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Novel application of local GAC adsorption to remove organic matters and pesticides in rural drinking water treatment

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Novel application of granular activated carbon (GAC) using renewable and low cost materials to remove pesticides and organic matters was studied in the rural water treatment plant (WTP), in Hai Duong province, Vietnam. The treatment efficiencies of the a series of processes such as coagulation/ flocculation, horizontal sedimentation, rapid sand filtration and adsorption using three types of GAC (activated carbon from bituminous coal, from bamboo and from coconut shell) and the overall treatment trains were evaluated by studying several parameters such as turbidity, COD, pesticides (chlorpyrifos, diazinon, carbofuran). Results show that the two locally produced adsorbents have a similar efficiency to commercial activated carbon for adsorption. The removal efficiency was somewhat lower for both materials, but in the same order of magnitude. The removal of pesticides and organic matter with a column filled with activated carbon derived from bamboo was found the best among the two locally produced materials, approximately 30% and 23% higher than those observed for the column filled with activated carbon from coconut shells. It concludes that bamboo-derived activated carbon adsorption can be employed on an alternative to commercial activated carbon and could be a feasible option for drinking water treatment.

Introduction

Many water treatment plants (WTPs) in rural ares in Vietnam are producing drinking water from surface water using conventional treatment technologies (coagulation – flocculation – horizontal sedimentation – rapid sand filtration – disinfection by chlorine). The occurrence of pesticides in surface waters is a fast growing concern for Vietnamese WTPs. Conventional treatment alone usually does not provide sufficient organic matter and pesticide removal to maintain the concentration below the requirements for safe and clean water in Vietnam (Thang 2009). Due to high cost of commercial activated carbon, applied advanced treatment technologies for removing organic matter and pesticides in WTPs are often restricted by economical considerations low income and rural areas in Vietnam. Faced with challenges of providing safe drinking water, low cost – high performance, flexible and feasible technologies should be studied to provide safe drinking water in these areas. Activated carbon produced from renewable, waste-to-product and cheap raw materials such as bamboo and coconut shell, can be considered as low-cost and high performance adsorbents (Palanisamy & Sivakumar 2009). Bamboo and coconut grow abundantly in Vietnam and have been traditionally used to construct various living facilities, handicraft and domestic fuel. Because of their properties (they are strong, tough and low-cost), it is proposed to convert bamboo and coconut shell into activated carbon to make better use of these cheap and abundant agricultural and industrial wastes (Hameed et al. 2007). The use of locally produced activated carbon should allow for an improvement of tap water quality in low income and rural areas in developing countries, using a method that is not only low-cost but also with a high adsorption performance. Eventually, the forthcoming stringent drinking water standards should be met. This requires (i) to efficiently remove organic matter and pesticides in each treatment unit; (ii) to evaluate and compare the adsorption efficiency of two low cost granular activated carbons (GACs) compared to a commercial GAC, to explore their potential to respond to the high performance aspect for considerations in low income and rural areas.

Materials and methods

Materials

Activated carbon

The granulated activated carbons were selected from different raw materials: activated carbon made from bituminous coal (ACBC) (Norit Activated carbon Co., the Netherlands); activated carbon made from bamboo (ACB) (Ha Bac Activated Carbon Co., Hoa Binh, Vietnam) and activated carbon made from coconut shell (ACCS) (Tra Bac Activated Carbon Co., Ben Tre, Vietnam). ACBC was a commercial activated carbon, and was used as a reference in comparison with the two local activated carbons. ACB and ACCS were local activated carbons based on local traditional bamboo handicraft villages and coconut processing factories in Vietnam as waste after producing goods.

Pesticides

Three pesticides commonly used in agricultural activities in Hai Duong, which may remain in high concentrations in surface water sources, were selected: diazinon, chlorpyrifos, carbofuran. The pesticides were purchased from Hai Duong's pesticide shop with trade names Basudin 50EC (common name: diazinon), Pyrinex 20EC (chlopyrifos), Furadan 3G (carbofuran).

