



## Adsorption of PM2.5 emissions from laser printer by paper waste aerogel

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### Abstract

Paper waste, rich in cellulose fibers have become recycled material for pollutant adsorption. This work investigated the feasibility of using aerogel produced from paper waste to capture particulate matter (PM2.5) particles emitted from laser printer devices during their printing job. This study also determined the adsorption behavior using an appropriate isotherm model in the batch process. The preparation conditions for adsorbent aerogel included 3 types of paper waste (2-used sided paper; 2P, color and newspaper), the pretreatment process and production procedures for waste paper aerogel (WPA) and waste paper/chitosan aerogel (WP-CSA). Experiments were conducted in a simulated chamber under actual printing environmental conditions. The results showed that (1) the paper-based aerogels were capable of capturing particle emissions from the printing, (2) the pretreatment process played a significant role in adsorption capacity and (3) the aerogel obtained from 2P without treatment and WPA process showed the best removal efficiency, up to 12  $\mu\text{g}/\text{m}^3$  or 63.2% compared with baseline. At equilibrium, this adsorption process obeyed the Freundlich model with an  $R^2$  value of 0.83. This implied that the adsorption sites on the aerogel surface were heterogeneous in nature and presented a strong interaction between PM2.5 and adsorbent fibers. They revealed a maximum adsorption capacity of 0.002 mg/g. Overall, this conversion, an environmentally friendly alternative to reuse paper waste, showed excellent adsorption capacity and reduced health risks from exposure to PM2.5 from laser printer devices.

**Keywords:** Adsorption, Aerogel, Laser printer, Paper waste, PM2.5 removal

### 1. Introduction

Modern society trends lead people to spend more than 90% of their daily life indoors resulting in a growing demand for fresh, healthy, and comfortable indoor environments [1]. Increasing concerns regarding indoor air quality (IAQ) have included monitoring indoor air pollutants affecting the health status and working performance among office workers [2]. Many related studies have reported that health complaints and poor IAQ were statistically associated [3-5]. One general parameter considered is inhaled particulate matter (PM) due to its adverse impacts on human health mechanisms and being easily monitored [6-8]. PM is a mixture of solid particles and liquid droplets suspended in air. Sources of PM indoors include cleaning processes, walking and playing activities, furnishings, IT equipment and electronic devices [9]. It varies widely in size defined by a diameter of 10 microns or less (PM10) and 2.5 microns or less (PM2.5). Because the smaller sized PM2.5 can affect the lower respiratory system, its toxicology and epidemiology have widely been considered. Some researchers have reported that PM2.5 was one of the causative factors of human nonaccidental death and damage to the human respiratory system [10].

Laser printer devices have been widely used in daily life in many places including schools, offices and other indoor environments [11]. Generally, printing operations use a photosensitive drum to attract toner powder and

fuse it on a page using pressured and heated rollers. Tang et al. (2011) [12] studied the emission rate of fine and ultrafine particles including PM2.5 and made the comparison between standby, printing and working phases of  $22.0 \pm 13.0$ ,  $27.0 \pm 13.0$ , and  $36.0 \pm 27.0 \mu\text{g}/\text{m}^3$ , respectively. In principle, this finding showed a potential risk from exposure to PM2.5 emitted from laser printers. Similar to case reports on cultured cells using a mouse model, exposure to PM2.5 from a printing room significantly increased inflammation, fibrosis, apoptosis and an abundance of pathogenic bacteria [13]. Therefore, it could be said that the fine PM components create a potential threat to individuals spending time in printing rooms and significantly degrades IAQ [14].

