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Benzene Adsorption by Plant Leaf Materials: Effect of Quantity and Composition of Wax

Chairat Treesubsuntorn · Parinda Suksabye · Sawitree Weangjun · Fonthip Pawana · Paitip Thiravetyan

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Abstract Twenty-one plant leaf materials were screened for benzene adsorption efficiency in the static system, and the leaf material from *Dieffenbachia picta*, *Acrostichum aureum*, *Ficus religiosa*, *Lagerstroemia macrocarpa*, *Alstonia scholaris*, and *Dracaena sanderiana* were found to have high potential for benzene removal. The relation between quantity and composition of wax to benzene removal efficiency was studied. Although high quantities of wax occurred in some leaf materials, low benzene removal was clearly found if compared with other plant materials with the same wax quantity. Alpha-linoleic acid and dodecyl cyclohexane were found to be the main composition in plant leaf materials with high benzene adsorption, and it might be a key factor for benzene removal.

Keywords Plant leaf material · Cuticular wax · Benzene

1 Introduction

Benzene, a volatile organic compound (VOC), is extracted from petroleum industries and widely used as an additive, an intermediate, and/or a solvent in many manufacturing industries. Nowadays, the emission of benzene from many sources can cause ambient air pollution problems, even though several organizations and countries have standard guidelines on ambient benzene concentrations (Pollution Control Department 2007; World Health Organization 2000). Benzene concentration in the atmosphere was found to be higher than the local standard guidelines in many places around the world. In addition, 9 cohort and 13 case–control studies confirm that benzene can clearly induce acute myelogenous leukemia and other cancers (Schnatter et al. 2005). From many researches, benzene is classified in the 1A group, which is composed of high potential carcinogens in the human body by IARC (Guieysse et al. 2008). Allergies, asthma, dizziness, tremors, restlessness, eye irritation, and nervous-system disorders were also known to be caused by benzene (Wolverton 1996). Benzene can also be accumulated and stablized in the environment. Benzene removal by plants was studied, and strong evidence showed that some species of plants could uptake benzene well (Liu et al. 2007; Orwell et al. 2004; Treesubsuntorn and Thiravetyan 2012; Wolverton et al. 1989). Ugrekhelidze et al. (1996) reported that plants could uptake gaseous benzene through the stomata and wax on the surface of the leaf. In addition, from a previous study, although the plants were grown under dark conditions, the plants could still grow and uptake benzene only through cuticular wax because the stomata was closed under dark conditions (Orwell et al. 2004; Treesubsuntorn and Thiravetyan 2012). The benzene accumulation in cuticular wax of plant leaves was found in many researches (Collins et al. 2000; Environmental Agency 2009; Gorna-Binkul et al. 1996; Kylin et al. 1994; Poborski 1988; Riederer 1990; Slaski

C. Treesubsuntorn · P. Thiravetyan (✉)
School of Bioresources and Technology, King Mongkut's University of Technology Thonburi,
Bangkok 10150, Thailand
e-mail: paitip.thi@kmutt.ac.th

P. Suksabye · S. Weangjun · F. Pawana
Department of Urban and Industrial Environmental Management, Science and Technology Faculty, Suan Dusit Rajabhat University,
Bangkok 10300, Thailand

et al. 2000; Tsiros et al. 1999). Benzene that contains 2.13 of log Kow, a logarithm value of octanol/water partition coefficient, can transport easily into the plant (Kamath et al. 2004). Nowadays, activated carbon was widely applied to treat benzene however high cost for control, and secondary waste disposal was a problem. The use of plant leaf material for benzene adsorption was of interest as a low-cost adsorbent. However, a few studies reported on the application of plant leaf material for benzene adsorption. In this present study, 21 plant leaf materials were screened for benzene adsorption in a static system. The composition of wax might be more important than quantity of wax in benzene adsorption (Topp et al. 1986), so not only new adsorbent from plant leaf material was improved but also the effect of wax quantity and composition of wax of each plant leaf material was analyzed as criteria for the selection of plant materials for benzene adsorption. This research aims to study the application of plant leaf material for benzene removal.