Pilot plant GAC filter

The pilot plant (capacity: 30 m³/day) was installed at the Cam Giang WTP in Hai Duong province, Vietnam. The pilot system was designed to test various process schemes consisting of a series of processes such as coagulation/ flocculation, sedimentation, rapid sand filter (RSF) and GAC adsorption. Three GAC filters (ACB, ACCS, ACBC) were set up to evaluate and compare the adsorption efficiency of two low cost GACs compared to a commercial GAC. Polyaluminum chloride was used as a coagulant with feed concentration about 10 – 30 mg/L depending on the result of jar test. Backwashing of both RSF and GAC filter was practiced once per day and was designed as follows: an initial air-water backwashing step, followed by a water backwash. Three GAC filters (height 3 m, internal diameter 0.2 m) were filled with 1 m of the three types of GACs and 0.3 m silica sand. The filtration rate was 5 m/h. The scheme shown in Figure 1 shows the treatment processes.

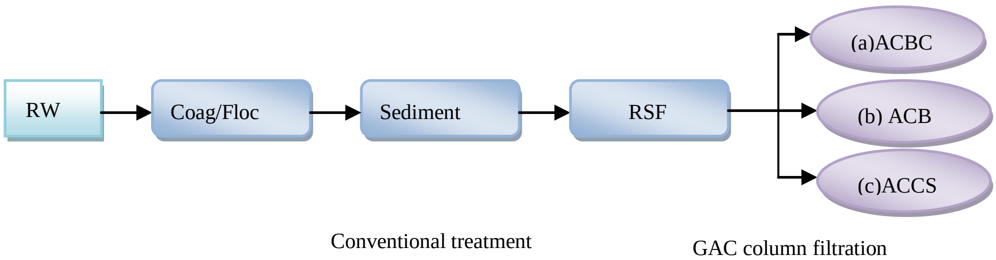

Figure 1. Schematic diagram of the pilot plant

Source: WEDC – Source

RW-Raw water, Coag/Floc-Coagulation/Flocculation, Sediment-Sedimentation, RSF-Rapid sand filtration, ACBC-Activated carbon from bituminous coal, ACB-Activated carbon from bamboo, ACCS-Activated carbon from coconut shell, GAC-Granular activated carbon.

Analytical methods

Water samples were collected from various locations to analyze turbidity, chemical oxygen dissolved (COD) and pesticides (diazinon, chlorpyrifos, carbofuran). Turbidity was measured by turbidity meter (Hana HI 98703 – Italy). The COD was analyzed by using the ASTM D1252 standard.

Diazinon and chlorpyrifos were identified and quantified with a single injection of extract using GC/MS. The concentration of carbofuran was determined with a HPLC 1200 Agilent –MS/MS API 4000 Applied Bio System.

Results and discussion

Rejection of turbidity and organic matters

Figure 2 compares the treatment efficiency of the treatment train consisting of conventional unit processes and three GAC filters (ACB, ACCS, ACBC) with respect to turbidity. The GAC filters are here operated with the same purpose as the RSF, i.e., to remove suspend solids (in addition to their adsorptive capacity, which will be described further on). The average turbidity of the effluent after RSF decreased from 32.6 to 3 NTU (average efficiency of turbidity reduction 90%). From the removal values shown in Figure 2, turbidity is generally well removed by coagulation and RSF, hence, it is frequently present only at low concentrations in GAC adsorber influent (Matilainen et al. 2006). Hence, treated water flows through the GAC filters in which turbidity is nearly completely removed; therefore, the turbidity removal efficiencies of the three GAC filters used in this study are similar. The average removal efficiency of turbidity in the three GAC filters is 97% with the ACBC and ACB filter and 96% with the ACCS filter. In the Vietnamese drinking water standard, the turbidity value of treated water should be below 2 NTU; the effluent after each of the three GAC filters meets this standard.

