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Characterization of wastewater from dairy industry in Palestine and its adsorption on biowaste

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ABSTRACT

This study aims at determining the pollution loads of wastewater discharged from local Palestinian dairy factories. Wastewater samples were collected from different dairy processes and pollution loads were determined. Furthermore, the technical feasibility of using adsorption process for treating such a wastewater was investigated using stirred batch and continuous packed-bed adsorption experiments with sawdust and activated carbon as adsorbents. Approximately, 477,000 m^3 of dairy wastewater in Palestine is annually discharged without proper treatment. The average measured chemical oxygen demand (COD) is 68,000 mg/L and the average total suspended solids (TSS) is 188,000 mg/L, which are much higher than values reported in the literature. The annual pollution loads released from the dairy industry in Palestine is estimated as 22,180 ton of COD, 210 ton of total dissolved solids, and 8,080 ton of TSS. Batch adsorption results showed that activated carbon particles requires more than 18 h to reach equilibrium while only 2 h are required in the case of sawdust adsorbents. However, both adsorbents achieve almost the same COD removal efficiency of about 65%. For the continuous packed-bed adsorption, increasing the bed height, increases the adsorption efficiency and the time required for the saturation of the adsorbent.

Keywords: Adsorption; Dairy wastewater; Wastewater treatment; Sawdust; Activated carbon

1. Introduction

Dairy industry is one of the most growing food processing sectors in the world. Water is heavily used in dairy processing steps for cleaning, heating, cooling and sanitization. Typically, processing of 1 m^3 of milk generates about 2.7 m^3 of wastewater, while the exact value is dependent on the end product [1]. The wastewater effluent from dairy industry is considered as one of the most polluted industrial effluents in terms of chemical oxygen demand (COD), biological oxygen demand (BOD), and total suspended solids (TSS) [2]. However, the pollution load depends on the production processes. Shete and Shinkar [2] reported that wastewater generated from cheese processing is more concentrated with COD, BOD and TSS than that released from milk processing. For instance, the wastewater released from a cheese factory has a COD of approximately 85,000 mg/L compared with 10,251 mg/L in the case of milk processing [2]. Likewise, the TSS from cheese factory was measured

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at nearly 9,000 mg/L compared with 5,800 mg/L for the wastewater produced from milk processing factory [2].

When released to the environment without proper treatment, dairy wastewater rapid decomposes and causes depletion of the dissolved oxygen in the receiving water body [2]. Furthermore, the casein precipitates from dairy wastewater decomposes into a highly odorous black sludge. In addition, the degradation of soluble organics and suspended solids in the dairy effluent release gases and odor and leads to eutrophication [2]. This makes dairy industry one of the largest source of contaminated wastewater from food processing in many countries [3].

Various techniques are widely applied for the treatment of dairy wastewater including activated sludge process, aerated lagoons, aerobic bioreactor [4], trickling filters [5], sequence batch bioreactors [6], anaerobic sludge blanket, membrane bioreactors [7], nanofiltration and others [8,9]. However, these techniques are rather complicated, expensive, energy consuming and cannot achieve the effluent discharge standards thresholds of 50 ppm BOD and 250 ppm COD as set by the world bank limits [2].

Adsorption is a promising physico-chemical treatment method that has been used for treating various types of industrial wastewater. Several studies reported the treatment of dairy wastewater using conventional adsorbents including activated carbon, silica gel, copper oxide nanoparticles, alumina, clay, marl and limestone particles [2,10,11]. Various adsorbents were also used for the removal of organic pollutants from dairy industry including activated carbon (AC), bagasse fly ash (BFA) and fly ash [12], biosorbent-water hyacinth (*Eichhornia crassipes*) [13], biochar [14], coconut shell activated carbon, laterite-red-colored-clay-rich soil [15], acid mine drainage sludge and coal fly ash [16].

In Palestine, there are more than 200 dairy manufacturing firms ranging from large production companies to small start-up [17]. The wastewater generated from these factories is generally discharged into the close surroundings, open wadies, or sewer network without sufficient or proper treatment. To the authors knowledge, the total annual pollution loads from dairy industry in Palestine was not previously determined.

