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Versatile applications of freshwater and marine water microalgae in dairy wastewater treatment, lipid extraction and tetracycline biosorption

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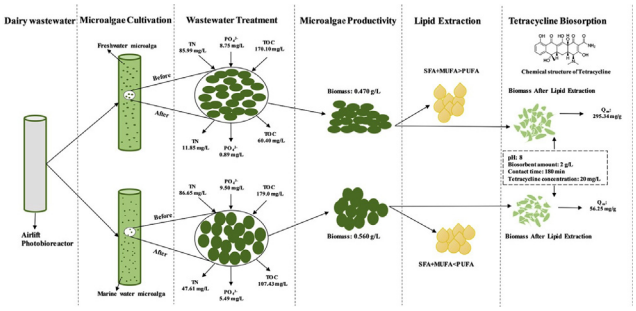
GRAPHICAL ABSTRACT



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ABSTRACT

In this study, freshwater (*Scenedesmus quadricauda*, *Sq*) and marine water (*Tetraselmis suecica*, *Ts*) microalgae were used for the treatment of dairy wastewater (DWW). *Sq* and *Ts* showed the highest biomass productivity as 0.47 and 0.61 g/L, respectively. Removal efficiencies of total nitrogen (TN), phosphate (PO_4^{3-}), and total organic carbon (TOC) were observed as 86.21, 89.83 and 64.47% by *Sq* and 44.92, 42.18 and 40.16% by *Ts*, respectively. After wastewater treatment, lipids were extracted from microalgal biomasses. Fatty acid methyl esters (FAMEs) analysis revealed that saturated fatty acids (SFAs) are dominant in *Sq* and polyunsaturated fatty acids (PUFAs) in *Ts*. After lipid extraction, removal of tetracycline (TC) from water by microalgal biomasses was also investigated. Maximum adsorption capacities of *Sq* and *Ts* were found to be 295.34 and 56.25 mg/g, respectively. Results of this study revealed the versatile applications of microalgae for wastewater treatment, lipid production and TC removal from water.

1. Introduction

During the past several decades, microalgae and their products/metabolites have gained a remarkable attention for many environmental applications. This growing interest in microalgae research is related to the particular features of microalgae such as high growth rate, CO_2 biofixation ability, promising feedstock of biofuel and synthesizing a range of valuable products (Ji et al., 2014). Microalgae are able to grow in wide range of environments including freshwater to seawater. Most species of microalgae are able to convert energy of sun light to biochemical compounds (protein, lipid and carbohydrate) by photosynthesis process (Souliès et al., 2016). Review of the literature

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shows a wide range of applications of microalgae and derived compounds such as, wastewater treatment, bioenergy production, human food and health, animal and aquaculture feed and extraction of vitamins, antioxidants, pigments, triglycerides and polysaccharides (Mata et al., 2010).

Among different applications of microalgae, wastewater treatment with microalgae and simultaneously using the harvested biomass as substrate of bioenergy production are two noticeable applications of microalgae. Microalgal-based wastewater treatment is a promising alternative technique as wastewater contains nitrogen, phosphorous, carbon compounds and other macro and micronutrients that are required for the growth of microalgae. Cultivation of microalgae in wastewater reduces the concentration of contaminants and produces microalgal biomass (Daneshvar et al., 2018). Microalgal species such as *Chlorella* sp. and *Dunaliella* sp. have been used for wastewater treatment and biomass production for > 75 years (Yeşilova et al., 2018). In recent years, different species of microalgae have been applied in the treatment of domestic, agricultural, and industrial effluents such as dairy (Lu et al., 2015), pulp and aquaculture (Daneshvar et al., 2018), and piggery wastewaters (Wang et al., 2016). Among different industries, food processing and dairy industries produce a considerable amount of wastewater with high concentration of nutrients that is a favorable medium for cultivation of microalgae (Monroig et al., 2013).

