PROJECT REPORT

The Effects of Iron Oxide Nanoparticles on the Adsorption of TCE by PAC in Water Treatment

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The Effects of Iron Oxide Nanoparticles on the Adsorption of TCE by PAC in Water Treatment

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ABSTRACT

Current best available technology (BAT) for the removal of volatile organic compounds (VOCs) during drinking water treatment is the use of activated carbon. The presence of nanoparticles, as well as the presence of natural organic matter (NOM), may adversely affect the adsorption of VOCs onto activated carbon. This research focuses on the potentially inhibiting effects of humic acid (HA), a NOM, and nanoparticulate iron oxide on the adsorption of TCE by powdered activated carbon (PAC). Six isotherms, prepared using various combinations of TCE, PAC, HA, and Fe_2O_3, were allowed to equilibrate for fourteen days, were filtered, diluted and then analyzed for TCE concentration using GC-FID. Results indicate that the presence of nanoparticulate Fe_2O_3 increases removal of TCE from solution and suggests a direct relationship between nanoparticulate concentration and TCE removal over a range of PAC concentrations. The presence of HA was found to decrease the adsorption of TCE onto PAC. In addition to these comparative studies, Freundlich constants (k and 1/n) were calculated for each combination, providing a mechanism for calculating the amount of PAC required to achieve a target reduction in TCE concentration. Information generated from this study may be utilized by drinking water treatment operators in redesigning facilities to ensure effective removal of TCE and other VOCs.

Key Words: isotherm, NOM, nanoparticles, VOC, Humic Acid

1. INTRODUCTION

Concerns with the production of disinfection byproducts via the interaction of organic material with chlorine during drinking water management (USEPA 2009), has prompted some water companies to implement activated carbon treatment. Activated carbon (AC) is widely accepted as the best available technology for adsorbing remaining organic contaminants after sedimentation and filtration (Russell et al. 1992). Contaminants are trapped in the pores of the carbon matrix, preventing them from being distributed to the consumer. Consequently, materials that interfere with or bypass the carbon treatment process merit investigation. Two materials of concern are nanoparticles and natural organic matter.

The rapid growth of nanotechnology has been both predicted and realized in the last two decades (Rao and Cheetham 2001). Emergence of nanoscience in scores of industrial and scientific developments has generated a concern for environmental impact as an immeasurable quantity of nanoparticles (NPs) is released into natural resources. Although a large percentage of these particles are released accidently via spills in transit between manufacturing facilities and production sites, as well as through general degradation of materials, some NPs, including iron oxide nanoparticles are released by design to remediate contaminated sediment, soils, and groundwater (Li et al. 2006). While this practice is widespread in the US, the UK has placed a moratorium on the use of this use of NPs because of unknown potential environmental effects (Klaine 2006).

One potential impact of the release of NPs is their effect on the treatment of drinking water. In particular, NPs may compete with the adsorption ability of activated carbon, which is used to remove volatile organic compounds (VOCs) during the drinking water treatment process.