The present study aims at characterizing wastewater effluents form selected local Palestinian dairy factories and determining the annual pollution loads released from dairy sector. Besides, it aims at investigating the technical feasibility of treating such a wastewater using two adsorbents obtained from biowastes (sawdust and activated carbon). The methodology of the study was based on field sampling and experimental approach. Wastewater samples were collected from different dairy processes. Then, the pollution loads in terms of COD, BOD, TSS, total dissolved solids (TDS), and pH were determined. The technical feasibility of utilizing sawdust as adsorbent for reducing organic load in dairy wastewater using batch and continuous adsorption was then investigated.

2. Materials and methods

2.1. Materials

Dairy wastewater samples were collected from two local dairy companies A (case study A) and B (case study B), located in Hebron, Palestine. Fig. 1 presents the process flow chart and shows the locations at which the wastewater samples were taken from both companies. The samples were stored in a refrigerator at 4°C after adding 2 mL of concentrated sulfuric acid per one litter of dairy wastewater to prevent natural degradation. Sawdust of mixed natural wood was obtained from a local carpentry (Al-Saeed Carpentry, Yata, Palestine). The activated carbon was obtained from Sigma-Aldrich through a local supplier in Palestine. The reagents were used for wastewater characterization included sulfuric acids (98% purity), ferrous sulfide (FS), potassium dichromate and potassium hydrogen phthalate. The chemical reagents used for wastewater characterization were obtained from Sigma-Aldrich through a local supplier in Palestine.

2.2. Methods

2.2.1. Wastewater characterization

The rates of the generated wastewater from each process in the two studied factories were determined through field data collected from the production processes. Wastewater samples were characterized in terms of pH, COD, chloride content, TSS and TDS. The pH was determined by using pH Bench Meter (Milwaukee MI150, US). The chloride content was determined using the argentometric titration method. The TSS were determined by weighing the material left after the evaporation and drying of the sample in the oven at a specific temperature of 105°C for 24 h. The TDS was measured using TDS meter (JENWAY 4510 bench conductivity meter, UK). The COD of wastewater samples was measured using 5220 C. closed reflux titrimetric method, which uses potassium dichromate as a strong oxidizing agent to oxidize the organic content under acidic conditions. Samples were then digested for approximately

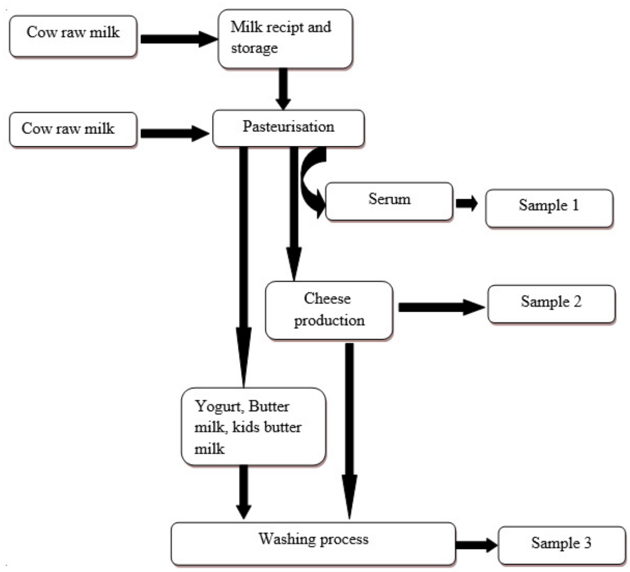

Fig. 1. Process flow chart for dairy industry showing the locations from which the wastewater samples were collected.

2 h at 150°C using COD digestion device (ROCKER, COD Reactor CR25, Taiwan) [18]. The COD values were then obtained from the following mass balance equation:

$$\text{COD}\left(\frac{\text{mg}}{\text{L}}\right)=\frac{(A-B)\times 8000\times M}{V_S} \quad (1)$$

where A is the volume of FS used for blank sample (mL), B is the volume of FS used for sample, M is the molarity of FS, V_s is the volume of sample in mL (2.5 mL).