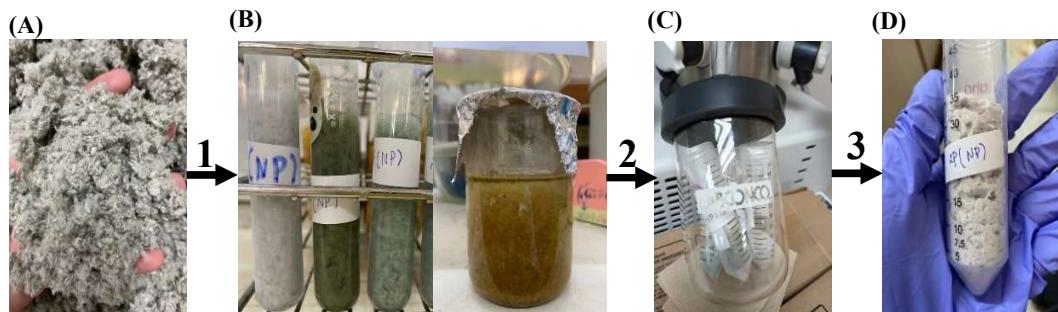
Adsorbent aerogel is a light, porous material exhibiting many excellent properties in adsorption efficiency including transparency, extremely high porosity, large surface area and high mechanical strength. Due to the advantages of high porosity and surface areas, aerogels were frequently used as adsorptive materials for various purposes both in aqueous and gaseous phases [15,16]. They can be produced from many cellulose and cellulosic-based materials including paper waste. Many researchers have focused their experiments on aqueous pollutants; however, few have conducted research in the gaseous phase. Li Z, et al [15] researched using untreated office paper waste to make adsorbent aerogel through a conventional wastepaper aerogel (WPA) production process to cleanup oil spills and organic pollution. They reported a low adsorption capacity by direct use of waste paper aerogel. This capacity has been increased by adding chitosan powder to the production process. Their findings indicated chitosan increased adsorption capacity corresponding to several related researchers [17,18].

Paper waste is rich in cellulose fiber posing a considerable adsorption potential concerning pollutants. Recycling and using paper waste can save resources and incorporate an environmentally-friendly approach. This work aimed to study the use of various paper waste materials obtained from offices to remove PM2.5 emitted from laser printers so as to determine their adsorption capacity and behaviors. The results would constitute one method to reduce health risk effects from exposure of PM2.5 emitted by laser printers.

## 2. Material and methods

### 2.1 Adsorbent aerogel preparation

Adsorbent aerogel materials were prepared for batch experiments using a  $3 \times 2 \times 2$  factorial design method. Independent variables included three types of paper waste (2-sided used paper; 2P, color and newspaper), two types of pretreatment processes (pretreatment and without pretreatment) and two methods of aerogel production processes WPA and waste paper/chitosan aerogel (WP-CSA). The details are presented in Figure 1 and described below.



**Figure 1** Production process for adsorbent aerogel (A) paper waste raw material, (B) paper waste fiber after pretreatment, (C) freeze-drying and (D) paper-based aerogel adsorbent.

#### 2.1.1 Materials and chemicals

The 2P, color and newspaper were collected from academic offices and shredded mechanically in small pieces. Chemical agents used in this study included sodium hydroxide (NaOH), hydrochloric acid (HCl), ethanol, urea and chitosan. All chemicals were analytic grade and used as received without further purification unless otherwise stated. Deionized water (DI) was used throughout the whole process of experiments.

#### 2.1.2 Paper waste pretreatment process

In the experiments, paper waste samples were categorized to be samples with pretreatment (P) and without pretreatment (NP). For the pretreatment process, paper wastes were dispersed in a 0.5 M NaOH solution (1:20 w/v) at room temperature for 4 h in permanent agitation on a 200 rpm shaker. Then they were washed with

distilled water and soaked in 0.5 M HCl solution (1:20 w/v) for 4 h at a stirring speed of 200 rpm [19]. Pretreated paper waste was further repeatedly washed with distilled water and dried at a temperature of 40°C for 48 h [20].

### 2.1.3 Aerogel production process

To prepare WPA, 4 g paper waste was poured in 100 mL of NaOH/urea solution and stirred at 1000 rpm for 10 min at room temperature [15]. Then 100 mL 0.5 M ethanol was added to the mixture and dried at room temperature for 24 h. A homogenous mixture was obtained and further washed with DI water until pH = 7. After that, the mixture was freeze dried to generate aerogel. WPA was obtained after freeze drying for 48 h [21].