2 Materials and Methods

2.1 Plant Leaf Materials Preparation

Twenty-one plants including *Homalomena rubescens*, *Citrus hystrix*, *Musa paradisiaca*, *Mangifera indica*, *Catura metet*, *Lagerstroemia inermis*, *Cananga odorata*, *Cassia siamea*, *Bougain villea*, *Litchi chinensis*, *Coccinia grandis*, *Dieffenbachia picta*, *Attacus atlas*, *Polyalthia longifolia*, *Acrostichum aureum*, *Ficus religiosa*, *Alstonia scholaris*, *Anthurium andraeanum*, *Plerocarpus indicus*, *Lagerstroemia macrocarpa*, and *Dracaena sanderiana* were purchased from plant shops in Thailand. A leaf of each plant was cut and dried at 60 °C for 2 days. The dry leaves of the plants were powdered by a Retsch ultra centrifugal mill with 14,000 cycles/min and dried again at 60 °C for 2 days. Each plant leaf material weighing 0.2 g was put in a fumigator for passive benzene adsorption.

2.2 Benzene Removal in a Static System

Glass modified desiccator with 30 cm of height, 12.86 cm of radius, and 15.6 l was applied in the static experiment. Plant leaf materials, each containing about 0.2 g, were placed in modified desiccators with an injection and sampling pot on the top of the chamber under indoor conditions (three repetition chambers in each treatment). Desiccators were closed and sealed by parafilm at the temperature of 32 °C and a pressure of 760 mmHg. The temperature and pressure were used to calculate the mole concentration (M_c) as present in Eq 1:

$$M_c = 24.47 \times \frac{760}{P} \times \frac{T + 273.15}{298.15} \tag{1}$$

$$20\ ppm = 10^6 \times \frac{W}{M_w} \times \frac{M_c}{V} \tag{2}$$

$$\rho = \frac{W}{V_b} \tag{3}$$

where M_c is the mole concentration, P (mmHg) and T (°C) are the pressure and temperature, respectively. The M_W (molecular weight of benzene) and V (volume of the chamber [l]) were substituted to calculate W (benzene weight [g]) using Eq. 2. ρ (benzene density [g/ml]) and the weight of benzene was used to predict V_b (benzene volume [ml]) using Eq. 3. 99 % pure benzene, which was purchased from Panreac (made in E.U.), should be injected at the 1±0.1 μl for 18±2 ppm benzene concentration. In this system, 4 h was required to achieve benzene equilibrium. In a previous study, 3 days was required to saturate benzene in 0.2 g plant leaf material, so a sample of 0.3 ml benzene contaminated gas in the chamber was collected by gastight syringe and directly injected and analyzed by gas chromatography (GC) every 24 h for 3 days.

2.3 Crude Wax Extraction for Quantity Analysis

For the extraction of hydrophobic wax, 1 g of 21 plant leaf materials was immersed in 50 ml of hexane at 4 °C overnight. The extracted solution was filtrated by Whatman number 2 filtration paper. After the filtration method, nitrogen was applied for hexane evaporation on the ceramic cup. Four decimal balances were used to measure the weight of the crude wax.

2.4 Wax Composition Analysis

Eight plant leaf materials including *D. picta*, *A. aureum*, *C. siamea*, *A. scholaris*, *D. sanderiana*, *L. Macrocarpa*, *P. longifolia*, and *M. paradisiaca* were selected because these leaf materials showed some interesting characteristics. For example, *D. picta*, *A. aureum*, *A. scholaris*, *D. sanderiana*, and *L. Macrocarpa* have high benzene removal potential and high weight of wax, but *M. paradisiaca* has high benzene removal potential although low weight of wax was found. For the *P. longifolia*, low quantity of wax and low benzene removal efficiency were found, but *C. siamea*, with high wax weight, has low benzene adsorption. These plant leaf materials were selected in order to study the relation between benzene uptake and wax composition. These plant leaf materials were extracted by hexane chilling of the plant material. Pure cuticular wax (10 mg) was immersed in 10 ml of hexane for cuticular wax dilution. N,O-Bis(trimethylsilyl)trifluoroacetamide (BSTFA) and 1 % trimethylchlorosilane (TMCS) were used for cuticular wax solution derivatization (Beck and Lynn 1997). 0.5 μl of the sample was applied as the injection volume. GC-mass spectrometry (MS) was used to analyze the composition of wax by a 30 m DB5 capillary column temperature programmed at 80–250 °C and the split mode of 1:100. The electron impact mode (70 eV) and 30–550 atomic mass units scannings were conditioned for mass analysis.