The principle mechanism by which contaminants are captured onto AC is electrostatic charge. The large surface area of AC, an average of 900 cm^2 per gram for granular activated carbon (GAC), allows AC to attract and adsorb large quantities of oppositely-charged particles. The charge on AC depends on the pH of its surroundings and, if placed into an aqueous solution, the solution's ionic strength (Letterman 1999). Fig. 1 shows the pH-charge relationship. Similarly, the surface charge of some NPs changes with surrounding pH and ionic strength (see Table 1.) Thus coagulation, flocculation and sedimentation processes cause some percentage of NPs to aggregate and settle out. Consequently, NPs may be attached onto AC, competing with target contaminants for adsorption sites or may work as adsorption sites themselves for contaminants. The first scenario may result in incomplete removal of target contaminants (usually VOCs), given a concentration of AC and specific contact time, while the second scenario may allow VOCs to bypass AC treatment regardless of operational parameters.
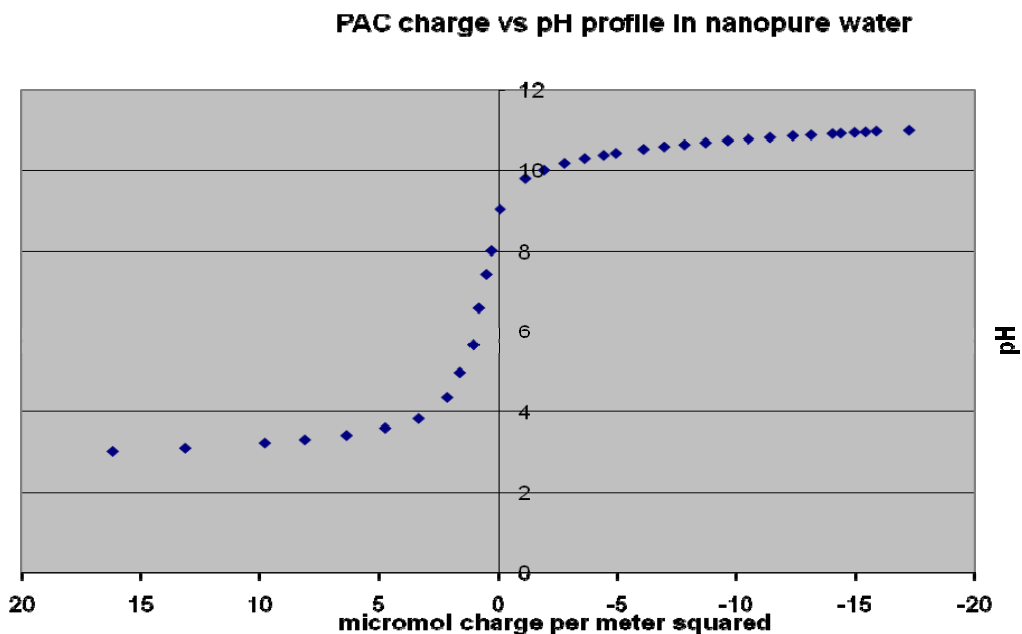

Figure 1. Charge vs. pH for PAC

Table 1. Zeta Potential and pHpzc (pH of Point Zero Charge) of the NPs and the PAC

Parameter	pHpzc (0.001MKH$_2$PO$_4$)	pHpzc (0.001M KH$_2$PO$_4$ +5mg/l Humic Acid)	Charge (millivolts) (0.001MKH$_2$PO$_4$)	Charge (0.001M KH$_2$PO$_4$ +5mg/l Humic Acid)
SiO$_2$	1.9	<1.8	pH 7 (-37.9)	pH 7 (-35.0)
TiO$_2$	2.4	< 1.8	pH 7 (-39.4)	pH 7 (-45.3)
Fe$_2$O$_3$	7.7	< 1.8	pH 7 (+17.8)	pH 3.5 (-28.0)
PAC	3.2	2.5	pH 7 (+28.8)	pH 7 (-41.3)

Natural organic matter (NOM) also interferes with adsorption of organic compounds onto AC. 10 Large molecular weight NOM physically blocks VOCs from contacting AC, while low molecular weight NOM fills adsorption sites. NOM also may change the electrostatic charge on nanoparticles so that they will not settle out during sedimentation processes associated with water treatment (refer to Table 1).

2. GOALS AND OBJECTIVES

This research focused on the effects of nanoparticles and NOM on the adsorptive capacity of carbon used for the removal of volatile organic compounds (VOC). In particular, we looked at the effect of nanoparticulate iron oxide in the presence and absence of humic acid, a low molecular weight form of NOM, on the adsorption of trichloroethylene, TCE, onto powdered activated carbon (PAC). TCE was chosen as the VOC of interest due to its presence as a groundwater contaminant (Chen and Wu 2000). Secondary objectives were to calculate Freundlich isotherm constants for all TCE, PAC, NP and HA combinations and generate data on the kinetics of TCE adsorption onto PAC. The project also was designed to provide middle and high school teachers with research experience.

3. RESEARCH STUDY DETAILS

3.1 Stock Solution Preparation

25000 mg/L TCE and 60 mg/L PCE stock solutions were prepared by mixing reagent grade chemicals in methanol. The solutions were capped with Teflon septa lids and stored in a refrigerator.

3.2 Calibration Curve Preparation

An eight-point calibration curve was prepared by diluting TCE stock solution with autoclaved nano-pure water, which had been buffered with 0.001 M KH_2PO_4 and adjusted to pH 7 with 10N NaOH. PCE was also added as an internal standard. Samples were analyzed using an

Archon 5100 Purge & Trap Autosampler connection to a Tekmar 3000 Purge & Trap

Concentrator. The purge and trap unit was interfaced to a Hewlett Packard 6890 gas

chromatograph (GC) equipped with a flame ionization detector (FID).   An Agilent DB-WAX

column (J&W 123-7063) (60 m X 320 micrometers inside diameter with 0.50 micrometer film

thickness was used.) The flow rate of the helium carrier gas was set at 2.5 milliliters per minute

