

## Evaluation of Quantitative Determination of Dissolved Organic Matter Adsorption Processes Using Charcoals and Activated Carbon

Asa MIURA<sup>1\*</sup>, Eisaku SHIRATANI<sup>2</sup>, Ikuo YOSHINAGA<sup>1</sup>,  
Tadayoshi HITOMI<sup>1</sup>, Koji HAMADA<sup>1</sup> and Kyoji TAKAKI<sup>1</sup>

<sup>1</sup> Department of Rural Environment, National Institute for Rural Engineering  
(Tsukuba, Ibaraki 305–8609, Japan)

<sup>2</sup> Rural Development Bureau, Ministry of Agriculture, Forestry and Fisheries of Japan  
(Chiyoda, Tokyo 100–8950, Japan)

### Abstract

In this study the processes of dissolved organic matter (DOM) adsorption by charcoals and activated carbon were quantitatively evaluated by the adsorption rate model. Five types of charcoal and a commercial granular activated carbon (GAC) were used to remove DOM eluted from rice-straw in an adsorption test. The adsorption rate model was considered as the adsorption process in the early steps which showed significant adsorption changes. The results of the adsorption test indicated that the DOM adsorption rate and volume treated by charcoals carbonized at high temperature (group-2) showed significantly higher levels than those of charcoals carbonized at low temperature (group-1). Although group-2 adsorbed less DOM than GAC after 6 hours, the adsorption rate and adsorbed volume of group-2 were greater than those of GAC under 6 hours. The results of the evaluation by the adsorption rate model showed that the calculated value agreed well with the measurements obtained from the adsorption test. It was suggested that DOM could move to the adsorption surface of group-2 more rapidly than that of the GAC in the early adsorption process. In the latter process the volume of DOM adsorption from sample water treated by group-2 was exceeded by that of the GAC which has a large specific surface area. The DOM adsorption processes depended on the specific surface area of charcoals/GAC as well as the DOM transfer rate and desorption.

**Discipline:** Agricultural environment

**Additional key words:** adsorption rate equation, specific surface area, transfer rate

### Introduction

Dissolved organic matter (DOM), which includes humic substances derived from stems and leaves<sup>15,21</sup> can have negative effects on the water environment. Discharges and accumulations of DOM, especially recalcitrant-DOM, can remain for long periods of time in bodies of water<sup>15</sup>. DOM can be a carrier of toxic chemical substances since it can combine with substances such as heavy metals and pesticides in the environment<sup>1,5,14,16</sup>. Furthermore, DOM includes humic substances which can be a precursor to trihalomethane during the drinking water treatment process. One of DOM sources is known to be farmland. DOM discharges from paddy fields during

puddling<sup>9</sup>. Therefore, it is important to remove DOM from agricultural drainage for conservation of water environments.

Activated carbon, which is a popular adsorption material, is used to remove residual organic matter with small molecules after coagulation using Al<sup>3+</sup> or Fe<sup>2+</sup>, Fe<sup>3+</sup> during the drinking water treatment process<sup>19</sup>. Activated carbon cannot adsorb humic substances with large molecules because the pore diameter of activated carbon is too small for humic substances<sup>8,19</sup>. Activated carbons undergo carbonization and activation treatment in the production process, so they require more time to make and the cost is higher.

Wood charcoal has recently been attracting attention as an environmental purification material instead of acti-

---

Present address:

<sup>2</sup> Department of Rural Technologies, National Institute for Rural Engineering (Tsukuba, Ibaraki 305-8609, Japan)

\*Corresponding author: e-mail [asam@affrc.go.jp](mailto:asam@affrc.go.jp)

Received 21 February 2007; accepted 30 July 2007.

vated carbons. We reported in a previous study that the wood charcoal made from thinned wood could adsorb DOM<sup>12,13</sup>. We also showed that wood charcoal carbonized at 1,050°C has the potential to remove humic substances<sup>13</sup>. The Japanese government is promoting the use of waste woods as a biomass resource in our country<sup>10</sup>. A report on the utilization of woody biomass in 2005 showed that most thinned wood and tree material remaining in forests were not used<sup>11</sup>. Accordingly, wood charcoal can play an important role in using this recyclable material resource in the environment since it can be made from waste woods.