Removal of COD by RSF and GAC adsorption is shown in Figure 3. It should be noted that for the purpose of the study, conditions were chosen in which COD is partially removed so that a comparison of materials can be made. The average removal efficiency of organic matter is 34% with RSF, 80% with the ACBC filter, 76% with the ACB filter and 68% with the ACCS filter. From the fractional removal values of COD before and after RSF shown in Figure 3, it can be seen that RSF alone would remove little COD, but with GAC adsorption, higher removal rates of COD could be achieved. Physical effects are largely governed by the molecular weight of organic pollutants relative to the pore size distribution of GAC (Velten et al. 2011). In RSF, the COD fraction that is present as suspended solids is removed. Organic pollutants adsorption primarily takes place in mesopores (2-50 nm width) and large micropores (1-2nm width) (Cheng et al. 2005). Physical properties of reference GACs in this study are given: the iodine number of ACB was found to be the lowest (690 mg/g), while the corresponding value was higher for ACCS (800 mg/g) and for ACBC (960 mg/g) (Pham et al., 2012). The molasses number and methylene blue number was the highest for ACBC, followed by ACB and much lower for ACCS. These results confirm that the commercial activated carbon (ACBC) had a higher mesopore, macropore and micropore content than the two local activated carbons (ACCS and ACB). ACCS had a higher micropore content and a lower mesopore and macropore content than ACB. Comparing the overall efficiency the three GAC filters using these water quality parameters, train A (conventional process + ACBC filter) achieved almost the same performance than trains B and C, in which the ACB and ACCS filters were used. The average COD of the effluent after ACB filter decreased from 7.8 to 1.9 mg/l (removal efficiency 75.6%) and after ACCS filter decreased from 7.8 to 3.68 mg/l (removal efficiency 52.8%). The removal of organic matter with the ACB filter was approximately 23% higher than observed for the ACCS filter. This was expected since bamboo-based carbons have been shown to remove more COD than their coconut-based counterparts due to their higher mesopore volume (higher methylene blue number, as given above) (Newcombe 2002). Furthermore, ACB has a wider range of pore sizes than ACCS, which would make it less susceptible to fouling by organic matter. In view of the Vietnamese drinking water standard, the COD value of treated water should be below 2 mg/l and the effluent after ACB filter meets this standard.

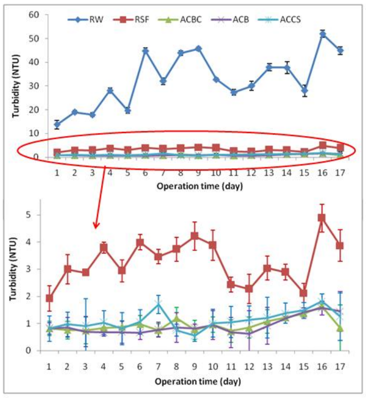

Figure 2. The variation of turbidity in different GAC column at filter velocity 5 m/h. Error bars (± standard deviation) present variability from triplicate experimental values
RSF- Rapid sand filtration, ACBC-Activated carbon from bituminous coal, ACB-Activated carbon from bamboo, ACCS-Activated carbon from coconut shell

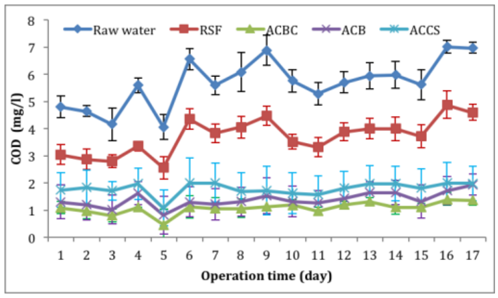

Figure 3. The variation of COD in different GAC column at filter velocity 5 m/h. Error bars (± standard deviation) present variability from triplicate experimental values
RSF-Rapid sand filtration, ACBC-Activated carbon from bituminous coal, ACB-Activated carbon from bamboo, ACCS-Activated carbon from coconut shell