2.3. Adsorption experiments

2.3.1. Batch adsorption

The required diluted wastewater samples were mixed with the required amounts of the activated carbon or sawdust adsorbents. The obtained mixture was kept under stirring at a rate of 70 rpm. The COD was measured at different time intervals by taking sample from the solution. The adsorption capacity of the adsorbents; q_t in (mg/g), was obtained as a function of time from the COD data, using mass balance as follows:

$$q_t=\frac{V\left(\text{COD}_0-\text{COD}_t\right)}{m} \quad (2)$$

where COD_0 is the initial COD of wastewater (mg/L), COD_t is the obtained COD (mg/L) at a time (t), m is the mass of adsorbent in mg and V is the volume of wastewater for each batch (0.1 L).

The efficiency of the adsorption process was expressed as the percentage reduction in the COD, as given by the following equation:

$$\text{Percentage removal of COD}=\frac{\text{COD}_0-\text{COD}_t}{\text{COD}_0}\times 100\% \quad (3)$$

2.3.2. Continuous adsorption in fixed bed column

The continuous adsorption experiments were performed using a packed bed column made of glass with an internal diameter of 4 cm, purchased from (Elettronicaveneta mod. LSA/EV, Italy). A schematic representation of the adsorption setup is shown in Fig. 2. The sawdust particles were packed inside the bed. The bed performance was investigated at two different heights of 10 and 30 cm. The dairy wastewater stream was fed from the top of the bed using a pump (APK 2100). The inlet and outlet concentrations of COD were measured and then the adsorption break-through curves were obtained, by dividing the exit COD_e by the inlet COD_o.

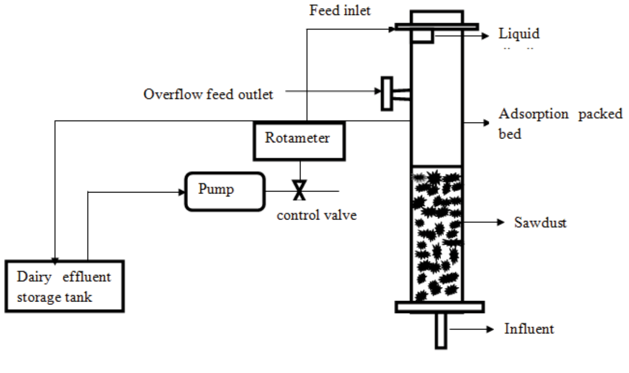

Fig. 2. Schematic representation of the continuous adsorption packed-bed setup.

3. Results and discussion

3.1. Pollution loads

The measured rates of generated wastewater from the three main processes (yogurt production, cheese production and washing stages) at the two dairy factories are reported in Table 1.

Table 1 indicates that there are noticeable variations in the rates of wastewater discharge between the two case studies. The total average wastewater discharged from case B is estimated at 62 m^3/d compared to 15 m^3/d from case A. The variations in the rates of wastewater discharged from both cases is ascribed to the difference in the size of the companies and the applied technologies in manufacturing processes. The measured physio-chemical characteristics of the wastewater from the two case studies, A and B, are listed in Tables 2 and 3, respectively. Tables 2 and 3 show that the wastewater from dairy manufacturing processes is heavily concentrated in term of organics (COD), chloride, pH, TSS and TDS. These parameters are higher than those reported in the literature [2]. The COD of the wastewater from case A is much higher than that of B. This may be due to less process and washing water used than case B, resulting in diluted streams. It can be also noticed that the COD and TSS of the wastewater resulting from cheese production process are much higher than those resulting from yogurt production process in both cases. This can be ascribed to the high amounts of whey protein, lactose and fat content in the wastewater stream during cheese production. On the other hand, the TDS in the yogurt effluent (~900 mg/L) is higher than that in the cheese wastewater (~680 mg/L). This can be due to the presence of the residues from the added ingredients (such as sweeter, sucrose, artificial sweeteners and stabilizers) during yogurt production for modifying certain properties of the yogurt.