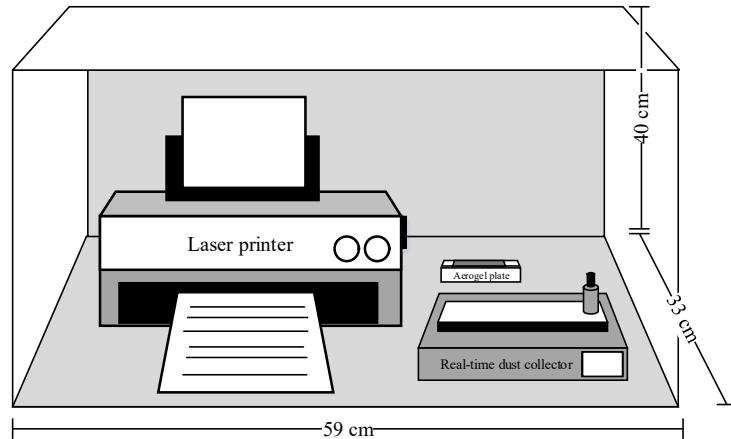
Similar to WPA, 100 mL NaOH/urea aqueous solution (7 wt%: 12 wt%) was kept at -20°C for 1 h. Then chitosan was added to the solution and stirred at 1000 rpm at room temperature. After 30 sec, 4 g of paper waste was added and stirred 10 min to form a homogeneous mixture. Stable hydrogel was washed with DI water until pH = 7. WP-CSA was obtained after freeze drying for 48 h.

### 2.2 Morphology characterizations

Aerogel morphologies were investigated using scanning electron microscopy (SEM, FEI- Quanta 250). Before testing, the samples were kept dry before SEM. Then cross-sections were sputter-coated with a thin layer of platinum before observing.

### 2.3 Batch adsorption experiments

Adsorbent aerogel materials were prepared for batch experiments using a 3×2×2 factorial design method. Independent variables included three types of WP (2P, color and newspaper), two types of pretreatment processes (P and NP), and two types of aerogel production processes (WPA and WP-CSA). Gaseous particle samples were collected using a study chamber described in Figure 2. The inner volume was about 0.08 m<sup>3</sup> (59 cm × 33 cm × 40 cm). This reactor was constructed using cardboard material. During experiments, the average temperature and humidity were measured in a range from 25±2°C to 38 to 40%, respectively. Each test was performed using a laser printer (Brother HL-2130 series) with a printing speed of 20 pages/min and about continuous 30 pages in a printing process. The DustTrak II Aerosol Monitor with a PM2.5 selective cyclone was employed after printing 10 min to monitor the real time mass concentration of PM2.5 by a flow rate of 1.52 L/min. All experiments were conducted in triplicate.



**Figure 2** Experimental setup.

### 2.4 Calculations

#### 2.4.1 Removal efficiency

The removal efficiency of PM2.5 by adsorbents was calculated from adsorption percentage (RE, %) and calculated using Equation 1.

$$RE (\%) = \frac{X_0 - X_1}{X_0} \times 100 \quad (1)$$

where  $X_0$  is the concentration of PM2.5 concentration without aerogel adsorbent added ( $\text{mg}/\text{m}^3$ ) and  $X_1$  is the residual concentration of PM2.5 after contact with the aerogel adsorbent ( $\text{mg}/\text{m}^3$ ) [22,23].

#### 2.4.2 Adsorption capacity

To evaluate the removal efficiency, at equilibrium, the adsorption capacity equation was employed and calculated according to Equation 2.

$$Q_t = \frac{(C_0 - C_e)V_d}{M_b} \quad (2)$$

where  $Q_t$  is the concentration of PM2.5 adsorbed onto the surface of aerogel adsorbents ( $\text{mg}/\text{g}$ ),  $C_0$  is the initial or background concentration of PM2.5 in the experimental setup ( $\text{mg}/\text{m}^3$ ),  $C_e$  is the PM2.5 concentration at equilibrium ( $\text{mg}/\text{m}^3$ ),  $V_d$  is the volume of solution and  $M_b$  is the amount of an adsorbent [24-26].

#### 2.4.3 Adsorption isotherms

To describe the relationship between the number of pollutants adsorbed by the adsorbents and the remaining pollutant concentration in the experiments at equilibrium, adsorption isotherm studies were conducted using the same initial PM2.5 concentration. The appropriate adsorbent was tested for its ability to adsorb the PM2.5 concentration. Results were plotted in two-parameter sorption isotherm models, i.e., the Langmuir and Freundlich models. The Langmuir isotherm was used to determine the adsorption of PM2.5 on the surface of the adsorbent on the monolayer and equivalent sites on the surface while the latter model was based on multilayer adsorption with a heterogeneous surface [27,28]. They were expressed mathematically using Equations 3 and 4, respectively.