2.5 Gas Analysis

The GC (Broker, UK) was used to analyze benzene concentrations. Benzene was measured by a 105-m length and 0.53 mm ID column. The diphenyl/dimethyl polysiloxane phase was filled as the stationary phase. N_2 was used as a carrier gas. The FID cylindrical electrode detector with the detection limit of 3×10^{-12} g/s and the dynamic range of 10^7 was used. The benzene uptake by biomaterials from the plant was calculated with the following equations:

$$\Delta ppm = ppm_{control} - ppm_{treatment} \tag{4}$$

$$W = \Delta ppm \times \frac{V}{M_c} \times \frac{M_w}{10^6} \tag{5}$$

Where $ppm_{control}$ and $ppm_{treatment}$ are the remaining benzene concentrations in the control and treatment systems, respectively. The differences between $ppm_{control}$ and $ppm_{treatment}$ were used as the benzene adsorption by

Table 1 Benzene adsorption efficiency in 3 days of the experiment by various leaf materials

Plant species	Benzene adsorption efficiency (μ mole/g of adsorbent)		
	Day 1	Day 2	Day 3
Homalomena rubescens	0.92 ± 0.01^{a}	0.94 ± 0.02^{a}	1.10 ± 0.28^{a}
Citrus hystrix	$7.64\pm0.33^{d,e}$	$8.66\pm8.70^{c,d,e,f}$	$9.19\pm2.63^{b,c}$
Musa paradisiaca	$9.01\pm1.99^{d,e,f}$	$15.20\pm3.29^{g,h}$	15.05 ± 1.14^{e}
Mangifera indica	$7.67\pm3.45^{d,e}$	$7.56\pm3.54^{b,c,d,e}$	$6.79\pm3.05^{b,c}$
Catura metet	1.42 ± 0.19^{a}	$3.75\pm0.52^{a,b}$	3.58 ± 0.04^{a}
Lagertroemia inermis	$3.80\pm0.16^{a,b,c}$	$6.25\pm3.63^{b,c,d}$	9.45 ± 0.88^{c}
Cananga odorata	$4.67\pm0.23^{b,c}$	$5.59\pm1.32^{b,c}$	6.36 ± 0.24^{b}
Cassia siamea	$3.52\pm0.34^{a,b,c}$	$4.63\pm1.11^{a,b,c}$	$7.66\pm0.17^{b,c}$
Bougain villea	$3.38\pm0.37^{a,b}$	$5.89\pm0.30^{b,c,d}$	$7.06\pm1.17^{b,c}$
Litchi chinensis	$3.29\pm0.59^{a,b}$	$10.94\pm0.25^{e,f,g}$	23.46 ± 2.10^{h}
Coccinia grandis	$4.62\pm1.84^{b,c}$	$4.37\pm2.28^{a,b,c}$	$6.64\pm0.33^{b,c}$
Dieffenbachia picta	$10.73\pm0.25^{f,g}$	15.77 ± 0.17^{h}	$19.37\pm0.25^{f,g}$
Attacus atlas	$3.48\pm0.37^{a,b,c}$	17.49 ± 0.46^{h}	$17.08\pm0.21^{e,f}$
Polyalthia longifolia	$6.34\pm0.91^{c,d}$	$7.98\pm0.44^{b,c,d,e,f}$	9.39 ± 0.51^{c}
Acrostichum aureum	$10.97\pm0.63^{g,h}$	$14.59\pm0.54^{g,h}$	$18.90\pm0.54^{f,g}$
Figus religiosa	$10.61\pm0.12^{f,g}$	$10.53\pm0.41^{d,e,f,g}$	$18.33\pm1.12^{f,g}$
Lagerstroemia macrocarpa	$10.94\pm0.54^{f,g}$	$12.59\pm0.03^{f,g,h}$	20.07 ± 0.88^{h}
Altonis schoris	$9.61\pm0.52^{e,f}$	$14.25\pm0.38^{g,h}$	$20.57\pm1.62^{g,h}$
Anthurium andraeanum	$12.55\pm4.69^{g,h}$	$12.56\pm0.21^{f,g,h}$	$12.41\pm0.46_{d}$
Plerocarpus indicus	13.88 ± 2.52^{h}	16.01 ± 2.81^{h}	$17.82\pm4.35^{f,g}$
Dracaena sanderiana	$11.00\pm4.68^{f,g}$	$16.02\pm1.62^{g,h}$	$19.00\pm2.8^{f,g}$

One-way ANOVA and Duncan multiple range test followed the column group classification for benzene removal efficiency comparison of each plant species at the 95 % confidence level biomaterials from plants (Δppm), and the benzene adsorption by plants was calculated for the weight (W) of benzene adsorption by biomaterial from plants (g) following Eqs. 4–5. V is the volume of the system (l). M_c is the mole concentration of benzene. M_W is the molecular weight of benzene. The weight of benzene uptake per weight of plant leaf material was reported.

2.6 Statistical Analysis

One-way ANOVA, Duncan's multiple range tests and correlation coefficient were used for statistical analysis by SPSS version 19 at 95 % confidant levels.