In this study, the DOM adsorption rate and volume of charcoal was compared with those of activated carbon in order to use charcoal as an adsorption material which can be an environmentally-friendly recyclable material in Japan. If the adsorption capability of charcoal performs as well as or better than that of activated carbon, it could be a useful application for overabundant resource materials such as thinned wood. The data on DOM concentration treated by activated carbon was added to the results of the DOM adsorption test in our previous report<sup>13</sup>. Additionally, the adsorption processes were evaluated by the coefficients with the adsorption rate model equation and the physical properties for charcoals and activated carbon. The adsorption processes for charcoals and activated carbons are also discussed.

## Materials and methods

### 1. Preparation of charcoals and activated carbon

We used five types of charcoal and an activated carbon in order to examine the DOM adsorption process. The charcoals were made from thinned wood from cedar (*Cryptomeria japonica* D. Don). The activated carbon was a commercial granular activated carbon made from coal.

The five types of charcoal were carbonized at 650°C to 1,050°C in intervals of 100°C (hereafter C650, C750, C850, C950, and C1050) at the carbonization factory of the Forestry Cooperative of Fukushima Prefecture, Japan. The starting carbonization temperature, 650°C was decided by the criteria described in our previous study<sup>13</sup>. The carbonization temperature interval of 100°C was the setting limit of the carbonization equipment. Two kinds of manufacturing methods, a one-stage method and two-stage method, were used for the carbonization process. This carbonization factory generally operates using the two-stage method for the hard charcoal carbonized at more than 750°C. In other words, carbonization is performed once for charcoal carbonized at less than 750°C, and twice at more than 750°C. The carbonization process is different due to differences in the sizes and kinds of raw

materials used or the carbonization furnace. For example, in the case of the one-stage carbonization with high temperature, charcoal is carbonized imperfectly or is burned out, thereby the material yield decreases. Therefore the high temperature charcoal is produced by a two-stage method. The carbonization furnace at the carbonization factory in this study was not an adequate facility for carbonizing charcoal at high temperature.

The temperature is controlled with room temperature airflow from an air blower which has a number of nozzles. The airflow prevents an excessive temperature increase which allows the raw material to be added in the furnace continuously producing the charcoal without burning itself out. The carbonizing temperature is controlled by a temperature sensor in the center of the furnace. The finished charcoals are removed from the furnace continuously.

In the one-stage method, dried wood chips were carbonized at 650°C or 750°C using a batch treatment furnace (hereafter C650 and C750: group-1). In the two-stage method, wood chips carbonized at 700°C during the one-stage carbonization process were used as the starting material, and they were further carbonized up to 850°C, 950°C or 1,050°C in a rotary-type kiln (hereafter C850, C950 and C1050: group-2) following a similar process to the one-stage method. The carbonization process used in this study was similar to the method reported by Pulido-Novicio et al.<sup>17,18</sup>, in which high temperature carbonization used a two-stage method and low temperature carbonization used a one-stage method.

The granular activated carbon (Shirasagi WH5C, Japan EnviroChemicals, Ltd.; hereafter GAC) was made from coal and selected as a reference to compare the performance of the charcoals. The GAC was produced as follows. After crushing and shaping, carbon material was carbonized at between 700°C and 800°C and then activated with water vapor at between 900°C and 1,000°C<sup>3</sup>. The manufacturer reported that this GAC adsorbs humic substances relatively easily compared to the other activated carbons available<sup>4</sup>. Although the GAC has the larger specific surface area, the larger number of pores and the smaller size of pores, the raw material of GAC are confined, including coal and palm shell<sup>3,4</sup>.

The physical properties of the charcoals and GAC are shown in Table 1. These data were measured by the Brunauer-Emmett-Teller (BET) method with nitrogen gas. There are significant differences between charcoals and GAC in the pore volume and the specific surface area. The pore volume of group-1 was 1/7–1/17 times, and that of group-2 was 1/4–1/5 times as large as that of the GAC. The specific surface area of group-1 was 1/10–1/12 times, and that of group-2 was 1/4 times as large as that of the

**Table 1. Physical characteristics of the charcoals and GAC used**

Adsorbent	Group-1		Group-2			GAC
	C650	C750	C850	C950	C1050	
Particle diameter (mm)	1.15	1.40	1.40	1.50	1.41	1.62
Mean pore diameter (nm)	$4.62 \times 10$	$5.63 \times 10$	$2.33 \times 10$	$3.90 \times 10$	$2.19 \times 10$	$2.97 \times 10$
Pores distribution	Macro-pores (%)	0.01	—	0.01	0.01	0.01
	Meso-pores (%)	37.19	47.10	13.00	17.63	14.50
	Micro-pores (%)	62.80	52.90	87.00	82.36	85.49
Pore volume (mL g <sup>-1</sup> )	0.04	0.09	0.17	0.15	0.12	0.68
Specific surface area (m <sup>2</sup> g <sup>-1</sup> )	$0.55 \times 10^2$	$0.78 \times 10^2$	$2.25 \times 10^2$	$2.04 \times 10^2$	$2.24 \times 10^2$	$9.41 \times 10^2$
Standard deviation of surface area	12.87	3.87	24.98	9.47	15.53	62.04