$$\frac{C_e}{q_e} = \frac{1}{q_{\max}K_L} + \frac{C_e}{q_{\max}} \quad (3)$$

$$q_e = \left(\frac{1}{n}\right) \ln C_e + \ln K_F \quad (4)$$

where  $q_{\max}$  is the adsorption capacity of the adsorbent at equilibrium ( $\text{mg}/\text{g}$ ) in the homogeneous layer,  $K_L$  is the Langmuir adsorption constant,  $C_e$  is the equilibrium concentration of PM2.5 ( $\text{mg}/\text{m}^3$ ),  $K_F$  is the Freundlich equilibrium constant, and  $n$  is an affinity constant between the adsorbates and the adsorbents. In the sense of control, when  $n$  is greater than one implies stronger interaction between an adsorbent and metallic ion [22,29,30].

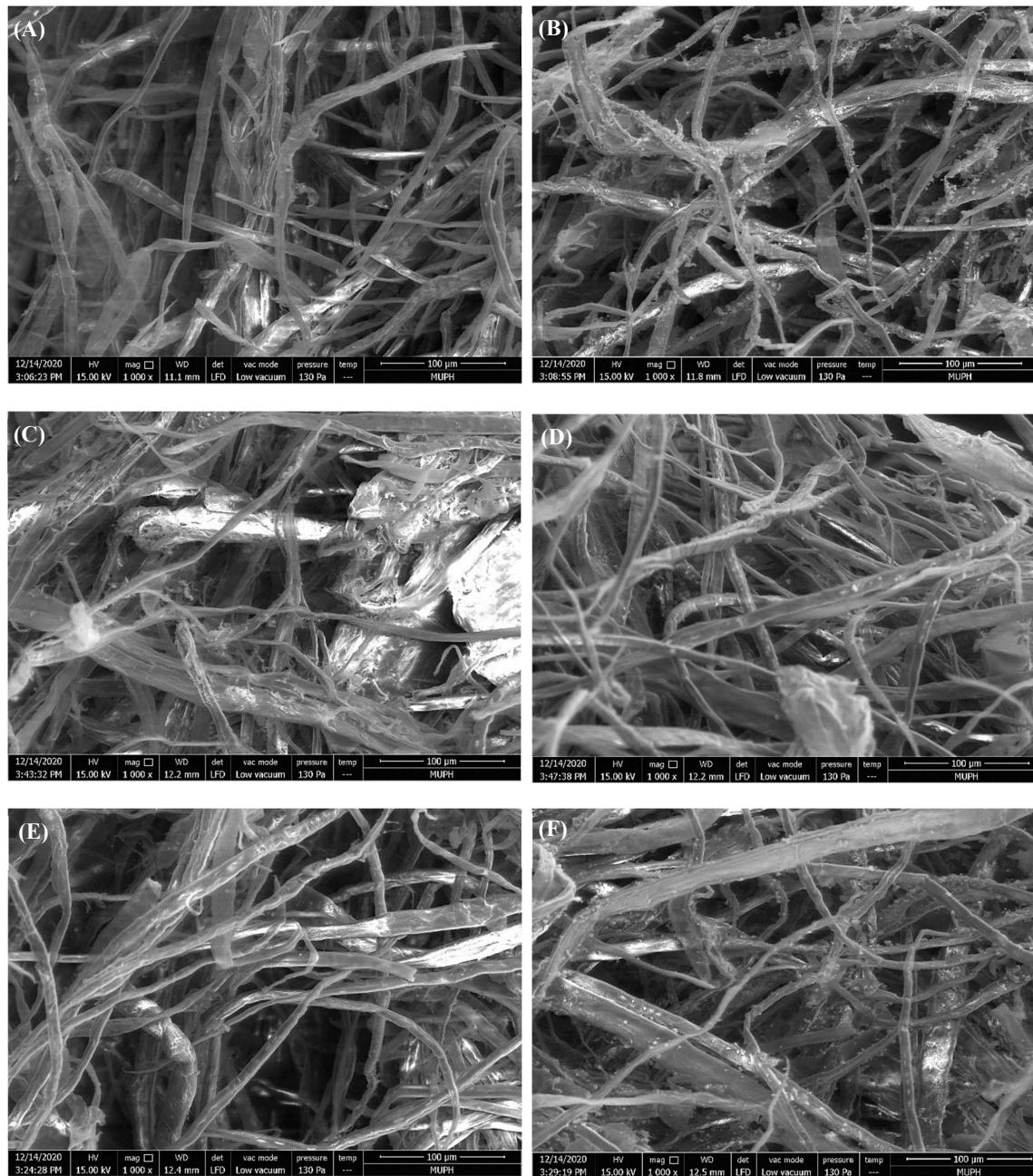
#### 2.5 Data analysis

Descriptive statistics including central tendency, variation and percentiles were used to calculate all parameters using Excel spreadsheet. To investigate the different associations of adsorption capacity and aerogel preparation methods, the analysis of variance (ANOVA) with a significance level of 95% Confidence Interval (CI) was evaluated. All statistical analyses were performed using Statistical Package for Social Sciences (SPSS, Version 18.0, SPSS Ltd., USA).