(mL/min). The detector makeup gas flow rate was set at 30.0 mL/min. The flow rates of

hydrogen and air flame gases were set at 35.0 mL/min and 400.0 mL/min, respectively. The

retention times for TCE and PCE were 10.4 min and 11.2 min, respectively. Results were

plotted using Microsoft Excel and appear as Fig. 1. For the line of best fit the R^2 value was 0.99.
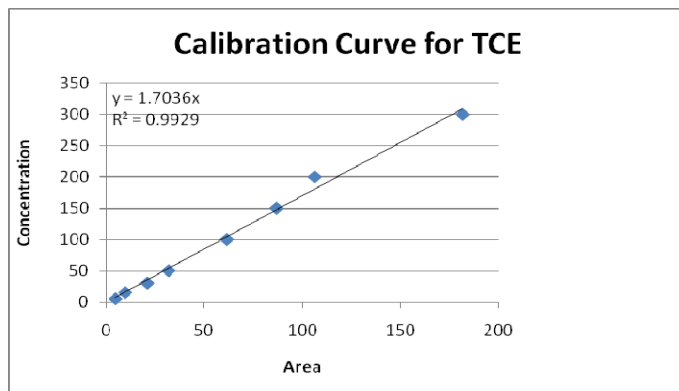

Figure 2. Calibration Curve for TCE Analysis

3.3 Carbon Preparation

Prior to the beginning of the RET project, PAC was prepared by rinsing granular

activated carbon several times with nano-pure water to remove the fines, drying in an oven at

105o C for two days to remove any moisture, crushing in a carbon mill and sieving using a 120

mesh sieve. This size fraction was then stored in a desiccator until use.

3.4 Isotherm Preparation

Samples designed to show the effects of iron oxide nanoparticles and humic acid on the adsorption of TCE onto PAC were prepared using the combinations shown in Table 2.

Table 2. Isotherm Sample Combinations

	TCE	PAC	HA	Fe_2O_3
Baseline1	✓	✓		
Baseline 2	✓	✓	✓	
Set A	✓	✓		1.0 mg/L
Set B	✓	✓		0.5 mg/L
Set C	✓	✓	✓	1.0 mg/L
Set D	✓	✓	✓	0.5 mg/L

For each combination, four liters of autoclaved nano-pure water, which had been buffered with 0.01M KH_2PO_4, and the pH adjusted to 7.0 with 10N NaOH was prepared. For Sets "Baseline 2", "Set C" and "Set D" humic acid was added to make a concentration of 10 mg/L. For "Set A" through "Set D" nanoparticles size Fe_2O_3 was added to achieve the concentrations listed in Table 2. The augmented water was stirred, using a magnetic stirrer, for 30 minutes.

For each combination, 300 µL of TCE stock solution were introduced to the 4-liter glass dispensing jar, nitrogen gas was introduced into the headspace to prevent volatilization of TCE and the solution was mixed for 30 minutes. Accurately weighed (± 0.2 mg) masses of PAC were placed in 250-mL glass amber bottles (see Table 3.) The bottles were then completely filled with solution sealed with Teflon-lined caps and covered with Parafilm. Each set of bottles was

accompanied by a minimum of two blanks to check for any TCE volatilization, adsorption onto

the walls of the bottles, and biodegradation. The bottles were then allowed to equilibrate in a

rotary tumbler for a period of 14 days.

Table 3. Mass of PAC Added to Each Bottle

Bottle Number	Mass of PAC in mg ($\pm$ 0.2 mg)
1	1.7
2	2.7
3	3.9
4	5.2
5	6.9
6	9.7
7	12.5
8	19.0

3.5 Sample Analysis

After fourteen days, sample bottles were removed from the tumbler. An aliquot of each

bottle was filtered using pre-rinsed 0.45 nanometer filter paper. The samples were diluted (to

yield results in the range of the calibration curve) in 50 ml volumetric flasks and an internal

standard of 25 µL of PCE was added to each vial. The vials were sealed with Teflon septum

caps and covered with Parafilm. The samples were analyzed for TCE and PCE concentrations

using the instrumentation described above. Data were plotted using Excel and Sigma Plot

software programs.

3.6 TCE Adsorption Kinetics Study

Kinetic studies were conducted in order to observe the rate of TCE adsorption over time.