3 Results

3.1 Benzene Adsorption by Biomaterials in a Static System

Benzene-contaminated air (20 ppm) was used to fumigate 21 plant leaf materials. The remaining benzene concentration was analyzed every day and calculated to the micromole per gram of adsorbents (μmole/g of adsorbents). The result is shown in Table 1. The biomaterials from *L. chinensis*, *D. picta*, *A. aureum*, *F. religiosa*, *L. macrocarpa*, *A. scholaris*, and *D. sanderiana* were found to have high benzene adsorption efficiency at day 3 of the experiment. However, *L. chinensis* showed slow benzene adsorption at day 1. Therefore, the biomaterials from *D. picta*, *A. aureum*, *F. religiosa*, *L. macrocarpa*, *A. scholaris*, and *D. sanderiana* were sustainable for application.

3.2 The Relation Between Benzene Adsorption and Quantity of Wax in Each Plant Leaf Material

The crude wax was extracted by the use of hexane as a solvent and the quantity was measured. Benzene adsorption efficiency in the unit of μmole per gram of adsorbent at day 3 of the static experiment and the quantity of wax in each plant is shown in Table 2. The group classification was separated by the Duncan multiple range test following the column comparison. High benzene adsorption by plant leaf materials was shown in high quantities of wax materials.

For the result, high benzene adsorption was found in high quantities of wax materials, as shown in Fig. 1. Each graph was classified following the increasing wax quantity that was grouped by Duncan multiple range tests. Plant leaf materials that occurred in the same graph were shown to have nonsignificant differences in wax weight. The increasing quantity of wax was considered to be one of the factors affecting benzene adsorption, but the same wax quantity group still showed a significant difference in benzene adsorption between different plant materials. Statistical correlations of the relationship between benzene adsorption and wax quantity were analyzed, and a logarithmic tendency was found (Fig. 2). Although the *M. paradisiaca* leaf material had low wax quantities, high benzene adsorption was found. In contrast, the *C. siamea* leaf material can adsorb benzene lower than other plant leaf materials that have nonsignificant differences of wax quantity. The result suggests that not only the quantity of

Table 2 Benzene adsorption efficiency by each plant leaf material and their wax weight

Plant species	Wax weight (g/g of absorbent)	Benzene adsorption efficiency (μmole/g of adsorbent) at day 3
Homalomena rubescens	0.01 ± 0.00^{a}	1.10 ± 0.28^{a}
Citrus hystrix	$0.06\pm0.01^{b,c}$	$9.19\pm2.63^{b,c}$
Musa paradisiaca	$0.06\pm0.02^{b,c}$	15.05 ± 1.41^{e}
Mangifera indica	$0.05\pm0.00^{b,c}$	$6.79\pm3.05^{b,c}$
Catura metet	0.01 ± 0.00^{a}	3.58 ± 0.04^{a}
Lagertroemia inermis	$0.05\pm0.00^{b,c}$	9.45 ± 0.88^{c}
Cananga odorata	0.04 ± 0.01^{b}	6.36 ± 0.24^{b}
Cassia siamea	0.16 ± 0.04^{d}	$7.66\pm0.17^{b,c}$
Bougain villea	$0.05\pm0.00^{b,c}$	$7.06\pm1.17^{b,c}$
Litchi chinensis	0.21 ± 0.02^{e}	23.46 ± 2.10^{h}
Coccinia grandis	$0.05\pm0.00^{b,c}$	$6.64\pm0.33^{b,c}$
Dieffenbachia picta	0.15 ± 0.01^{d}	$19.37\pm0.25^{f,g}$
Attacus atlas	0.10 ± 0.00^{d}	$17.08\pm0.21^{e,f}$
Polyalthia longifolia	0.07 ± 0.01^{c}	9.39 ± 0.51^{c}
Acrostichum aureum	0.10 ± 0.00^{d}	$18.90\pm0.54^{f,g}$
Figus religiosa	0.10 ± 0.00^{d}	$18.33\pm1.2^{f,g}$
Lagerstroemia macrocarpa	0.18 ± 0.03^{e}	20.48 ± 0.88^{h}
Altonis schoris	0.11 ± 0.01^{d}	$20.57\pm1.62^{g,h}$
Anthurium andraeanum	0.14 ± 0.02^{d}	12.41 ± 0.46^{d}
Plerocarpus Indicus	0.10 ± 0.02^{d}	$17.82\pm4.35^{f,g}$
Dracaena sanderiana	0.25 ± 0.00^{e}	$19.00\pm2.8^{f,g}$