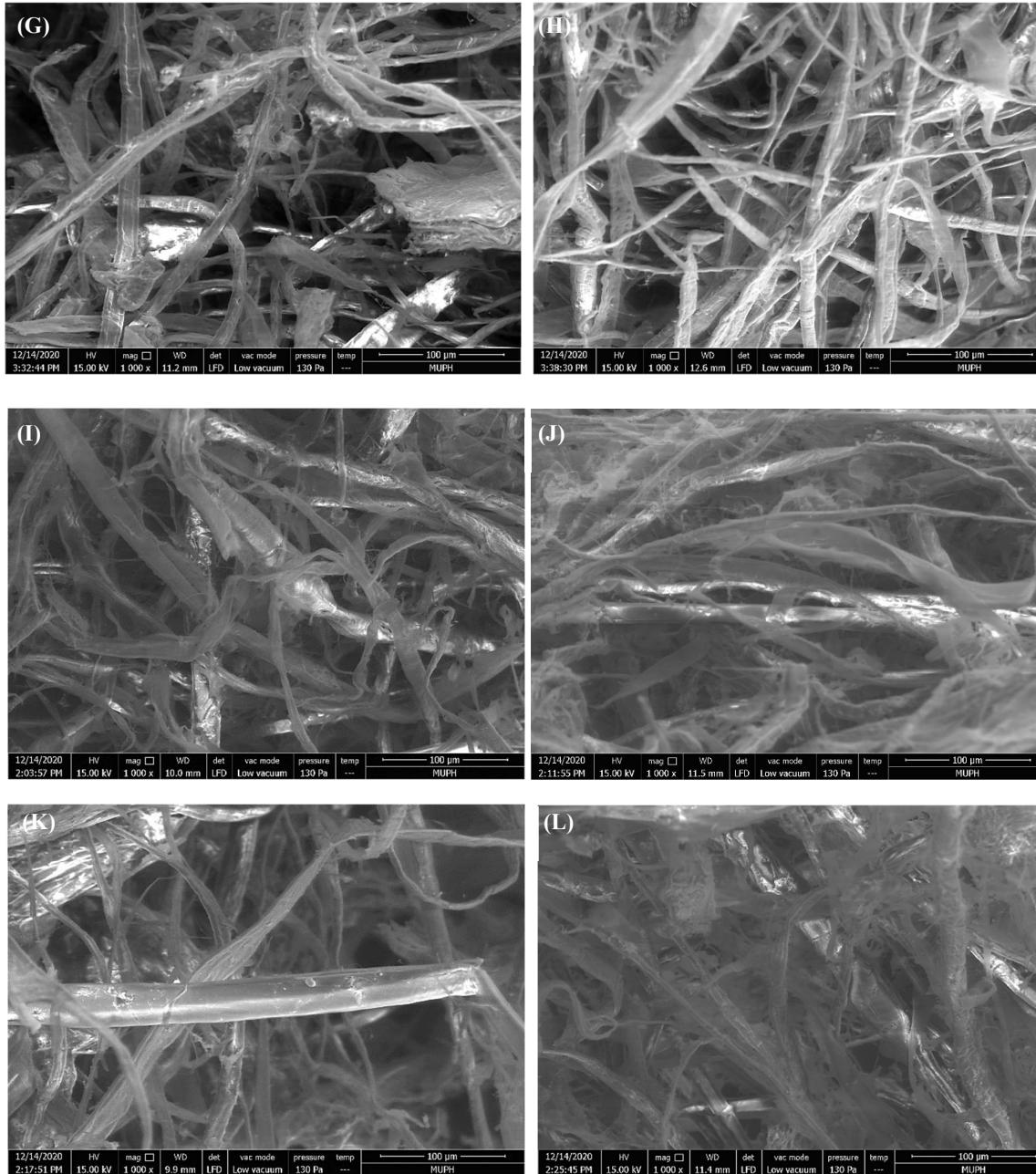
### 3. Results and discussion

#### 3.1 Characterizations of aerogel

The microstructures of aerogel adsorbents produced from paper waste are shown in Figure 3. Each experiment showed various fiber sheet structures in the lateral fiber dimension. The dimension network structure presented a pore size diameter from 33 to 58  $\mu\text{m}$  as detailed in Table 1. Related research reported that the appearance of lamellas becomes the key difference for WPA and WP-CSA methods. The study of Li Z, et al [15] described that increasing chitosan content may raise the lamellas and enhance mechanical strength, acid resistance, and high adsorption capacity. Conversely, results in this study cannot clarify lamellas occurrence; and further, the similar adsorption capacity among the two production methods found is discussed in the next section.



**Figure 3** SEM micrographs of aerogel adsorbents (1,000x magnification) (A) 2P/P/WPA; (B) 2P/NP/WPA; (C) 2P/P/WP-CSA; (D) 2P/NP/WP-CSA; (E) Color/P/WPA; (F) Color/NP/WPA.



**Figure 3 (Continued)** SEM micrographs of aerogel adsorbents (1,000x magnification) (G) Color/P/WP-CSA; (H) Color/NP/WP-CSA; (I) Newspaper/P/WPA; (J) Newspaper/NP/WPA; (K) Newspaper/P/WP-CSA; and (L) Newspaper/NP/WP-CSA.

### 3.2 PM2.5 removal

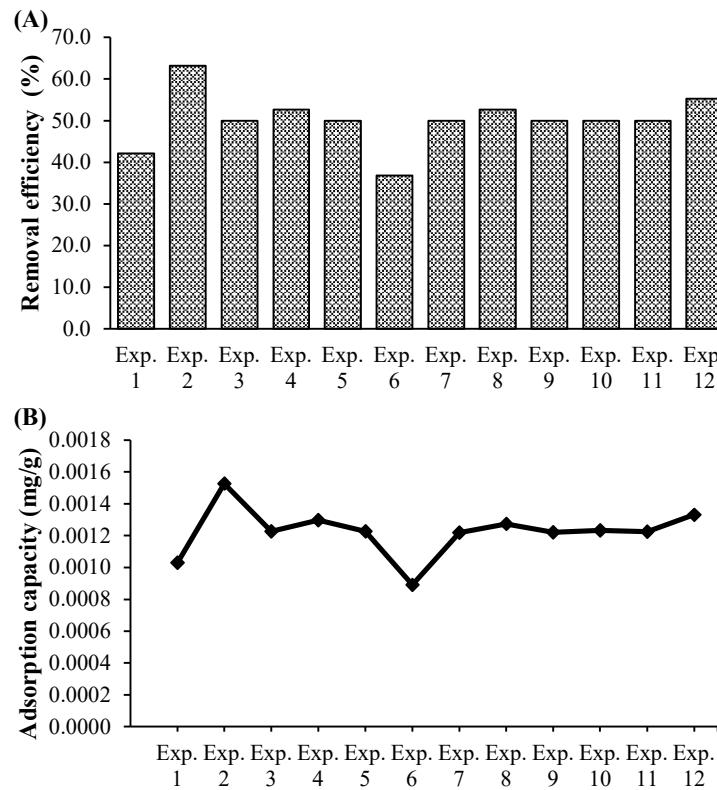
