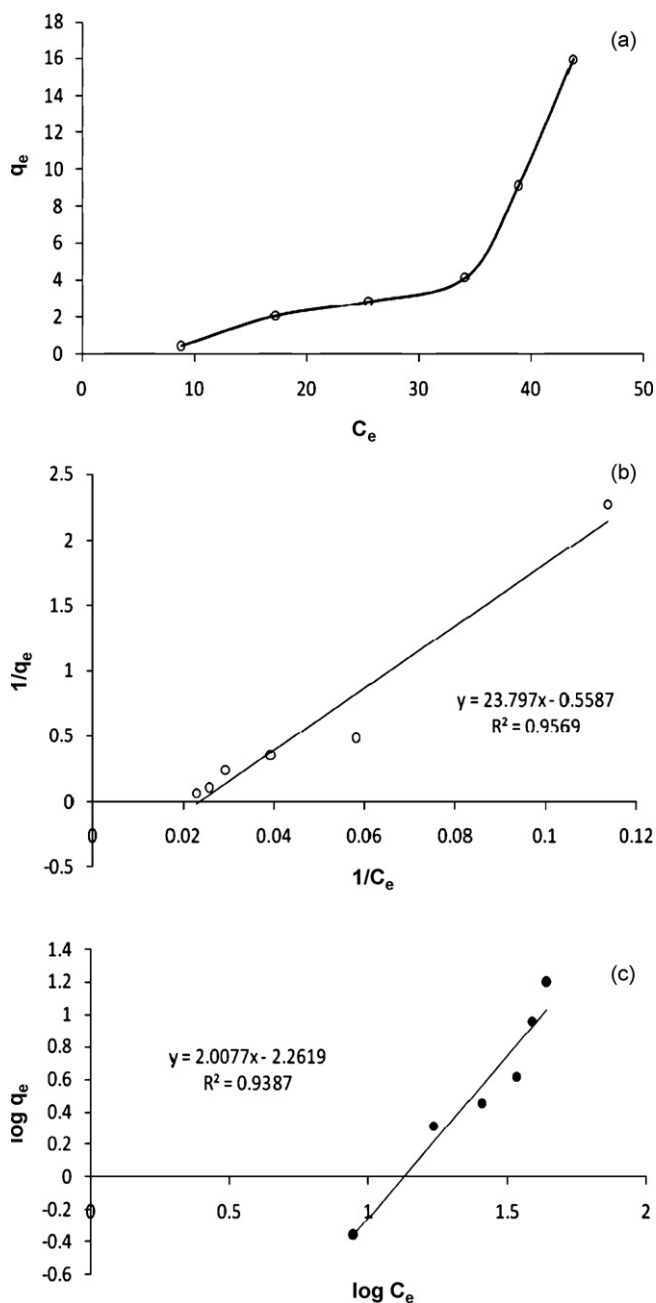| pH	Langmuir isotherm			Freundlich isotherm		
	Q_0 (mg g^{-1})	K_L (L mg^{-1})	R^2	K_F ((mg/g)(L/mg)$^{1/n}$)	n	R^2
3	22.02	0.003	0.98	27.14	0.79	0.91
6	7.73	0.009	0.99	35.08	0.77	0.96
7	6.03	0.006	0.96	21.9	0.8	0.96
9	5.27	0.011	0.99	20.7	0.83	0.96
10	4.95	0.01	0.9	17.09	0.89	0.89

Fig. 6. Plots of C_e versus q_e (a), Langmuir (b) and Freundlich (c) isotherms of triclosan sorption onto montmorillonite at pH 3.

and montmorillonite as revealed from the regression coefficients in all the pHs. However, the convex upward nature of the isotherms (Figs. 5a and 6a) clearly indicates that they can be better fitted by the Freundlich model [32].

Table 5
Calculated isotherm parameters of Langmuir and Freundlich models for sorption of triclosan onto montmorillonite at various pHs.

pH	Langmuir isotherm			Freundlich isotherm		
	Q_0 (mg g^{-1})	K_L (L mg^{-1})	R^2	K_F ((mg/g)(L/mg)$^{1/n}$)	n	R^2
3	3.3	0.012	0.96	2962.8	0.43	0.93
6	2.4	0.012	0.98	559.8	0.52	0.93
7	1.79	0.023	0.99	182.76	0.5	0.97
9	0.42	0.018	0.98	63.27	0.7	0.98
10	0.24	0.025	0.86	54.14	0.76	0.81

4. Conclusion

Experimental results indicated that the sorption capacity of activated carbon, kaolinite and montmorillonite for triclosan removal was higher under acidic rather than in alkaline conditions. At higher and lower pH values, proton activity has a pronounced affect on sorption as the anionic and cationic species assume greater importance to overall sorption. The results further revealed that, irrespective of the types of sorbents, at lower pH, sorption capacity increased with an increase in the ionic strength of solution. The presence of HA at elevated concentrations reduced the sorption of triclosan onto activated carbon. However, during the triclosan sorption onto clays HA showed much greater affinity for triclosan than clay indicating that organic matter content could be the major factor controlling the sorption of triclosan in soil.

Acknowledgement

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