One-way ANOVA and Duncan multiple range test followed the column group classification for benzene removal efficiency comparison of each plant species and wax quantity group classification at the 95 % confident level

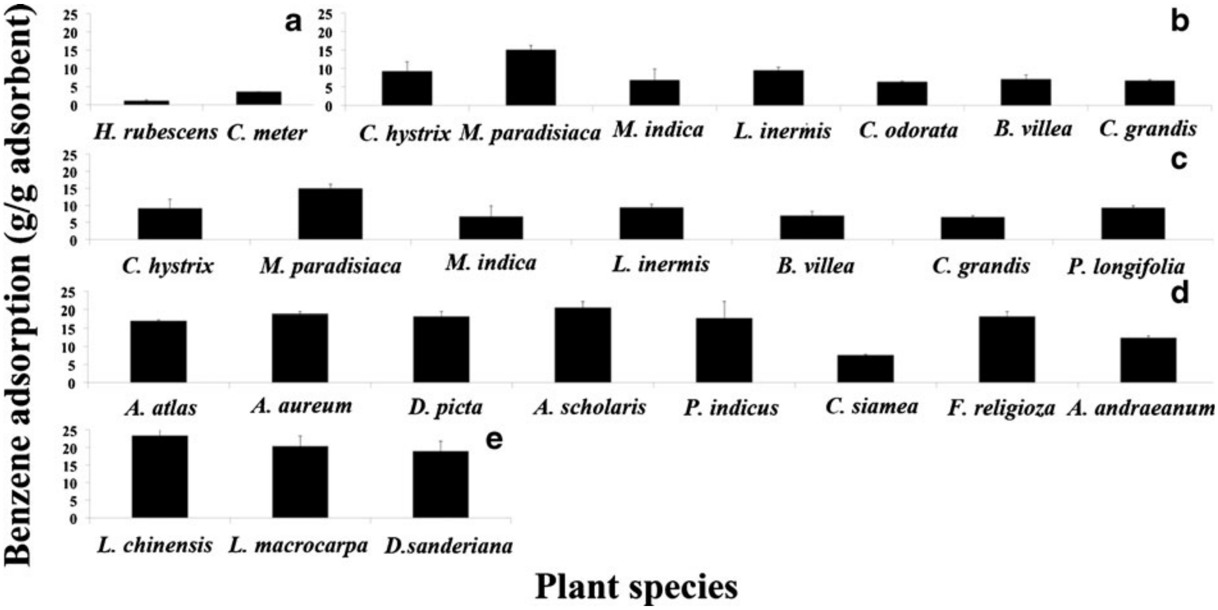


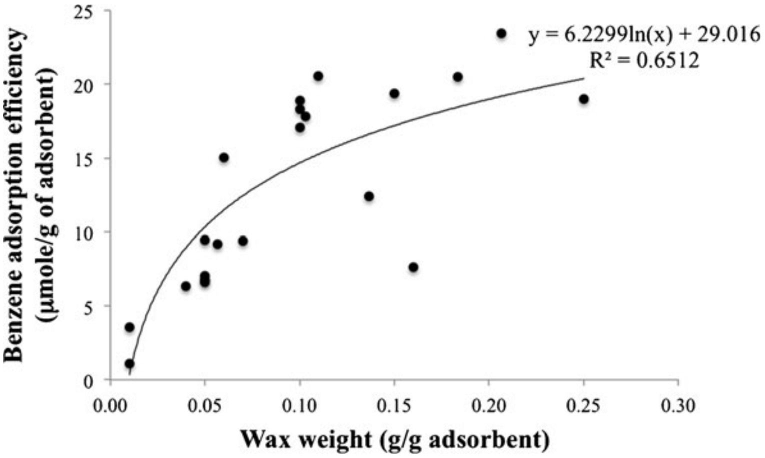
Fig. 1 Comparison of benzene adsorption efficiencies of plant leaf materials in nonsignificantly different wax quantities (Duncan multiple range test classification): **a** 0.01–0.03 mg/g of absorbent, **b** 0.04–0.06 mg/g of absorbent, **c** 0.05–0.07 mg/g of absorbent, **d** 0.1–0.16 mg/g of absorbent and **e** 0.18–0.25 mg/g of absorbent (average and SD in each dot and error bar, respectively)

wax but also the composition of wax affects benzene adsorption by plant materials.