The concentration of PM2.5 emitted from the laser printer was  $19.0 \pm 4.9 \mu\text{g}/\text{m}^3$ . The result corresponded to the study of [11] reporting the average PM2.5 concentration emitted from laser printer was  $23.3 \pm 4.5 \mu\text{g}/\text{m}^3$ . Figure 4 shows the adsorption performance of aerogel adsorbents including adsorption capacity and removal efficiency. The adsorption capacities of aerogel adsorbents were 0.89 to  $1.53 \mu\text{g}/\text{g}$  while the values of removal efficiency ranged from 36.8 to 63.2%. The adsorption capacity value of these adsorbents was quite low compared with other studies due to the different focused boundaries on the gaseous phase of indoor air pollutants. Most related studies were conducted using aqueous media including groundwater, wastewater, and contaminated water [21,31]. The adsorption of PM2.5 on aerogel adsorbents produced from waste color paper

and newspaper revealed a comparable value about 9.1 to 12.1  $\mu\text{g}/\text{m}^3$  while the higher adsorption capacity was carried out by 2P-paper waste as shown in Table 1. Notably, applying 2P with the condition shown in experiment 2 presented the best result of final PM2.5 concentration (7.1  $\mu\text{g}/\text{m}^3$ ). Therefore, only the aerogel adsorbent from Exp. 2 was further used in the next experiment and discussed in section 3.3.