GAC. These differences in physical properties between the charcoals and GAC depend on the following differences in pore structure. The physical pores of the porous structure are classified into macro-pores (> 50 nm), meso-pores (= 2–50 nm) and micro-pores (< 2 nm). Generally, the micro-pores of GAC, which has strong adsorptivity<sup>20</sup>, hold the largest share of its surface area<sup>23</sup>. The pore distribution for charcoals and GAC are shown in Table 1. The structure of charcoal micro-pores was less developed than that of GAC. The distributions of the micro-pores of group-2 were larger than those of group-1.

Pulido-Novicio et al.<sup>17</sup> reported differences in the physical properties of charcoal produced in the carbonization process by the one-stage and two-stage methods. According to their results, the difference between 1-step and 2-step carbonization was as follows. For example, the specific surface area and the total pore volume of charcoal carbonized at 1,000°C with a two-stage method were larger than those of a one-stage by 20 times and 15 times, respectively. Both the specific surface area and the total pore volume of a two-step method for charcoal carbonized at 700°C were 2 times larger than those of a one-stage method. The development of pore structures was significant when carbonizing at more than 800°C with a two-stage method. Therefore, the high temperature carbonization using a two-stage method seems to result in perfectly carbonized charcoal, and would contribute to adsorbing DOM.

## 2. Treatment of DOM solutions with charcoals

Solutions including DOM used in this study were prepared from rice straw immersed in tap water and aerated for more than three months (hereafter rice-straw water). The rice-straw water showed a brownish color.

We conducted a test of DOM adsorption in order to understand the DOM adsorptive process onto the char-

coals and GAC. The DOM adsorption experiment was performed using a batch-wise test. The experimental procedure is as follows. Five types of charcoals and GAC (12 g each) were placed in a bottle with 800 mL of the rice-straw water. The sample waters in the bottles were continuously aerated at 20±1°C. The rice-straw water without charcoal or GAC was used as control water. We took the sample waters by amounts of about 20 mL from the bottles after 0, 0.25, 1, 3, 6, 24, and 144 hours of immersing the charcoals in the rice-straw water. We did not add water to the sample water in the bottles. This method had no negative effect on the following TOC concentration because every sampling volume was less than 3% for the residual solution volume in the bottle. The samples were filtered through washed membrane filters (pore size, 0.2 µm; AJ0511-155, Whatman, USA). The DOM adsorption onto charcoals was estimated by determining the total organic carbon (TOC) concentration. The TOC in the samples was analyzed as nonpurgeable organic carbon by means of the 680°C combustion catalytic oxidation/nondispersive infrared method with a TOC analyzer (TOC-V, Shimadzu, Japan).

## 3. Calculation of adsorption rate

Many adsorption rate equations have been proposed by researchers<sup>6</sup>. Our study was based on the adsorption rate equation by Langmuir which has two independent phenomena, adsorption and desorption, simultaneously<sup>6</sup>. The adsorption phenomena in the adsorption rate process have some elementary steps because the adsorption onto the porous adsorbent is complicated. Generally, the adsorption processes for liquid phase are explained as follows<sup>2,23</sup>: (1) the process of adsorbate diffusion toward the charcoals/GAC in solution, (2) the process of DOM diffusion in the liquid laminar film on the surface of the charcoals/GAC, (3) the process of adsorbate diffusion within

the particle pores of charcoals/GAC, and (4) the process of adsorption onto the pore surface of charcoals/GAC. The processes controlling the adsorption speed (the rate-determining processes) are steps (2) and (3). Step (2), which is the early step of the adsorption process, is rapid, while step (3), which is the latter, is slow. It is suggested that the speed in the adsorption process depends on step (2). That is, the adsorption phenomenon at the early stage of the process could determine the immediate adsorptive effect. Supposing that these typical adsorption processes can apply to the DOM adsorption process of the charcoals/GAC in this study, we focused on the liquid laminar film diffusion (step (2)) of charcoals/GAC in order to examine the adsorption rate differences in the early stage between the charcoals and GAC.