Removal efficiency for pesticides

To investigate and compare the removal efficiency of pesticides for the overall removal efficiency of the conventional treatment alone and conventional treatment combined with three GAC filters, the pilot was run with fresh GAC, using three different inlet pesticide concentrations (1, 10 and 100 µg/L). Figure 4 shows the average removal of pesticides for the conventional process and three GAC filters at an inlet concentration of 100 µg/l after seventeen days running. The results show that the conventional treatment alone was ineffective to remove pesticides, but these constituents could be removed by GAC. Several studies on lab-scale and full scale drinking water treatment systems have reported removals lower than 50% for several pesticides and micro-pollutants by conventional treatment processes (Pham et al. 2008). The results in this study also show that the removal of pesticides in conventional treatment processes is lower than 50% (17.2% with chlorpyrifos, 8.5% with diazinon and 5.0% with carbofuran). The results in Figure 4 also demonstrate that the GAC process was sufficient to remove pesticides and adsorption onto activated carbon, and is driven mainly by hydrophobic interactions. Sorption of pesticides to particles depends on their physico-chemical properties. For hydrophobic compounds, sorption can be predicted from the octanol–water partition coefficient (K_{ow}) and the treatment results in pesticide removals that tend to be larger for hydrophobic pesticides (Ballard & MacKay 2005). Reungoat et al. (2010) and Westerhoff et al. (2005) observed that the trend in the removal efficiency of micropollutants (including pesticides) by activated carbon can be predicted from log Kow values. The adsorption capacities of the pesticides studied follow the order: chlorpyrifos (log K_{ow} = 4.96) > diazinon (log K_{ow} = 3.81) > carbofuran (log K_{ow} = 2.32). This explains the differences observed in the removal efficiencies of these compounds. The removal of pesticides at inlet concentration of 100 µg/l by ACCS is the lowest, followed by ACB and the highest for ACBC. In particular, ACB shows a better adsorption capacity than ACCS and almost equal to ACBC. The removal of pesticides by the ACB filter process was approximately 30% higher than that observed for ACCS filtration. Thus, the pilot scale results in this study show that ACB is effective for pesticide adsorption, suggesting that ACB generally contains a higher volume of mesopores (higher methylene blue number, as given above) which allows a quicker access to adsorption sites, resulting in faster adsorption of pesticides (Ho & Newcombe 2010). The higher fraction of mesopores and macropores of ACB compared to ACCS also acquires a reasonable surface area of adsorbing the multilayer coverage of pesticide molecules. Thus, among the two local activated carbons, ACB has a better adsorption capacity for the removal of pesticides.

The concentration of pesticides remaining in the effluent of three GAC filters is given in Table 1. It is observed that for the initial pesticides concentration of 1 µg/L, the remaining concentration of pesticides after two local GACs filters were usually below 0.1 μg/L and meet the WHO drinking water standard for pesticides. Some studies in Vietnam detected pesticides in surface water sources with concentration ranging from 0.003 to 1.7 μg/L (Lamers et al. 2011). The cost for more installation and operation of GAC filters in the existing WTP with capacity of 7,000 m^3/day, with an empty bed contact time of 20 minutes, was estimated on the basis of the results obtained in this study, as given: the unit cost for ACBC, ACB and ACCS filter installation would be 1,528 VND (equal to 0.0611 EUR), 1,058 VND (equal to 0.0423 EUR) and 1,081 VND (equal to 0.0432 EUR) per cubic meter, respectively. Hence, the two local activated carbons can be feasible for the removal of pesticides in rural WTPs in Vietnam. Among the two local activated carbons, ACB is preferred as a low-cost, high performance adsorption material.