The average of the measured COD values from all processes given in Tables 2 and 3 is estimated at 68,000 mg/L. This value is extremely high compared to values reported in the literature [2]. The Same observation also applies to the TSS, the average measured value of which is 188,000 mg/L. Based on the COD results, and using a BOD:COD ratio of

Table 1
Total amount of daily wastewater discharged from each dairy manufacturing process for both cases in cubic meter per day

	Yogurt (m^3/d)	Cheese (m^3/d)	Washing (m^3/d)
A	2	0.8	12
B	10	15	37

Table 2
Characteristics of wastewater samples collected from case study A

	Yogurt production	Cheese production	Soda washing	Acid washing	Dumped to sewer system (kg/d)
COD ($\times10^{-3}$), mg/L	120	203.2	41.6	33.6	853.76
TDS, mg/L	862	671	282	314	5.83
TSS ($\times10^{-3}$), mg/L	4.1	72.1	13.2	2.2	158.28
Turbidity ($\times10^{-2}$), NTU	49.9	49	42.7	11.28	–
EC, µs/cm	14.07	8	23.3	5.2	–
Chloride ($\times10^{-3}$), mg/L	3	4.2	7.5	2	66.36
pH	5.7	6.5	7.7	2.11	–

Table 3
Characteristics of wastewater samples collected from case study B

	Yogurt production	Cheese production	Soda washing	Acid washing	Dumped to sewer system (kg/d)
COD ($\times10^{-3}$), mg/L	17.6	118.4	3.2	9.6	2188.80
TDS, mg/L	920	691	269	301	30.11
TSS ($\times10^{-3}$), mg/L	3.9	89.9	1.1	1.6	1437.45
Turbidity ($\times10^{-2}$), NTU	76.1	77	14.89	17.57	–
EC, µs/cm	15.07	10.9	43.9	4.86	–
Chloride ($\times10^{-3}$), mg/L	2	4	9	2	283.5

0.91 [19], the total dumped BOD is estimated at 1,386 kg/d which is extensively polluted effluent compared to the Palestinian standard for industrial wastewater, which allows up to 150 mg/L of BOD to be discharged [20].

The annual milk production in Palestine is approximately 182,000 tons (175,845 m^3) [21]. Based on the average value of wastewater generation of 2.71 m^3/m^3 of processed raw milk [1], and the total annual milk processing (175,845 m^3), approximately, 477,000 m^3/y of dairy wastewater is generated in Palestine. The annual pollution loads from the dairy industry in Palestine were determined in terms of total COD, TSS, TDS and Cl^- discharged into the environment based on the measured values in cases A and B. The estimated total pollution loads dumped into the sewer system are given in Table 4. These high pollution loads of the generated dairy wastewater in Palestine might be ascribed to the fact that the processing water and washing water used at local dairy plants are much less than those used in other countries, which might be due to the shortage of water caused by the complicated political situation in Palestine. These extremely polluted effluents from local dairy industry require corrective measures from the policy-makers and stake-holders to reduce the resulting environmental impacts. Several steps might be applied here, including installation of proper in-situ wastewater treatment units to reduce the organic load in the effluent streams.

3.2. Batch adsorption experiment

The technical feasibility of organics removal from dairy wastewater by its treatment with sawdust and activated carbon is demonstrated experimentally. Adsorption kinetics for both activated carbon and sawdust were performed. Fig. 3 presents the adsorption kinetics for both adsorbents using a adsorbent dose of 5 g/100 mL, for each case. Fig. 3 shows typical adsorption kinetics for types of adsorbents, where the concentration of the pollutant decreases with time at a decreasing rate until it approaches a plateau value. It shows that the adsorption onto activated carbon particles is slower than that on sawdust, as it takes more than 18 h to reach equilibrium, compared with 2 h in the case of sawdust. Also, the initial adsorption rate (initial slope) onto sawdust is much higher than that onto activated carbon. On the other hand, both adsorbents achieved almost the same final removal efficiency since they achieve a nearly similar final COD value. The adsorption efficiency [Eq. (3)] and the adsorption capacity [Eq. (2)] of sawdust and activated carbon are listed in Table 5. The table also lists the previously reported adsorption efficiency and adsorption capacity using clay, marl and stone cutting solid waste for the same type of wastewater and the same adsorbent dosage [12,22]. For the same dosage, stone cutting solid waste has almost the same adsorption efficiency for both activated carbon and sawdust. These results present a practical approach for

Table 4
Estimated total approximated organic loads dumped into the sewer system resulted from dairy industry in West Bank