DOM transfer was assumed to be one-dimensional diffusion in the liquid laminar layer between the surfaces (including pore wall surface) of the charcoals/GAC and the sample water. The transfer rate,  $N$  ( $\text{mg h}^{-1}$ ) is presented as follows<sup>7</sup>;

$$N = Ak_f(C(t) - C_e) \quad (1)$$

where  $A$  is the specific surface area of adsorbent per the amount of charcoals/GAC ( $\text{m}^2$ ),  $k_f$  is the mass transfer coefficient ( $\text{m h}^{-1}$ ),  $C(t)$  is the adsorption concentration of time  $t$  in solution ( $\text{mg m}^{-3}$ ), and  $C_e$  is the adsorption equilibrium concentration ( $\text{mg m}^{-3}$ ). The solution concentration of interface in contact with the surface of the charcoal is assumed to be the adsorption equilibrium concentration. DOM that passes through the liquid laminar layer for time  $dt$  is

$$Ndt = v_a \rho_a dq \quad (2)$$

where  $v_a$  is the volume of adsorbent ( $\text{m}^3$ ),  $\rho_a$  is the density of adsorbent ( $\text{mg m}^{-3}$ ), and  $dq$  is the variation in adsorption volume for the weight of the charcoals/GAC on minute time  $dt$  ( $\text{mg mg}^{-1}$ ).

Similarly, the relationship between the change in the DOM adsorption volume onto the surface of charcoals/GAC and the change of concentration in the sample water is shown as the following equation;

$$v_a \rho_a dq = VdC \quad (3)$$

where  $V$  is the volume of rice-straw water ( $\text{m}^3$ ) and  $dC$  is the change in the concentration of solution ( $\text{mg m}^{-3}$ ). Therefore, Eq. (1), Eq. (2) and Eq. (3) are represented as follows.

$$\left(\frac{dC}{dt}\right)_1 = -\frac{A}{V}k_f(C(t) - C_e) \quad (4)$$

In addition to the phenomena described above, the DOM can stay on the surface of the charcoals/GAC and move away from them. Therefore, DOM desorption from the charcoals/GAC can be expressed with the following equation<sup>6</sup>;

$$V \frac{dC}{dt} = \alpha V(C_0 - C(t))$$

$$\left(\frac{dC}{dt}\right)_2 = \alpha(C_0 - C(t)) \quad (5)$$

where  $\alpha$  is the desorption rate coefficient ( $\text{h}^{-1}$ ) and  $C_0$  is the initial concentration in solution ( $\text{mg m}^{-3}$ ).

These two differences in phenomenon (Eq. (4) and Eq. (5)) can occur simultaneously<sup>4</sup>.

$$\left(\frac{dC}{dt}\right) = \left(\frac{dC}{dt}\right)_1 + \left(\frac{dC}{dt}\right)_2$$

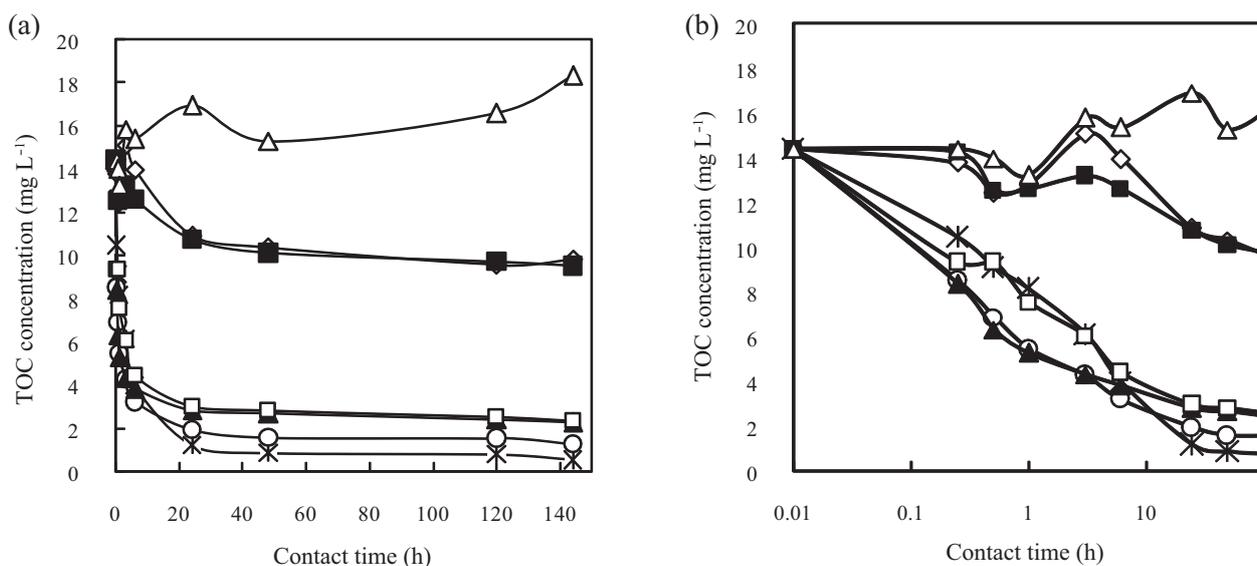
$$= -\frac{A}{V}k_f(C(t) - C_e) + \alpha(C_0 - C(t)) \quad (6)$$