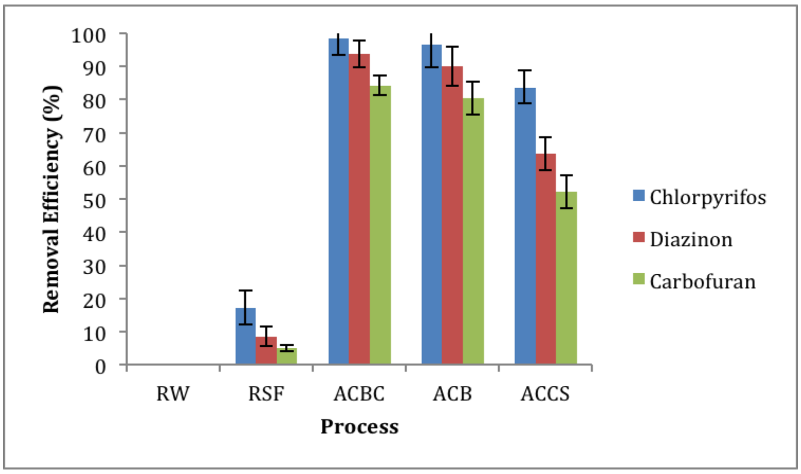

Figure 5. The average removal efficiency of pesticides with different GAC columns at filter rate = 5 m/h and inlet concentration of 100 μg/l pesticides during running period of seventeen days. Error bars (± standard deviation) present variability on the percentage removal of seventeen experimental values

RW-Raw water, RSF-Rapid sand filtration, ACBC-Activated carbon from bituminous coal, ACB-Activated carbon from bamboo, ACCS-Activated carbon from coconut shell

Table 1. The concentration of pesticides remaining in the effluent of three GAC filters

Conc. (μg/l)	The concentration of pesticides remaining in water after equilibrium (C, μg/L)								
	Chlorpyrifos			Diazinon			Carbofuran		
	ACCB	ACB	ACCS	ACCB	ACB	ACCS	ACCB	ACB	ACCS
1	0.002 ± 0.001 (99.8)	0.021 ± 0.005 (97.9)	0.039 ± 0.01 (96.1)	0.026 ± 0.015 (97.4)	0.042 ± 0.01 (95.8)	0.056 ± 0.02 (94.4)	0.068 ± 0.01 (93.2)	0.083 ± 0.025 (91.7)	0.107 ± 0.02 (89.3)
10	0.21 ± 0.05 (97.9)	0.32 ± 0.05 (96.8)	1.23 ± 0.24 (87.7)	0.51 ± 0.01 (94.9)	1.01 ± 0.21 (89.9)	3.03 ± 0.52 (69.7)	0.89 ± 0.20 (91.1)	1.57 ± 0.30 (84.3)	2.08 ± 0.4 (79.2)
100	1.50 ± 0.30 (98.5)	3.30 ± 0.78 (96.7)	16.30 ± 1.32 (83.7)	6.20 ±1.50 (93.8)	9.90 ± 1.45 (90.1)	36.40 ± 2.50 (63.6)	15.80 ± 1.75 (84.2)	19.50 ± 1.55 (80.5)	47.70 ± 3.60 (52.3)

[a] GAC filters rate = 5 m/h.
[b] The average concentration of pesticides remaining in water after equilibrium from seventeen experimental values (C, in μg/L).
[c] The error value represent ± standard deviation on variability from seventeen experimental values
[d] The average removal efficiency of pesticides from seventeen experimental values are given in parentheses (%).
ACBC - Activated carbon from bituminous coal, ACB - Activated carbon from bamboo, ACCS-Activated carbon from coconut shell, Conc. – Concentration.

Conclusion

The surface water source in Vietnam has a high concentration of organic matter and pesticides because of increasing anthropogenic, industrial, domestic and agricultural activities, natural organic matter, odor and pesticides problems. Pesticides were found to adsorb strongly on the surface of two local activated carbons. Pesticide removal depends on the hydrophobic characteristics of the compound. Comparing the pesticide removal efficiency of the two local activated carbons, ACB is observed to possess a higher adsorption capacity than ACCS. The removal of pesticides and organic matter by the ACB filter column process was approximately 30% and 23% higher than those observed for the ACCS filter column process. The present study concludes that the ACB can be employed as an alternative to commercial activated carbon and feasible option to provide safe drinking water in low income and rural areas.

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