Organic load	COD	TDS	TSS	Cl^-
Approximated value (ton/y)	22,180	210	8,080	2,160

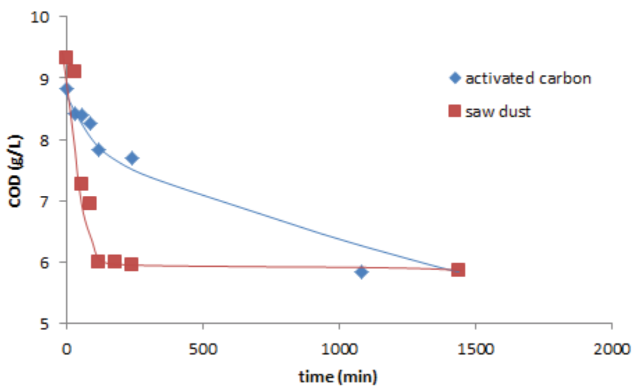

Fig. 3. Adsorption kinetics for both activated carbon and sawdust particles mixed with dairy wastewater (dose 5 g/100 mL) at a stirring rate of 70 rpm.

Table 5
Adsorbent efficiency and adsorption capacity for treating wastewater from dairy industry using sawdust, activated carbon (obtained from this study), compared to those for marl, clay and stone cutting solid waste (obtained from previous work [22]) for a dose of 5 g/100 mL

Type of adsorbent	Adsorption efficiency %	Adsorption capacity (q) (mg/g)
Activated carbon	64.3	0.964
Sawdust	65.9	0.965
Clay	29	3.2
Marl	35	4
Stone cutting solid waste	68	7.3

reducing environmental pollution of industrial wastewater in Palestine, using low cost, local abundant materials.

3.3. Continuous adsorption experiments

The breakthrough curves of the COD adsorption in a packed bed of sawdust are shown in Fig. 4 as plots of (COD_e/COD_0) as functions of time for two bed heights of 10 and 30 cm. Fig. 4 indicates that the higher the bed height, the higher the adsorption efficiency and the longer the time required for the saturation of the sawdust. Increasing the bed height increases the removal efficiency since pollutants can achieve more residence time to be contacted with sawdust particles and due to the higher availability of more adsorption sites. For instance, Fig. 4 shows that for a bed with a height of 10 cm, the COD_e/COD_o increases rapidly from 0 to 0.4 within a period of about 7 min, however, increasing the bed height causes a major shift in the time domain of the breakthrough curve. Hence, the saturation time is extended to about 25 min when a bed height of 30 cm is used.

4. Conclusion

The physico-chemical characteristics and the rates of wastewater generated from dairy industry in Palestine were determined. Approximately, 477,000 m^3 of dairy wastewater is annually produced in Palestine and discharged to the surroundings without proper treatment. The released wastewater is highly polluted with average values of COD and TSS of 68,000 and 188,000 mg/L, respectively. These values are higher than those reported for dairy effluents in other countries. The estimated annual pollution loads released from dairy industry in Palestine are 22,180 ton of COD, 210 ton of TDS, 8,080 ton of TSS and 2,160 ton of chloride content.

Installation of proper in-situ wastewater treatment processes can reduce pollution loads in the effluent streams. Batch adsorption using sawdust is faster than activated carbon, however, COD removal efficiency of about 65% can be obtained by both adsorbents at various time scale. For the continuous packed-bed adsorption, both the adsorption efficiency and the bed saturation time increase with increasing bed height.

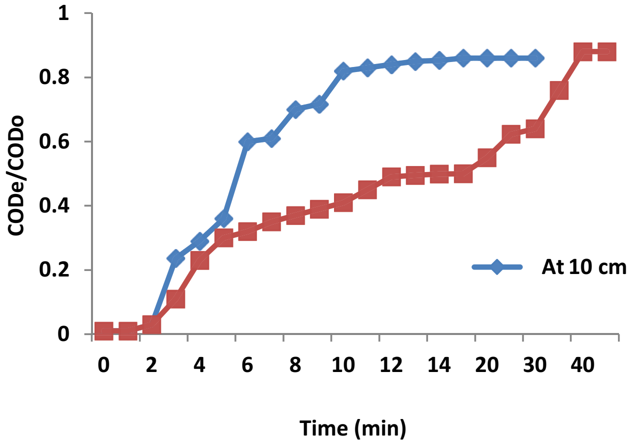

Fig. 4. Breakthrough curves of the adsorption treatment of dairy wastewater in packed bed of sawdust at two different bed heights (10 and 30 cm).

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