After wastewater treatment, lipid of harvested microalgae from wastewater can be converted to fatty acid methyl esters (FAMEs) by transesterification and can be used as a clean, cheap and renewable source of energy (biodiesel) (Kuo et al., 2015). Extraction of biodiesel from microalgae is economically reasonable especially when the price of microalgal mass production as feedstock is low. In this way, microalgal mass can be harvested directly in wastewater without extra investment for microalgae cultivation (Ma et al., 2016). The final cost of microalgal mass production and consequently, lipid and biodiesel extraction can be inexpensive by using wastewater as a cheap and easily available medium for cultivation of microalgae.

Another application of microalgae is their use as an efficient biosorbent for the removal of diverse contaminants such as pharmaceuticals from water. In particular, microalgal mass, after lipid extraction, can be applied for the removal of toxic aquatic pollutants such as pharmaceuticals. Pharmaceutical compounds are emerging contaminants and their low concentrations even in μg/L or ng/L cause serious ecological problems and health effects to human and animals (Xiong et al., 2018). For example, tetracycline (TC) antibiotic is widely used to treat a number of infections; due to its high demand, elevated concentrations of TC have been reported in surface waters, wastewater treatment plant, ground waters, and drinking water (Priya and Radha, 2017). The presence of TC in aquatic environment causes the bacterial resistance and therefore, it is necessary to remove it from water (Yeşilova et al., 2018).

Combining different applications of microalgae is an interesting idea to meet environmental challenges. One-time cultivation of microalgae in wastewater and achieving different goals at the same time can reduce the cost of investment and contributes towards environmental sustainability. In this study, freshwater (*Scenedesmus quadricauda*, *Sq*) and marine water (*Tetraselmis suecica*, *Ts*) microalgae were cultivated in dairy wastewater. Microalgal growth was monitored during 12 days by calculating dry algal masses. After cultivation period, wastewater treatment was evaluated by measuring nitrate (NO_3-N), total nitrogen (TN), phosphate (PO_4^{3-}), total organic carbon (TOC) and sulfate (SO_4^{2-}). After that, microalgae were harvested and lipids were extracted from the microalgal biomass. FAMEs profile of cultivated microalgae was analyzed to investigate the potential of microalgal lipid as biodiesel feedstock. Finally, the removal of tetracycline (TC) pharmaceutical from water was studied by rest of the microalgal biomasses, after lipid extraction. To the best of our knowledge, there is no study which reports the versatile applications of microalgae (freshwater and marine water) for different purposes, as reported in this study.

2. Materials and methods

2.1. Microalgae strains and cultivation conditions

Freshwater, *Scenedesmus quadricauda* (*Sq*) and marine water, *Tetraselmis suecica* (*Ts*) microalgae were purchased from the Culture Collection of Algae and Protozoa (CCAP, Scotland, UK). Main stocks of *Sq* and *Ts* were inoculated in 1.1 L photobioreactors including 1 L of Bold's Basal Medium (BBM) and modified Guillard's marine water enrichment solution (F/2), respectively. After centrifuging, *Sq* with optical density (OD) of 0.209 nm (0.052 g/L) and *Ts* with OD of 0.161 nm (0.073 g/L) were transferred to 12 L airlift photobioreactor including 10 L of dairy wastewater (DWW). Photobioreactors were aerated and mixed gently by injection of compressed air with 0.04% CO_2. Microalgae were grown under continues illumination of 110 μmol photon/m^2.s, provided by white fluorescent lamp at 25 °C for 12 days (Daneshvar et al., 2018).