3.3 The Relation Between Benzene Adsorption and Composition of Wax in Each Plant Leaf Material

The wax composition was analyzed by GC-MS and BSTFA, 1 % TMCS was used for cuticular wax solution derivatization and the composition of fatty acids in each selected plant leaf material is shown in Fig. 3. High alpha-linoleic acid and octadecanoic acid were clearly found in *A. scholaris* leaf materials, and the *A. scholaris* leaf material also showed high benzene removal materials. In the *C. siamea* leaf material, significantly lower benzene removal efficiency was found when compared with other plant leaf materials under the same high wax weight. However, high weights of wax occurred. Alpha-linoleic acid was not found in *C. siamea*, but these fatty

Fig. 2 The logarithmic tendency of relationship between benzene absorption and wax quantity



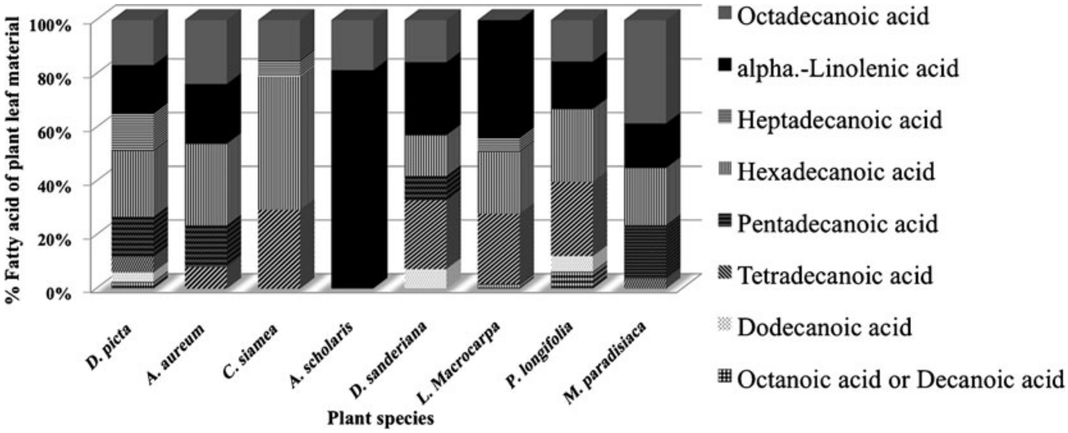


Fig. 3 Percentage of fatty acid composition in the wax of each plant leaf material

acids were also found in other species. On the other hand, the *A. scholaris* leaf material showed high benzene adsorption efficiency and contained high amounts of alpha-linoleic acid and octadecanoic acid. In *D. picta* and *A. aureum* leaf materials, high benzene removal efficiency were also found, and these two materials also contained alpha-linoleic acid and octadecanoic acid. From the results, the plant leaf materials that contained high alpha-linoleic acid had mostly high benzene adsorption efficiency. As shown in (Fig. 4a.), 0.44 of r^2 was found in the statistical correlations of % linoleic acid and benzene adsorption. In addition, the statistical correlations of octadecanoic acid and benzene adsorption have been shown very low r^2 of 0.040 (Fig. 4b).

The percent of alpha-linoleic acid and other fatty acid composition showed that the materials from *A. scholaris* that contained high quantities of wax also had high benzene removal efficiency. In addition, high alpha-linoleic acid composition was found in this species. In the *C. siamea* leaf material, low alpha-linoleic acid was found. So alpha-linoleic acid might enhance benzene adsorption in the plant leaf materials. The percent of alpha-linoleic acid and other fatty acid composition of each material was shown in Fig. 5.

Not only fatty acid but also alkane was observed in Fig. 6. High dodecyl cyclohexane in the *A. scholaris* leaf material was found to be higher than other plant leaf materials. This implied that dodecyl cyclohexane might also be involved in benzene adsorption.