**Table 1** Final concentration of PM2.5 in printing job after experimental processes.

Items	Aerogel preparation			Pore diameter ( $\mu\text{m}$ )	PM2.5 concentration ( $\mu\text{g}/\text{m}^3$ )
	Paper waste	Pretreatment	Methods		
Baseline	-	-	-	-	19.0 $\pm$ 4.9
Exp. 1	2P	P	WPA	46.95	11.2 $\pm$ 6.9
Exp. 2	2P	NP	WPA	33.01	7.1 $\pm$ 0.0
Exp. 3	2P	P	WP-CSA	54.82	10.2 $\pm$ 4.9
Exp. 4	2P	NP	WP-CSA	39.05	9.3 $\pm$ 6.1
Exp. 5	Color	P	WPA	36.31	10.2 $\pm$ 4.6
Exp. 6	Color	NP	WPA	41.82	12.1 $\pm$ 0.0
Exp. 7	Color	P	WP-CSA	46.26	10.0 $\pm$ 6.1
Exp. 8	Color	NP	WP-CSA	36.01	9.3 $\pm$ 5.9
Exp. 9	Newspaper	P	WPA	38.86	10.1 $\pm$ 4.1
Exp. 10	Newspaper	NP	WPA	54.12	9.9 $\pm$ 4.0
Exp. 11	Newspaper	P	WP-CSA	58.05	10.1 $\pm$ 2.0
Exp. 12	Newspaper	NP	WP-CSA	41.73	9.1 $\pm$ 4.2

P, Sample with pretreatment process; NP, Sample without pretreatment process.



**Figure 4** Adsorption performance of aerogel adsorbents: (A) removal efficiency (RE, %) and (B) adsorption capacity ( $Q_t$ , mg/g).

ANOVA was used to estimate significantly different adsorption capacities from the effect of types of Purified water (PW), production methods and pretreatment processes. Results of ANOVA test are presented in Table 2. No statistically significant differences were found between group means among types of PW and production process. Only the confounding factor of pretreatment process (with and without treatment process) played an important role in increasing adsorption capacity ( $F(4,7) = 4.550$ ,  $p = 0.040$ ). Therefore, the higher adsorption amount and removal efficiency presented in previous sections showed the best practice to produce

aerogel from paper waste without pretreatment process. This finding corresponded to one related study on the absorption performance for oils by cellulose-based aerogels from waste newspaper without any pretreatment exhibiting a good absorption performance for waste engine oil and showed an absorption capacity for chloroform [32].

**Table 2** One-way ANOVA results for adsorption capacity difference in aerogel preparation.

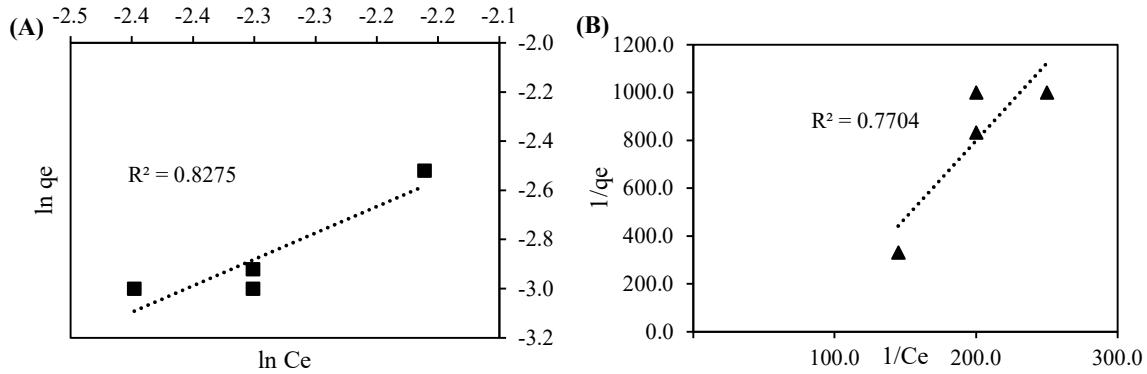
	Sum of squares	df	Mean square	F	Sig.
Types of PW					
Between groups	2.667	4	0.667	0.875	0.524
Within groups	5.333	7	0.762		
Total	8.000	11			
Methods					
Between groups	1.500	4	0.375	1.750	0.243
Within groups	1.500	7	0.214		
Total	3.000	11			
Pretreatment					
Between groups	2.167	4	0.542	4.550	0.040 <sup>a</sup>
Within groups	0.833	7	0.119		
Total	3.000	11			