Two kinetic studies using nanoparticulate TiO_2 and SiO_2 were conducted using 20 bottles for

each nanomaterial considered. Each bottle contained target initial concentrations of TCE of

1800µg/L, 0.015 mg of PAC. The nanoparticles concentrations were 0.5 mg/L, or 1.0 mg/l for TiO$_2$ and 5.0 and 10.0 mg/L for SiO$_2$. All bottles were buffered to a pH of 7.0 ± 0.2. The bottles were continuously tumbled. Sacrificial bottles were removed from the tumbler every 20 minutes initially for a period of 3 hours then after 5 hours, and followed by 10 hours.. Samples were taken from the sacrificed bottle, filtered, diluted, and analyzed in the GC.

4. RESULTS

The concentration of TCE present in each sample (C$_e$) was calculated using Equation 1.

Equation 1. **C$_e$ = A * S * D**

where: C$_e$ is the concentration of TCE in µg/L

   A is the area of the TCE peak

   S is the slope calculated for the calibration curve (1.7 for the experiments)

   D is the dilution factor for the sample

TCE removal per gram of PAC was calculated using Equation 2.

Equation 2. **q$_e$ = (B - C$_e$) * 0.25 / M**

where: q$_e$ is the TCE, in µg, removed per gram of PAC

   B is the mean concentration of TCE, in µg/L, reported in the blanks for the combination

   M is the mass of PAC, in grams, for the sample

   0.25 is the correction for using 250 mL sample bottles

PCE concentrations (the internal standard added to assess the accuracy of GC-FID results) were erratic. Review of procedures for addition of the PCE to samples suggests that significant

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PCE may have volatilized during the addition process. Analysis of TCE in water samples spiked with PCE before and after the analyses performed for this study indicate that the GC-FID was operating within normal parameters. Consequently, TCE areas on the chromatographs were not corrected for PCE response.

Figure 3 shows the relationship between the sample conditions and the amount of TCE removed from solution per gram of PAC. Data for each combination is presented in Appendix III.
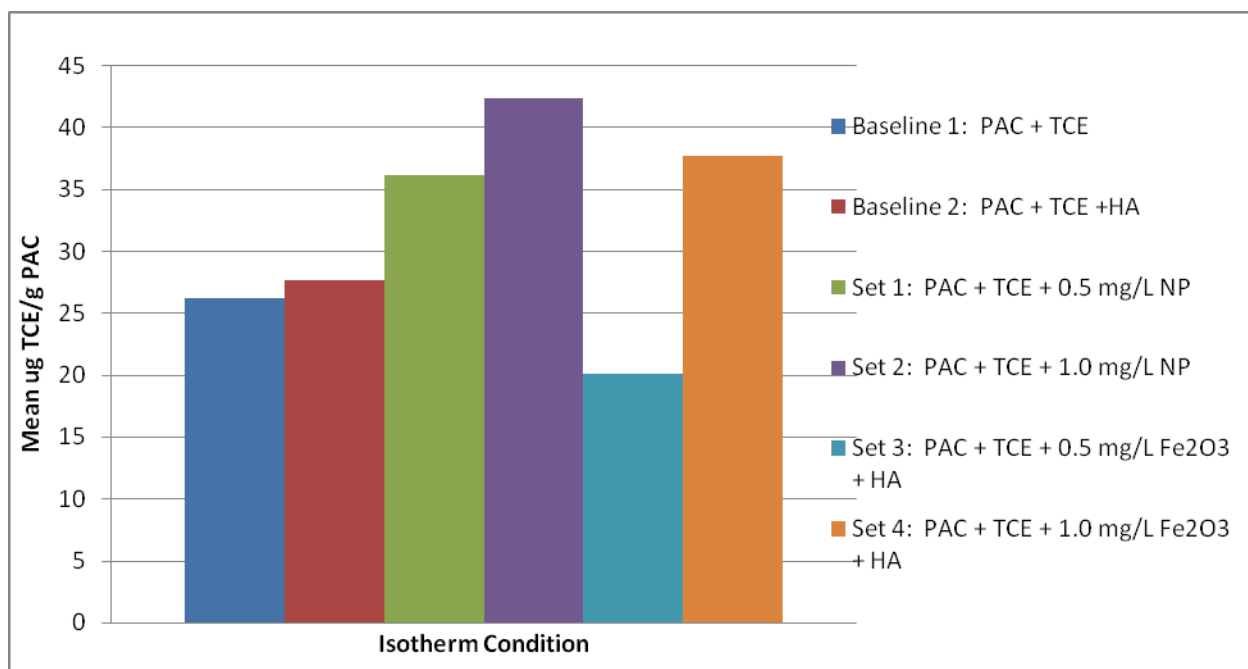