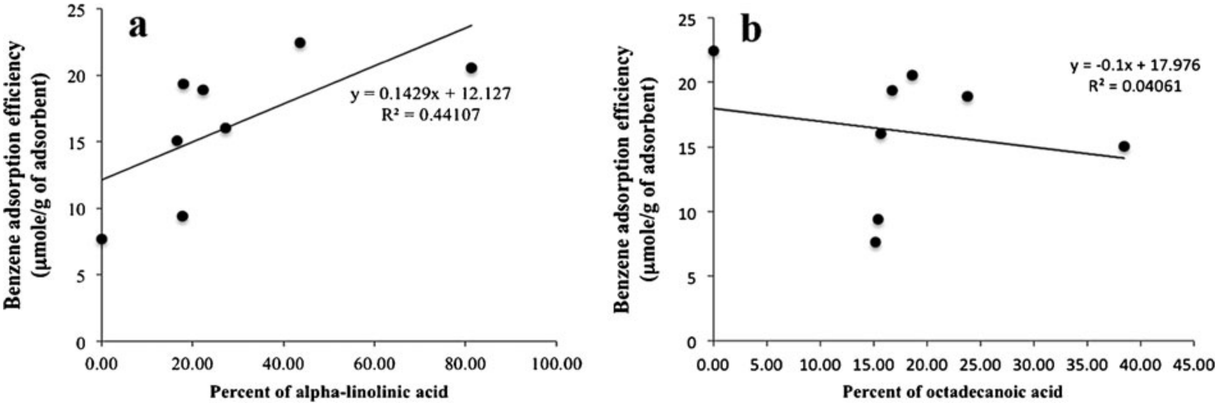


Fig. 4 The statistical correlations of alpha linolinic acid (%) and benzene adsorption (**a**) and octadecanoic acid (%) and benzene adsorption (**b**)

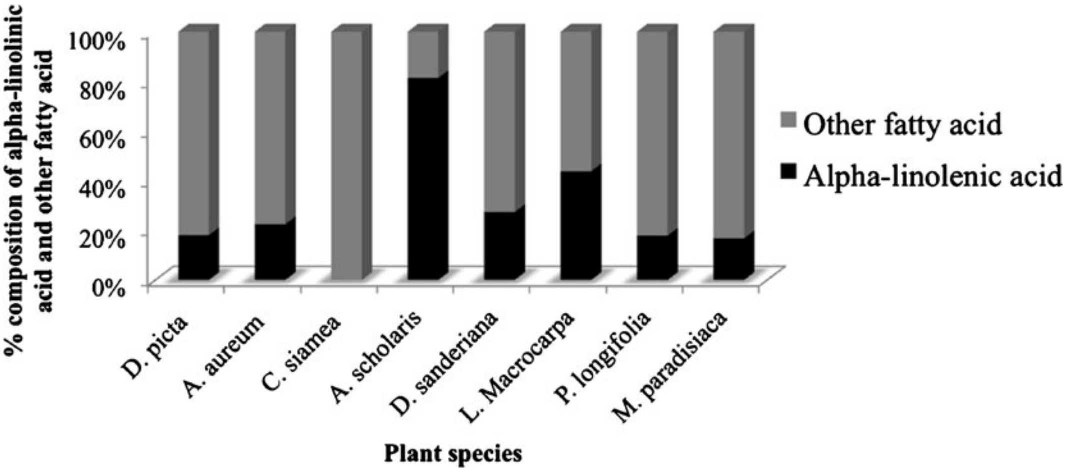


Fig. 5 Percentage of alpha-linolenic acid and other fatty acids composition in each plant leaf materials

The statistical correlations of dodecyl cyclohexane and benzene adsorption have been shown in Fig. 7.

4 Discussions

Gorna-Binkul et al. (1996) found that benzene and its derivatives contaminated orange peels and parsley. Benzene accumulation was also found in blackberries and apples (Collins et al. 2000). Benzene adsorption by avocado fruits was also reported (Jansen and Olson 1969). The theory of Kamath et al. (2004) showed that benzene, which contains 2.13 of log K_{OW} (log scale of octanal solubility per water solubility), could easily transform into the plant. In addition, the Environment Agency (2009) reviewed the VOCs contamination in plants. These results suggested that plants can clearly accumulate VOCs in the leaf, and that the species of plant is an important factor because some species can uptake high benzene, but some species of plant cannot (Binnie et al. 2002; Collins et al. 2000; Ugrekhelidze et al. 1996). Nowadays, a few studies have reported on the application of plant leaf as adsorbent for hydrophobic. In gaseous BTEX, plant leaf material was of interest as a low-cost adsorbent. From the 21 plant leaf materials screenings, *D. picta*, *A. aureum*, *F.religiosa*, *L. macrocarpa*, *A. scholaris*, and *D. sanderiana* leaf materials were found to have high benzene adsorption efficiency. The relation between benzene adsorption and the quantity of cuticular wax was found in logarithmic curve. The result suggested that the increasing of quantity of wax could increase benzene adsorption efficiency, but from the logarithmic curve in the high quantity of wax material group, although quantity of wax increased, benzene adsorption efficiency was stabilized or slowly

