1 Article

## 2 The impact of humic acid on metaldehyde adsorption onto 3 powdered activated carbon in aqueous solution<sup>†</sup>

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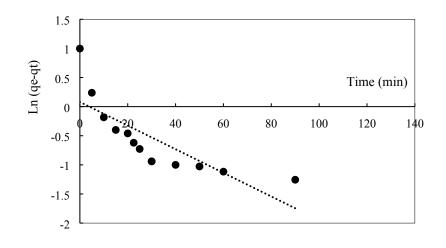
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<sup>†</sup> Electronic supplementary information (ESI) available.

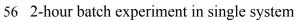
## 26 Adsorption experiment:

- 27 For single adsorption of metaldehyde, three experiments were done: 1) PAC dosage was varied
- 28 from 0.005g to 0.5 g using 500 mL of 1 mg L<sup>-1</sup> metaldehyde solution without adjusting the pH
- 29 of the solution for a 2-hour reaction time; 2) sample solutions were taken at different time
- 30 intervals between 0 minutes and 2 hours using 500 mL of 1 mg L<sup>-1</sup> metaldehyde solution and
- 31 0.05 g PAC without adjusting the pH of the solution; 3) pH of metaldehyde solution was varied
- 32 from 4 to 12 using 500 mL of metaldehyde solution and 0.05 g PAC for a 2-hour reaction time.
- 33 For single adsorption of HA, two experiments were done: 1) PAC dosage was varied from 0.05
- 34 g to 1 g using 500 mL of 30 mg L<sup>-1</sup> HA solution without adjusting the pH of the solution for
- 35 the 2-hour reaction time; 2) the sample solutions were taken at different time intervals between
- 36 0 minutes and 30 days using 500 mL of 30 mg  $L^{-1}$  HA solution without adjusting the pH of the
- 37 solution and 0.25 g PAC.
- 38 For binary adsorption of metaldehyde and HA, two experiments were done: 1) for the 500 mL
- 39 multi-component solution containing metaldehyde and HA, the concentration of HA in the
- 40 binary system was varied from 3 mg L<sup>-1</sup> to 90 mg L<sup>-1</sup> and the concentration of metaldehyde
- 41 was fixed at 1 mg  $L^{-1}$  without adjusting the pH of the solution using 0.05 g PAC for a 2-hour
- 42 reaction time; 2) sample solutions were taken at different time intervals between 0 minutes to
- 43 2 hours using 500 mL of multi-component solution containing 1 mg L<sup>-1</sup> metaldehyde and 30
- 44 mg  $L^{-1}$  HA without adjusting the pH of the solution and 0.05 g PAC.
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55 Figure 1. Pseudo-first order kinetic model fitting of 1 mg L<sup>-1</sup> metaldehyde with 0.05g PAC in



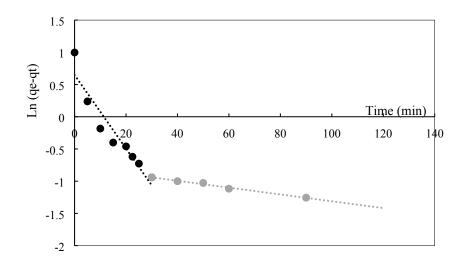
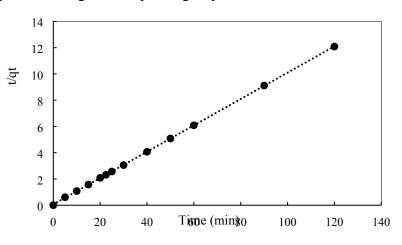






Figure 2. Two gradients plotting of pseudo-first order kinetic model



- 60 Figure 3. Pseudo-second order kinetic model fitting of 1 mg  $L^{-1}$  metaldehyde with 0.05g PAC
- 61 in 2-hour batch experiment in single system

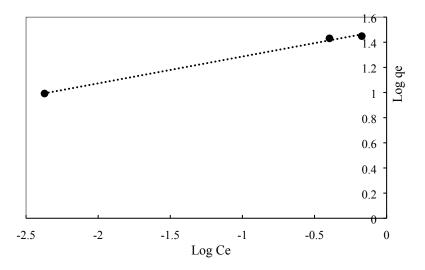
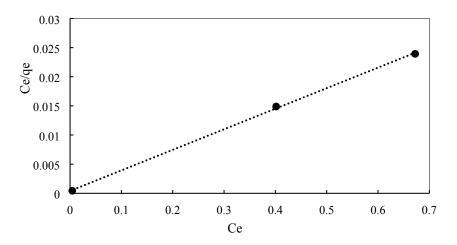




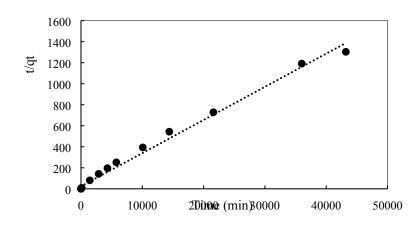
Figure 4. Freundlich isotherm fitting for single adsorption of metaldehyde



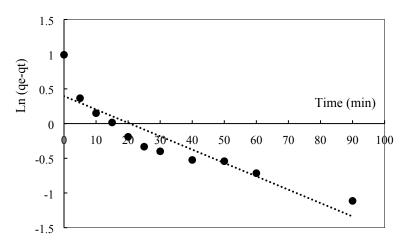
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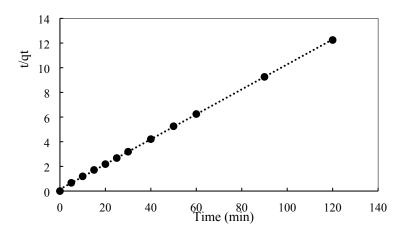
Figure 5. Langmuir isotherm fitting for single adsorption of metaldehyde