Figure 3. Average Amounts of TCE Removed per Gram of PAC for Each Condition

The log values of C_e and q_e were plotted for each combination. Figure 4 shows the plotted data for all combinations.
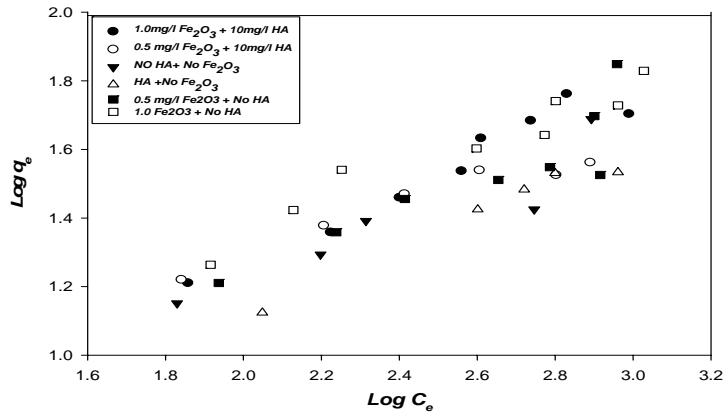
Figure 4. Adsorption of TCE in the Presence and Absence of Fe_2O_3 and HA

Table 4 lists regression analysis results for each combination as calculated by Excel. The Freundlich equation constant for each combination was calculated by solving equation 3 for k.

Equation 3. $Q_e = k * C_e^{1/n}$

where: k and 1/n are Freundlich equation constants

Table 4. Freundlich Isotherm Constants

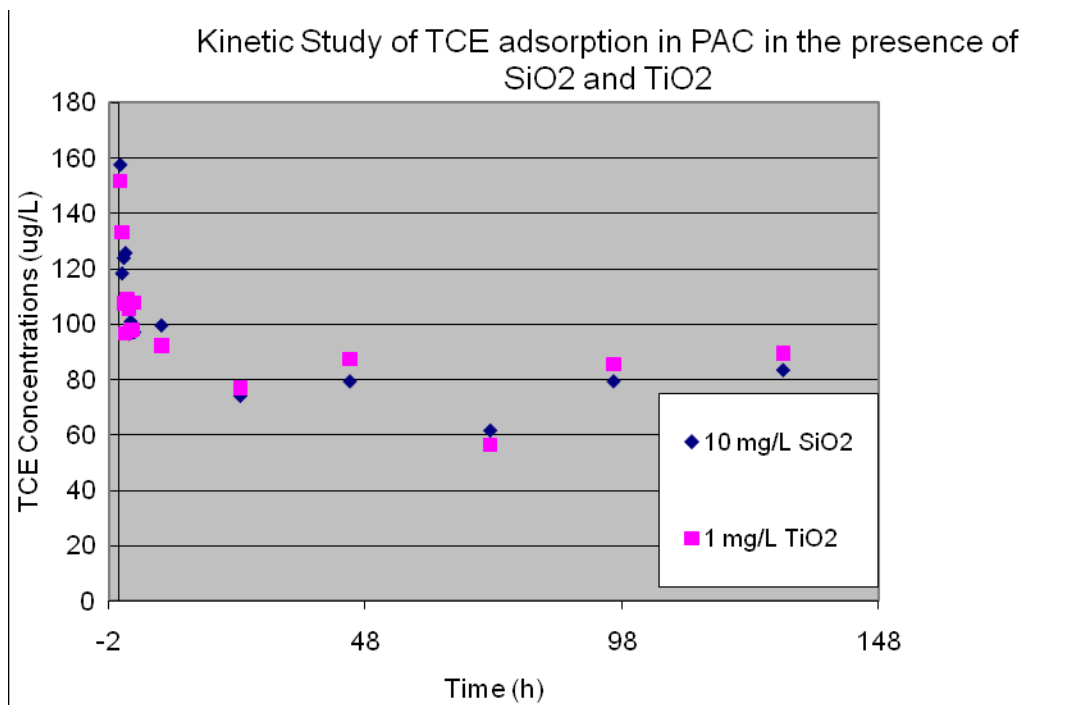| Isotherm Sample Combination	k	1/n	R^2
Baseline1	0.43	0.37	0.86
Baseline 2	0.48	0.25	0.97
Set A	0.47	0.29	0.80
Set B	0.43	0.49	0.93
Set C	0.22	0.79	0.72
Set D	0.51	0.25	0.94

Figure 5. Results of the TCE adsorption kinetics study.