Consequently, we estimated the DOM adsorption process by the charcoals/GAC with Eq. (6). The values of  $C_e$  were assumed to be the concentration in solution at 144 hours in this test. This is because the sample water concentration changes in this test were predicted to stabilize at 144 h and later. This equation indicates that the DOM concentration in the solution would depend on three parameters ( $k_f$ ,  $\alpha$ ,  $A$ ).

## Results and discussion

### 1. DOM removal experiments

The change in the TOC concentration in the sample water treated by the charcoals/GAC showed significant differences with contact time. Figure 1 (a) shows the relationship between contact time and the TOC adsorption by the charcoals/GAC. At the beginning of the test the rate of decrease in TOC concentration showed significant differences between group-1 and group-2/GAC (Fig. 1 (a)). After 0.25 hours of contact time, adsorption of group-1 was around 1–4%, that of group 2 was around 35–40%, and that of GAC was 27.4% of TOC. The DOM adsorption rate at the beginning was reported to depend on the pore size of the charcoal surface determined by the carbonization temperatures<sup>13</sup>. Group-2 and GAC having smaller pore sizes showed faster DOM adsorption rates. These results are explained by the fact that DOM could be adsorbed by molecular force<sup>7,23</sup> and Polanyi potential theory<sup>7,22</sup> depending on the pore size. Therefore, it was suggested that the difference in pore size on the sur-



**Fig. 1. Change of TOC concentration in sample water treated with 5 types of charcoal and GAC**

The x-axis in (a) is represented as a logarithmic scale in (b).  
 \* : Activated Carbon, ○ : 1050C, ▲ : 950C, □ : 850C,  
 ◇ : 750C, ■ : 650C, △ : Control.

face could determine the adsorption rate for group-1 and group-2/GAC.

During the first 6 hours in the adsorption test, the rate of TOC decrease by adsorption to charcoals carbonized at high temperatures (C950, C1050) was more rapid than that of the GAC. The X-axis in Fig. 1 (b) is represented as a logarithmic scale in order to closely examine the beginning of decreasing TOC concentration. The decrease in the TOC concentration for group-2, except for C850, was larger than that of GAC for the first 6 hours (Fig. 1 (b)). However, the decrease in TOC concentration of group-2 was less than that of GAC after 6 hours. It was considered that the adsorption volume of GAC was larger than that of group-2 in the latter adsorption process because GAC has a specific surface area and pore volume larger than group-2. The reason for the increased TOC at around 3 hours for control, C750 and C650 in Fig. 1 (b) was not clear. It seemed that the increased TOC for the control was due to TOC eluted from some fragments of rice-straw, and that of group-1 was due to the lack of molecular force between the charcoals and TOC.

These phenomena could be explained as follows. All steps from step (1) to step (4) would be proceeding simultaneously. In the early process step (2) was dominant, and in the latter process step (3) was dominant. When the volume of decreasing TOC concentration of group-2 was less than that of GAC, step (3) would be more dominant than step (2) in the adsorption process of GAC.