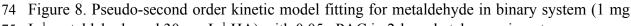
- 68 Figure 6. Pseudo-second order kinetic fitting of 30 mg L<sup>-1</sup> metaldehyde with 0.25g PAC in 2-
- 69 hour batch experiment in single system



- 71 Figure 7. Pseudo-first order kinetic model fitting for metaldehyde in binary system (1 mg L<sup>-1</sup>
- 72 metaldehyde and 30 mg  $L^{-1}$  HA) with 0.05g PAC in 2-hour batch experiment



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75  $L^{-1}$  metaldehyde and 30 mg  $L^{-1}$  HA) with 0.05g PAC in 2-hour batch experiment

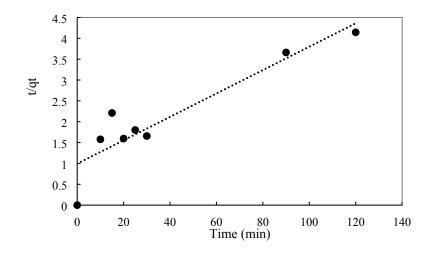


Figure 9. Pseudo-second order kinetic model fitting for HA of in binary system (1 mg  $L^{-1}$ metaldehyde and 30 mg  $L^{-1}$  HA) with 0.05g PAC in 2-hour batch experiment

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80 Table 1. Removal of metaldehyde and HA under different experiment conditions (p values are

81 from ANOVA single-factor statistic test for analysing concentrations of solutions before and

	Percentage removal	Initial concentration	Initial concentration (mg L <sup>-1</sup> )		m vialues	
	(%)	Metaldehyde	НА	PAC dosage (g L <sup>-1</sup> )	<i>p</i> value	
	$30.3 \pm 6.4$	1		0.01	2.28×10-7	
Removal of metaldehyde (single)	$57.8 \pm 2.7$	1		0.02	1.07×10 <sup>-9</sup>	
	$99.6 \pm 0.0$	1	n/a	0.1	4.07×10 <sup>-12</sup>	
	$100.0\pm0.0$	1		0.2	8.24×10 <sup>-16</sup>	
	$100.0 \pm 0.0$	1		1	1.45×10 <sup>-12</sup>	
Removal of HA (single)	$9.8 \pm 0.1$		30	0.1	8.5×10 <sup>-10</sup>	
	$14.8 \pm 0.1$		30	0.2	6.96×10 <sup>-11</sup>	
	$21.6 \pm 0$	n/a	30	0.5	6.72×10 <sup>-11</sup>	
	$25.1 \pm 0.1$		30	1	1.39×10 <sup>-12</sup>	
	$32.0 \pm 0.1$		30	2	9.8×10 <sup>-11</sup>	
Removal of metaldehyde (binary)	$98.6 \pm 0.2$	1	3	0.1	1.24×10 <sup>-18</sup>	
	$98.4 \pm 0.3$	1	9	0.1	8.48×10 <sup>-16</sup>	
	$97.8 \pm 0.5$	1	15	0.1	2.53×10 <sup>-18</sup>	
	$96.2 \pm 0.5$	1	30	0.1	1.37×10 <sup>-18</sup>	
	$89.9 \pm 1.4$	1	60	0.1	9.76×10 <sup>-16</sup>	
	$90.2 \pm 1.0$	1	90	0.1	1.31×10 <sup>-14</sup>	
Removal of HA (binary)	$20.5 \pm 0.5$	1	3	0.1	1.98×10 <sup>-7</sup>	
	$16.3 \pm 0.4$	1	9	0.1	2.71×10 <sup>-7</sup>	
	$14.8 \pm 0$	1	15	0.1	6.53×10 <sup>-11</sup>	
	$11.3 \pm 1.6$	1	30	0.1	1.11×10 <sup>-10</sup>	
	$7.7 \pm 0$	l	60	0.1	1.34×10 <sup>-10</sup>	
	6.5 ± 0	l	90	0.1	8.73×10 <sup>-10</sup>	
82 after treatments	)					
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Table 2. Comparison table of adsorption of metaldehyde and HA onto GAC and PAC

Sample solution (500 mL)	2 g/L GAC (purchased from sigma-aldrich, 20 mesh size, specific surface area = 650 m <sup>2</sup> g <sup>-1</sup> ) Removal (%)		0.1 g/L PAC (used in this study, specific surface area = 962 m <sup>2</sup> g <sup>-1</sup> ) Removal (%)		0.2 g/L PAC ( used in this study, specific surface area = 962 m <sup>2</sup> g <sup>-1</sup> ) Removal (%)	
	Metaldehyde	НА	Metaldehyde	HA	Metaldehyde	НА
Mono-component	99.70±0.15		99.6		100	
solution of metaldehyde						
(1 mg L <sup>-1</sup> )						
Multi-component	98.99±0.58	21.56	96.2±0.5	11.3 ±		
solution of				1.6		
metaldehyde (1 mg L <sup>-1</sup> )	$k_2 = 0.016 \text{ g}$		$k_2 = 0.069 \text{ g}$			
and HA (30 mg L <sup>-1</sup> )	mg <sup>-1</sup> min <sup>-1</sup>		mg <sup>-1</sup> min <sup>-1</sup>			
Mono-component		22.67		9.8±		14.8 ±
solution of HA (30 mg L <sup>-1</sup> )				0.1		0.1