5. DISCUSSION

Results of the TCE adsorption kinetics study indicate that the majority of TCE adsorption occurs onto PAC within the first 20 minutes. The implication of these results is that contact time between TCE-laden water and PAC did not influence the TCE concentrations removed during the study. Current contact times used to treat water with granular activated carbon prior to public distribution (approximately 15 minutes), therefore seems sufficient.

Referring to Fig. 3, the "Baseline 1" condition approximates the average µg of TCE adsorbed onto each gram of PAC. The increased removal of TCE from solutions in "Set A" and "Set B" is consistent with the expectation that nanoparticulate Fe_2O_3 would act as a TCE adsorption site,

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which would complement/compete with PAC for TCE removal. The results indicate that TCE-laden nanoparticles were captured by the 0.45 µm filter, resulting in an overall higher removal of TCE. What is not known is whether the amount adsorbed to the nanoparticles is comprised of only the amount beyond where the PAC becomes saturated (considered to be complementary to PAC removal), or the presence of nanoparticles prevented some PAC adsorption of TCE, which was masked by adsorption of TCE onto the particles (i.e., competition with PAC removal.) Investigation of this question was beyond the scope of this research, but the large difference in surface area between PAC and Fe_2O_3 nanoparticles (900 m^2/g versus 35 m^2/g) suggests complementary TCE removal. The implications of the first scenario for water treatment are that using sufficient amounts of activated carbon to remove TCE to levels below regulatory limits would render the presence of nanoparticulate iron oxide a non-issue. If the second scenario is correct, then some amount of TCE will pass through drinking water treatment systems, regardless of the amount of activated carbon and contact time used. Only nanofiltration, or possibly the use of magnetism to remove TCE-laden Fe_2O_3, would lower "soluble" TCE concentrations.

The presence of low molecular weight NOM, such as humic acid, lowers the adsorption of TCE onto activated carbon either by competing for PAC adsorption sites or blocking access to the carbon. The causal mechanism for the increased TCE removal in the "Baseline 2" samples is unclear. It may be due to insufficient mixing of HA resulting in lower concentrations of HA being dispensed into the adsorption bottles. The effect of HA on TCE adsorption was evident when comparing combinations A and C and B and D. In each case when HA was added to nanoparticles-containing solutions, TCE removal decreased.

The values calculated for the Freundlich constants (k and 1/n) provide a design tool to determine the amount of PAC that would be needed to adsorb a specific amount of TCE. Data from this study may be used as a baseline for future carbon bed filtration experiments.

6. CONCLUSIONS

The presence of nanoparticulate Fe_2O_3 increased the removal of TCE beyond that which was removed by PAC alone. This appears to be a direct relationship, as an increase in the amount of nanoparticles resulted in increased TCE removal. Conversely, the addition of humic acid lowered the adsorption of TCE by PAC, diminishing the increases observed when nanoparticles are present.

7. RECOMMENDATIONS

Given the limited scope of this study, more research is necessary to substantiate project conclusions. Replicate samples to allow statistical evaluation of differences between combinations should be performed. Also, since GAC is more commonly used in drinking water treatment, future research should investigate this form of carbon in addition to PAC. Freundlich constants determined by this research may be useful in further column filtration studies.

8. ACKNOWLEDGEMENTS

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10. APPENDIX I - NOMENCLATURE USED

1/n = Freundlich equation constant

A = area of TCE peak

B = mean concentration of TCE

C_e = TCE concentration

D = dilution factor

k = Freundlich equation constant

L = liter

M = mass of PAC

q_e = TCE removed per gram of PAC

R^2 = correlation coefficient

S = slope of calibration curve

µg = microgram

µL = microliter

11. APPENDIX II - PROJECT TIMELINE

June 19 -25   Prepare isotherm bottles for TCE with and without HA on PAC; also prepare calibration curve for TCE.

June 26 – July 3   Prepare isotherm bottles for TCE with 0.5 and 1.0 mg/L Fe_2O_3 without HA on PAC

July 5 – July 13   Analyze samples prepared June 19 – July 13. Prepare regression analyses for all experimental samples collected.

July 16 – July 20   Prepare isotherm bottles for TCE with 0.5 and 1.0 mg/L Fe_2O_3 and 10 mg/L HA on PAC

July 23 – July 24   Analyze samples prepared July 16 – July 20. Prepare regression analyses for all experimental samples collected.