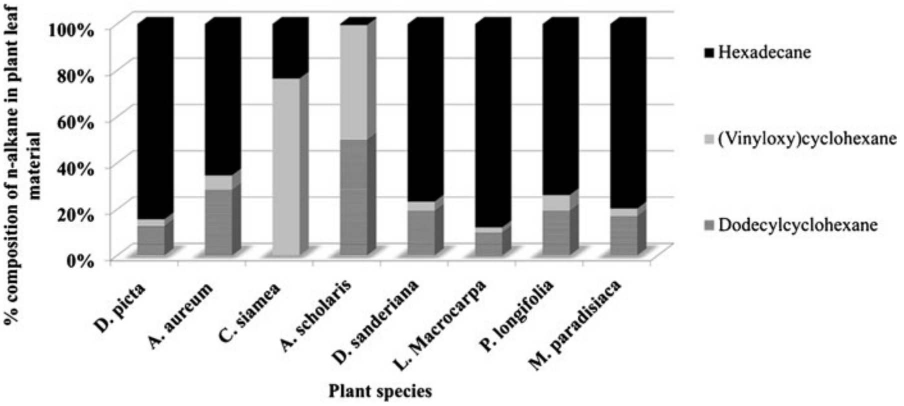


Fig. 6 Percentage of alkane composition in the wax of each plant leaf material

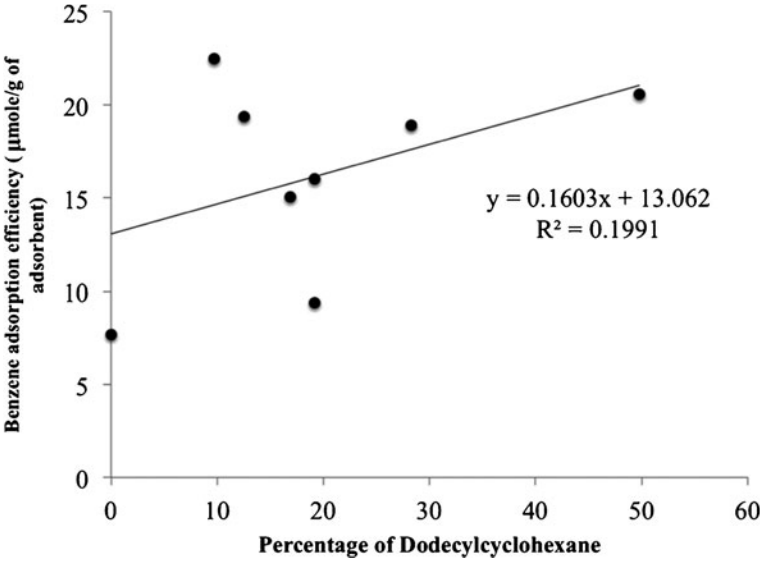


Fig. 7 The statistical correlations of dodecyl cyclohexane and benzene adsorption

increased. The thickness of wax, which was proposed as one of the affecting factors for benzene adsorption by plant leaf (Topp et al. 1986), could be applied to explain this result. Benzene can be adsorbed and transferred in a limiting layer of wax, so high quantity of wax, although thickness of wax was increased, benzene could be only adsorbed in a limitation layer. High benzene removal also occurred, but some species, although they contained high quantities of wax, low benzene removal still appeared. The composition of wax might be a main factor for VOC accumulation (Topp et al. 1986). On the other hand, alpha-linoleic acid was not found in the *C. siamea* leaf material, which had low benzene removal although high wax quantities were found. The increasing number of carbon on the fatty acid molecule can decrease the polar fatty acids and might increase the solubility in benzene (Hoerr and Balston 1994). However, in our study, although octadecanoic acid and alpha-linoleic acid had the same amounts of carbon, the result suggested that only linoleic acid could be affecting factor to increase benzene adsorption. In addition, the *A. scholaris* leaf material, which has high benzene removal efficiency, has higher dodecyl cyclohexane than other plant leaf materials. Therefore, alpha-linoleic acid and dodecyl cyclohexane might be involved in benzene removal. In addition, benzene adsorption by pure fatty acids and dodecyl cyclohexane would be further studied for benzene adsorption.

5 Conclusion

Leaf materials of *D. picta*, *A. aureum*, *F.religiosa*, *L. macrocarpa*, *A. scholaris*, and *D. sanderiana* that were found having high potential for benzene adsorption might be suitable adsorbents for a continuous system. In addition, the quantity and composition of wax were studied to relate with benzene removal efficiency. High quantities of wax material also showed high benzene removal efficiency. However, low benzene adsorption appeared in some plant leaf materials that contained high quantities of wax. On the other hand, although low quantities of wax appeared in some plant leaf materials, high benzene adsorption might be a possibility. The study of wax composition in selected materials, alpha-linolenic acid and dodecyl cyclohexane, found that this might be the major composition for benzene adsorption.

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