At the final stage of adsorption approaching the sta-

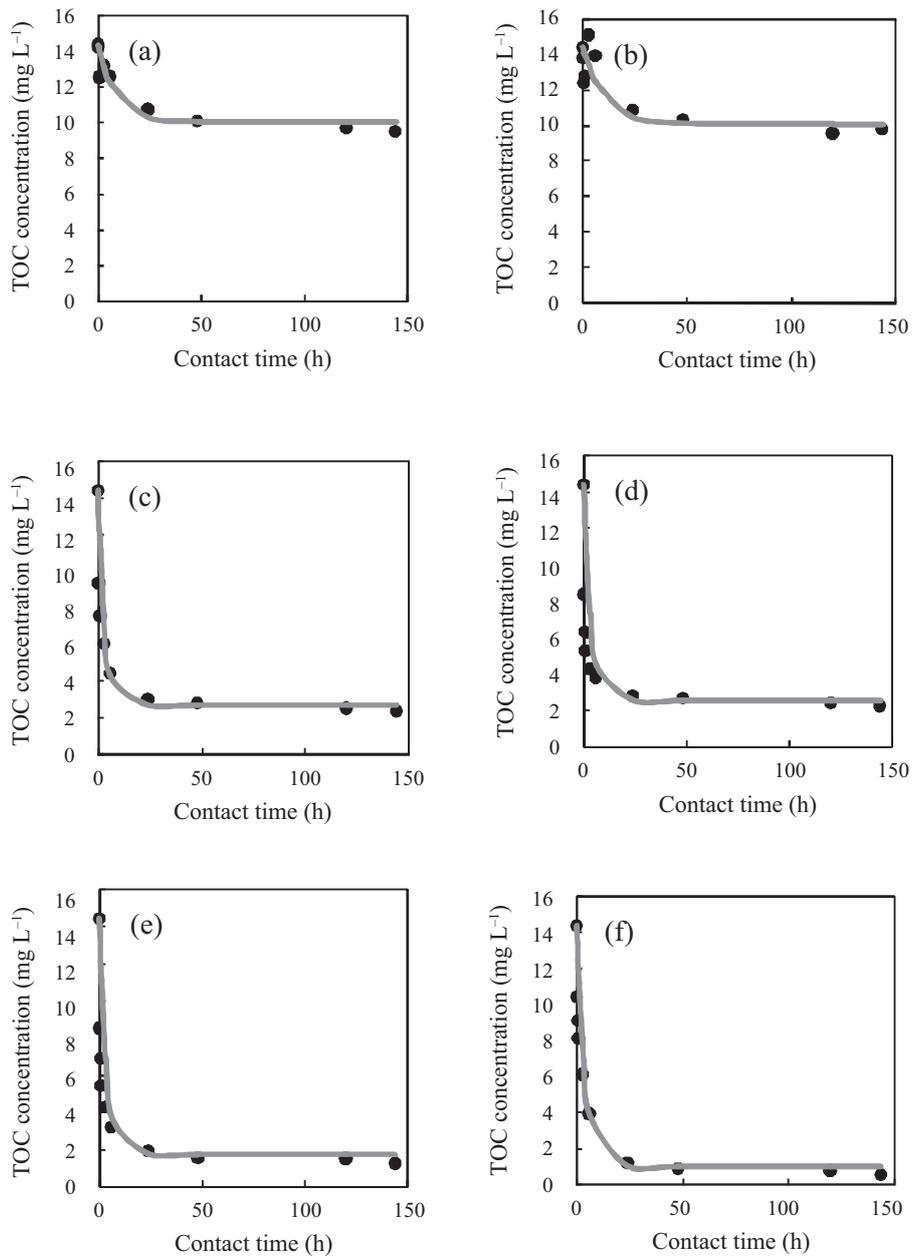
ble state, the adsorption volume depends on the specific surface area on the adsorbate<sup>7</sup>. GAC can adsorb adsorbate a little more than group-2 because of its large pore volume and specific surface area (Table 1). Consequently, the adsorption rates on C1050 and GAC reached 90% or greater at 144 hours and both showed equivalent adsorption for DOM.

## 2. Evaluation of adsorption rate by the adsorption model

The coefficients of Eq. (6) on the charcoals/GAC were compared with each other in order to understand the adsorption phenomena. The results of the adsorption test indicated the difference in adsorption rate between group-1, group-2 and the GAC. In particular, the results in the early process showed that the adsorption rate of group-2 was higher than that of the GAC. In this section, we evaluated the coefficients calculated by Eq. (6), in order to discuss the early process.

The changes in the measured TOC concentration in the sample water treated by charcoals and GAC agreed well with the concentrations calculated by the model equation (Eq. (6)). Figure 2 shows the plots of the measurements obtained from the test and the calculated values. We also obtained highly correlated correlation coefficients on all charcoals and GAC (Table 2).

The model parameters in the model calculations are shown in Table 2. We have considered the charcoal adsorption rate property by categorizing it into three



**Fig. 2. Comparison of TOC concentration between measurements in the adsorption test and calculated by the adsorption rate model onto (a) C650, (b) C750, (c) C850, (d) C950, as well as (e) C1050 and (f) GAC**  
 ● : Measured, — : Calculated.