July 27 – July 31   Prepare reports.

12. APPENDIX III - PROJECT DATA TABLES

Sample Results

Baseline (PAC +TCE)

Sample #	TCE Area	PCE Area	TCE/PCE	Cal. Curve slope	TCE Concentration (ug/L)	Mass of Carbon (g)	q_e (ug TCE Removed/gPAC)	Log Ce	Log qe	Avg. ug Removal of TCE/gPAC
Blank 1 (no PAC)	66.651	13.74125	66.6506	1.7	1133.0602	0				26.2155
B1	45.971	10.72786	45.97156		781.51652	1.8	48.82551111	2.892938	1.688647	
B3	45.778	10.94253	45.778		778.226	3.8	23.34435526	2.891106		
B4	65.605	13.29797	65.60543		557.646155	5.4	26.63953912	2.746359	1.425527	
B6	60.652	9.94956	60.65233		206.217922	9.4	24.65006059	2.314326	1.391818	
B7	46.385	10.46491	46.38488		157.708592	12.4	19.66434694	2.197855	1.29368	
B8	39.728	10.62764	39.72803		67.537651	18.8	14.16918283	1.829546	1.151345	

HA (PAC + TCE + HA)

Sample #	TCE Area	PCE Area	TCE/PCE	Cal. Curve slope	TCE Concentration (ug/L)	Mass of Carbon (g)	q_e (ug TCE Removed/gPAC)	Log Ce	Log qe	Avg. ug Removal of TCE/gPAC
HA1	53.803	12.35391	53.80346	1.7	914.65882	1.6	34.12521563	2.961259	1.533075	27.66608
HA3	37.108	12.23113	37.10783		630.83311	3.7	33.93426284	2.799914	1.530638	
HA4	61.814	14.3258	61.814		525.419	5	30.38206	2.720506	1.482617	
HA5	46.958	14.08886	46.9575		399.13875	6.9	26.59135688	2.601124	1.42474	
HA8	6.5772	14.82052	6.57719		111.81223	19.2	13.29749961	2.048489	1.12377	

(PAC + TCE +0.5 mg/L Fe2O3)

Sample #	TCE Area	PCE Area	TCE/PCE	Cal. Curve slope	TCE Concentration (ug/L)	Ave. ug/L TCE in no Pac Blank	q_e (ug TCE Removed/gPAC)	Log Ce	Log qe	Avg. ug Removal of TCE/gPAC
Blank 1 (no PAC)	79.58949	10.50233	7.57826977	1.7	1353.02133	1333.126315				36.16148
Blank 2 (no PAC)	77.2489	6.54355	11.8053503		1313.2313					
Blank 3 (no NP or PAC)	83.82436	9.13838	9.17278117		1425.01412					
Blank 4 (no NP or PAC)	87.39201	7.43248	11.7581225		1485.66417	Mass of Carbon (g)				
1	53.50303	7.56865	7.06903213		909.55151	1.5	70.59580083	2.958827	1.848779	
2	46.79563	8.56844	5.46139437		795.52571	2.7	49.7778338	2.900654	1.697036	
3	48.45021	7.7557	6.2470454		823.65357	3.8	33.51794375	2.915745	1.525277	
4	72.00633	7.3861	9.74889725		612.053805	5.1	35.34669167	2.78679	1.548349	
5	53.06091	6.28134	8.44738702		451.017735	6.8	32.4304625	2.654194	1.510953	
6	76.34149	7.90855222	9.65302977		259.561066	9.4	28.55226726	2.41424	1.455641	
7	50.86891	7.62035469	6.67539925		172.954294	12.7	22.83803191	2.237931	1.358659	
8	50.85028	7.73999966	6.56980391		86.445476	19.2	16.23282342	1.936742	1.210394	

(PAC + TCE +1.0 mg/L Fe2O3)