df, Degree of freedom; F, statistics value (sum of squares/mean square); Sig., significance.

<sup>a</sup> Difference between the two groups was significant at 0.05 level.

### 3.3 Adsorption isotherm

Results from the previous section presented that PM2.5 emitted during printing jobs was highly adsorbed by exp. 2 (2P/NP/WPA). Then to carry out more adsorption behavior, the adsorption isotherms were investigated to describe how PM2.5 interacted with this adsorbent. The Langmuir and Freundlich isotherm models were applied to explain about the two-parameter sorption conditions. Figures 5A and B show the linearized tendency obtained by Freundlich and Langmuir models, respectively. Values of the adsorption and the correlation coefficient are presented in Table 3. The isotherm experiment revealed the Freundlich isotherm fitted well with the correlation coefficient value ( $R^2$ ) of 0.83. It could be said that the adsorption of PM2.5 on aerogel adsorbent involved multilayer adsorption with active sites distributed unevenly on the surface of the adsorbent [16,33]. As Freundlich isotherm was valid for heterogeneous surface media, the high KF value represented the greater adsorption capacity of the adsorbent [34,35]. Therefore, the high value of 1/n or adsorption intensity (2.15) also confirmed that they represented a strong interaction between PM2.5 and aerogel. However, based on the plotting of 1/qe against 1/Ce values, the Langmuir model offered the maximum adsorption capacity ( $q_m$ ) of 0.002 mg PM2.5/g aerogel.



**Figure 5** Adsorption isotherm: (A) Freundlich and (B) Langmuir models.

**Table 3** Isotherm parameters for PM2.5 adsorption onto aerogel adsorbents.

Isotherm model	Estimated isotherm parameters				
	$q_{max}$ (mg/g)	$K_L$	$K_F$	$n$	$R^2$
Langmuir	0.002	77.16	-	-	0.77
Freundlich	-	-	7.85	0.47	0.83

#### 4. Conclusion

Paper waste could be adequate adsorbent material. Four types of paper wastes were used as raw material to produce aerogel adsorbent, namely, 2P, color and newspaper. From this finding, the removal efficiency and adsorption capacity were found from 36.8 to 63.2% and 0.89 to 1.53  $\mu\text{g/g}$ , respectively. The condition of 2P, without any pretreatment, and WPA method revealed a peak of adsorption capacity and presented a final PM2.5 concentration of 0.007  $\text{mg/m}^3$  compared with baseline (0.019  $\text{mg/m}^3$ ). Moreover, the ANOVA test was employed to find the confounding factors affecting efficiency. Results showed that the pretreatment process (with and without treatment) played an important role in increasing adsorption capacity ( $p<0.05$ ). At equilibrium, the adsorption isotherm models of Freundlich and Langmuir were employed to explain the adsorption behaviors and mechanisms. Results showed that the adsorption experiments fitted well with the Freundlich model with correlation coefficient ( $R^2$ ) of 0.83, while that of Langmuir was found to be 0.77. Optimum capacity was found to be 0.002  $\text{mg/g}$ . This implied that the adsorption site on aerogel surface was heterogeneous in nature and presented a strong interaction between PM2.5 and aerogel. Thus, the use of aerogel adsorbents could help alleviate health risk effects from exposure to PM2.5 emitted by laser printers. Moreover, the study of the use of synthesized adsorbent aerogel from paper waste and other residues will offer several benefits for emission control in working area environments and reduce the risk of exposure to indoor air pollutants.

#### 5. Ethical approval

This study protocol followed the principles of the Declaration of Helsinki and the Ethics Committee for Research Involving Human Subjects (COA. No. 185/2563).

#### 6. Acknowledgements

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