**Table 2. Parameters of adsorption rate model onto 5 types of charcoal and GAC**

Adsorbent	Group-1		Group-2			GAC
	C650	C750	C850	C950	C1050	
$K_f$ (m h <sup>-1</sup> )	$9.70 \times 10^{-8}$			$11.4 \times 10^{-8}$		$2.00 \times 10^{-8}$
$\alpha$ (h <sup>-1</sup> )	0.015			0.011		0.015
$C_e$ (mg m <sup>-3</sup> )	9.49	9.8	2.33	2.24	1.26	0.52
R	0.95	0.84	0.95	0.85	0.90	0.96

R: correlation coefficients.

groups. Categorizing the groups made it possible to clarify the rate properties of both charcoals and GAC. We have explained rate properties by categorizing groups in a previous report<sup>12</sup> using the results of the adsorption test in this study. These parameters were selected to minimize the square of the differences between the measurements and calculated values. The parameters of the mass transfer coefficient  $k_f$  were the liquid laminar film diffusion and the desorption rate coefficient  $\alpha$  on charcoals/GAC. As these parameters,  $k_f$  and  $\alpha$  most closely approached each value for the minimum square of differences between the measurements and calculated values, each parameter was assumed to be equivalent by group, group-1, group-2, and GAC. Their values are shown in Table 2. The specific surface area,  $A$  was given analytical values (Table 1). We discuss three parameters ( $k_f$ ,  $\alpha$ ,  $A$ ) below.

Parameter  $k_f$ , which implies the mass transfer coefficient, showed the largest value for group-2. The values of  $k_f$  in increasing order are as follows; GAC ( $2.00 \times 10^{-8}$ ) < group-1 ( $9.70 \times 10^{-8}$ ) < group-2 ( $1.14 \times 10^{-7}$ ). These results indicated that DOM translation speed was relatively high in the liquid laminar film of group-2, and was low in that of the GAC.

Parameter  $\alpha$  implies the adsorption rate coefficient. There was not so great a difference between each value. The value of group-2 ( $\alpha = 0.011$ ) was somewhat larger than the others ( $\alpha = 0.015$ ). This indicates that it was relatively hard to release adsorbed material from group-2.

These results were summarized as follows. In the early adsorption process the DOM adsorption volume by group-2 which had the largest  $k_f$  was larger than the GAC. In other words, it was suggested that DOM could move to the surface of charcoal carbonized at high temperatures more rapidly than that of the GAC. However, in the latter process the DOM adsorption volume by GAC which had the largest  $A$  exceeded those of group-2 and group-1. Group-1 adsorbed the least DOM because  $k_f$  and  $A$  were smaller than those of group-2. Therefore, it was clear that the factors affecting DOM adsorption were not only the DOM transfer rate and desorption, but also the specific surface area.

## Conclusion

In this study the DOM adsorption processes using five types of charcoal (C650, C750, C850, C950, and C1050) and commercial granular activated carbon (GAC) were quantitatively evaluated with the adsorption rate model based on the measurements obtained from the adsorption test. The adsorption rate model was considered to consist of an adsorption process in the early step, which has a liquid laminar film diffusion and a desorption

process by collision of DOM.

In the results of the adsorption test, the significant difference in the adsorption volume and rate were shown between group-1 (C650 and C750) with the one-stage method and group-2 (C850, C950 and C1050) with the two-stage method. Similar differences were also shown between group-2 and the GAC. Although the GAC with the largest specific surface area could adsorb DOM better than group-2 at reaching to the near stable state after 6 hours in the test, C950 and C1050 adsorbed DOM quicker and better than GAC in the first 6 hours. The DOM adsorption rates of C1050 and GAC reached over 90% at 144 hours. Consequently, the DOM adsorption volume of C1050 was not significantly less than that of GAC, and the rate of initial TOC concentration decrease by C1050 was quicker than that of GAC. Therefore, it was suggested that C1050 could be used for DOM adsorption as well as GAC.

The measurements from the adsorption test agreed well with the values calculated by the adsorption rate equation (Eq. (6)). The specific surface area of the charcoals/GAC as well as transfer rate in liquid laminar film and adsorption rate could influence the adsorption process.

In this study we evaluated the adsorption rate process of only step (2) focusing on the early process. In order to understand the adsorption rate process and create a model which more closely approaches measured values, we need to consider the latter process of step (3) in further research.