Sample #	TCE Area	PCE Area	TCE/PCE	Cal. Curve slope	TCE Concentration (ug/L)	Ave. ug/L TCE in no Pac Blank	q_e (ug TCE Removed/gPAC)	Log Ce	Log qe	Avg. ug Removal of TCE/gPAC
Blank 1 (no PAC)	84.41148	no peak	#VALUE!	1.7	1434.99516	1470.123365				42.43299
Blank 2 (no PAC)	88.54421	9.58021	9.24240805		1505.25157					
Blank 3 (no NP or PAC)	86.48043	9.7445	8.87479399		1470.16731					
Blank 4 (no NP or PAC)	95.60698	9.16986	10.4262202		1625.31866	Mass of Carbon (g)				
1	62.68066	6.18947	10.1269834		1065.57122	1.5	67.4253575	3.027582	1.828823	
2	53.76534	10.893	4.93576976		914.01078	2.6	53.47236394	2.960951	1.728129	
3	37.24828	13.09296	2.84490902		633.22076	3.8	55.05938191	2.801555	1.740831	
4	69.73747	11.45889	6.08588354		592.768495	5	43.8677435	2.772885	1.642145	
5	46.63945	11.12096	4.19383309		396.435325	6.7	40.06298657	2.598172	1.602743	
6	52.56838	9.40443	5.58974653		178.732492	9.3	34.71480841	2.252204	1.540515	
7	39.50038	8.47317	4.66181842		134.301292	12.6	26.50440621	2.12808	1.423318	
8	48.43658	no peak	#VALUE!		82.342186	18.9	18.3568939	1.915622	1.263799	

(PAC + TCE + 1.0 mg/L Fe2O3 + 10 mg/L Humic Acid)

Sample #	TCE Area	PCE Area	TCE/PCE	Cal. Curve slope	TCE Concentration (ug/L)	Ave. ug/L TCE in no Pac Blank	q_e (ug TCE Removed/gPAC)	Log Ce	Log qe	Avg. ug Removal of TCE/gPAC
Blank 1 (no PAC)	79.98708	14.78062	5.41161873	1.7	1359.78036	1300.06174				37.7448
Blank 2 (no PAC)	72.96136	13.93712	5.23503852		1240.34312					
Blank 3 (no NP or PAC)	75.60383	13.61351	5.55358831		1285.26511					
Blank 4 (no NP or PAC)	73.27322	14.08194	5.20334698		1245.64474	Mass of Carbon (g)				
1	57.44544	14.33252	4.00804883		976.57248	1.6	50.54519688	2.989704	1.70368	
2	39.75042	14.06439	2.82631668		675.75714	2.7	57.80598148	2.829791	1.761973	
3	32.12304	14.77746	2.17378629		546.09168	3.9	48.3314141	2.737266	1.68423	
4	47.877	13.4365	3.5632047		406.9545	5.2	42.93784808	2.609546	1.63284	
5	42.68264	13.73736	3.10704822		362.80244	6.8	34.4580625	2.55967	1.537291	
6	73.99689	14.23938	5.19663707		251.589426	9.1	28.80418445	2.400692	1.459456	
7	49.28894	13.59456	3.62563702		167.582396	12.4	22.83224484	2.224228	1.358549	
8	42.38684	14.22679	2.9793678		72.057628	18.9	16.24344063	1.85768	1.210678	

(PAC + TCE + 0.5 mg/L Fe2O3 + 10 mg/L Humic Acid)

Sample #	TCE Area	PCE Area	TCE/PCE	Cal. Curve slope	TCE Concentration (ug/L)	Ave. ug/L TCE in no Pac Blank	q_e (ug TCE Removed/gPAC)	Log Ce	Log qe	Avg. ug Removal of TCE/gPAC
Blank 1 (no PAC)	67.20206	13.33172	5.04076443	1.7	1142.43502	1144.61255				20.14382
Blank 2 (no PAC)	67.45824	12.93989	5.21320042		1146.79008					
Blank 3 (no NP or PAC)	81.73385	12.96593	6.30373988		1389.47545					
Blank 4 (no NP or PAC)	76.55395	13.5278	5.6590096		1301.41715	Mass of Carbon (g)				
1	69.95229	13.67928	5.11374064		1189.18893	1.6	-6.965059375			
2	61.73861	10.69818	5.77094515		1049.55637	2.9	8.194498276	3.021006		
3	45.6648	9.65296	4.73065257		776.3016	3.9	23.60967628	2.89003	1.37309	
4	74.66696	11.88503	6.28243765		634.66916	5.3	24.05393349	2.802547	1.381186	

5	47.46862	12.22536	3.88279936	403.48327	6.8	27.2474	2.605826	1.435325
6	76.18196	10.98695	6.93385881	259.018664	9.2	24.06505125	2.413331	1.381387
7	47.28648	14.35173	3.29482787	160.774032	12.4	19.83545399	2.206216	1.297442
8	40.79677	13.13676	3.10554277	69.354509	19.2	14.00075574	1.841075	1.146151