## References

1. Ginneken, V. L. & Blust, R. (2000) Determination of conditional stability constants of cadmium-humic acid complexes in freshwater by use of a competitive ligand equilibration-solvent extraction technique. *Environ. Toxicol. Chem.*, **19**, 283–292.
2. Ide, T. (1993) Water treatment engineering: theory and application. Gihodo Syuppan Co. Ltd., Tokyo, pp.738 [In Japanese].
3. Japan EnviroChemicals Co., Ltd. (2007) Activated carbon. <http://www.jechem.co.jp/shirasagi/abt/index.html> [In Japanese].
4. Japan EnviroChemicals Co., Ltd. (2004) Activated carbon, sales brochure for EnviroChemicals Co., Ltd. [In Japanese].
5. Kaschl, A., Romheld, V. & Chen, Y. (2002) Cadmium binding by fractions of dissolved organic matter and humic substances from municipal solid waste compost. *J. Environ. Qual.*, **31**, 1885–1892.
6. Keii, T. (1965) Kyuchaku. *Kyoryitsu Zensho*, **157**, 204 [In Japanese].
7. Kondo, S., Ishikawa, T. & Abe, I. (2002) Kyuchaku no kagaku, second edition. Maruzen Co. Ltd., Tokyo, pp.223

- [In Japanese].
8. Li, F. et al. (2003) Microcolumn test and model analysis of activated carbon adsorption of dissolved organic matter after pre-coagulation: effects of pH and pore size distribution. *J. Colloid Interf. Sci.*, **262**, 331–341.
  9. Maie, N., Watanabe, A. & Kimura, M. (2004) Chemical characteristics and potential source of fulvic acids leached from the plow layer of paddy soil. *Geoderma*, **120**, 309–323.
  10. Ministry of Agricultural Forestry and Fisheries of Japan, MAFF (2007) Biomass-Nippon homepage. <http://www.maff.go.jp/biomass> [In Japanese].
  11. Ministry of Agricultural Forestry and Fisheries of Japan, MAFF (2005) Biomass no riyoujyokyou (status of biomass utilization). [http://www.maff.go.jp/biomass/pdf/riyou\\_jyokyou.pdf](http://www.maff.go.jp/biomass/pdf/riyou_jyokyou.pdf) [In Japanese].
  12. Miura, A. et al. (2007) Removal of recalcitrant dissolved organic matter using carbonized recycled sugi wood. *J. Jpn. Soc. Water Environ.*, **30**, 39–44 [In Japanese].
  13. Miura, A. et al. (2007) Characteristics of the adsorption of dissolved organic matter by charcoals carbonized at different temperatures. *JARQ*, **41**, 211–217.
  14. Naidu, R. & Harter, R. D. (1998) Effect of different organic ligands on cadmium sorption by and extractability from soils. *Soil Sci. Soc. America J.*, **62**, 644–650.
  15. National Institute for Environmental Studies (2001) Studies on origin and dynamics of recalcitrant organic matter in lake and its effects on lacustrine ecosystem and water quality. Report of Special Research from the National Institute for Environmental Studies, Japan, SR-36, pp.38 [In Japanese]. <http://www.nies.go.jp/kanko/tokubetu/sr36/sr36.pdf>.
  16. National Institute for Environmental Studies (2004) Studies on mass balance of dissolved organic matter in lake and its functions and effects on lacustrine ecosystem and water quality. Report of Special Research from the National Institute for Environmental Studies, Japan, SR-62, pp.52 [In Japanese]. <http://www.nies.go.jp/kanko/tokubetu/sr62/sr62.pdf>.
  17. Pulido-Novicio, L. et al. (2001a) Adsorption capacities and related characteristics of wood charcoals carbonized using a one-step or two-step process. *J. Wood Sci.*, **47**, 48–57.
  18. Pulido-Novicio, L. et al. (2001b) Adsorption of mercury by sugi wood carbonized at 1,000°C. *J. Wood Sci.*, **47**, 159–162.
  19. Tanbo, N. (1983) Suido to toriharometan. Gihodo Shuppan Co. Ltd., Tokyo, pp.273 [In Japanese].
  20. Tatemoto, H. & Abe, I. (2000) Applications technology of activated carbon. Techno System, Japan, pp.660 [In Japanese].
  21. Tsutsuki, K. (1995) Characteristics and mechanism of formation of humic substances. *J. Jpn. Soc. Water Environ.*, **18**, 252–256 [In Japanese].
  22. Urano, K., Koichi, Y. & Nakazawa, Y. (1981) Equilibria for adsorption of organic compounds on activated carbons in aqueous solutions. I. Modified Freundlich isotherm equation and adsorption potentials of organic compounds. *J. Colloid Interf. Sci.*, **81**, 477–485.
  23. Yoshida, H. (2005) Handbook on porous adsorbents. Fuji Techno System, Japan, 77–90 [In Japanese].