Microalgal growth was recorded by converting the optical density (OD) to dry mass for each photobioreactor. During days 2, 4, 8 and 12 of cultivation time, 5 mL sample was taken and the optical density of microalgae was read by UV-spectrophotometer at 680 nm. Microalgal dry weight was calculated according to a linear relationship as follows:

$$Sq\ \text{dry weight}\,(g/L) = 0.2784 \times OD680 - 0.0061 \quad (R^2 = 0.9981) \tag{1}$$

$$Ts\ \text{dry weight}\,(g/L) = 0.4221 \times OD680 + 0.0054 \quad (R^2 = 0.9963) \tag{2}$$

2.2. Wastewater treatment using microalgae

Dairy wastewater was collected from a local milk producing factory in Lapinlahti, Finland. Initial pH of DWW was adjusted to 6.5–7.0 by adding 0.1 and 1 M of HCl and/or NaOH. For cultivation of *Ts* (marine microalga), 34 g/L sea salt (Havsalt company) was added to DWW. Both, freshwater and marine water microalgae were cultivated in DWW without adding extra macro and micronutrients. The initial (before adding microalgae) and final (after 12 days of cultivation) concentrations of nitrate (NO_3-N), total nitrogen (TN), phosphate (PO_4^{3-}), total organic carbon (TOC) and sulfate (SO_4^{2-}) were measured to calculate the removal efficiencies of target pollutants.

Samples were centrifuged and filtered using 0.45 μm membrane filters before analysis. NO_3-N, SO_4^{2-} and PO_4^{3-} were analyzed using HACH analysis kits and spectrophotometer (DR 2800 and DR 2010). TN and TOC concentrations were measured by TOC/TN Analyzer (multi N/C 2100S). The removal efficiencies of pollutants were calculated as follows:

$$\text{Removal efficiency}\ (\%) = \frac{(C_I - C_F)}{C_I} \times 100 \tag{3}$$

where C_I and C_F are the initial and final concentrations of target pollutants on initial and final days of experiment, respectively.

2.3. Lipid extraction and fatty acids profile

Sq and *Ts*, cultivated in DWW, were collected from photobioreactors by centrifuging them for 10 min at 5000 rpm. Harvested microalgal biomasses were freeze dried and then, lipid was extracted and converted to FAMEs according to a modified one-step in-situ transesterification method (IST) (Levine et al., 2011). Then, FAMEs composition was analyzed by a gas chromatography (Agilent Technologies 7890A) equipped with a 10 m capillary column (DB–WAX, 0.1 mm i.d., × 0.1 μm) using helium as a carrier gas (30.34 cm/s). Initial oven temperature was 40 °C, which was held for 0.5 min and then increased to 195 °C at the rate of 25 °C/min. The temperature was then increased to 205 °C at the rate of 3 °C/min. Finally, the temperature was increased to 230 °C at the rate of 8 °C/min and held for 4 min. Temperatures (both of injector and detector) were set at 250 °C. C17:0 (Sigma) was added as

an internal standard. All measurements were performed in duplicate and the mean values of results as ± standard deviation (SD) are presented.

2.4. Tetracycline removal from water by microalgal biomasses after lipid extraction

2.4.1. Preparation of biosorbent and adsorbate

After lipid extraction, the rest of the biomasses of *Sq* and *Ts* were collected. To remove unreacted solvents (chloroform and methanol) from the surface of microalgal residues, tap water was added to centrifuging tubes containing biomasses of *Sq* and *Ts*. Tubes were shaken for five min and then, microalgal biomasses were collected by centrifugation and supernatant was discarded. This procedure was repeated for two times with tap water and one time with deionized water. Finally, microalgal biomasses were collected by centrifugation and dried in an oven at 60 °C for 20 h, crushed and sieved to obtain particles < 45 µm. The powdered algal masses (without any further chemical or physical modification) were used as biosorbents for TC removal from water.

TC with purity ≥98.0% (NT), molecular formula of $C_{22}H_{24}N_2O_8{\cdot}xH_2O$ and molecular weight of 444.43 g/mol (anhydrous basis) was purchased from Sigma-Aldrich. A stock solution of TC (500 mg/L) was prepared and used for biosorption studies after making appropriate dilutions.

2.4.2. Biosorption studies

The batch biosorption studies were performed to evaluate the TC removal from water by microalgal biomasses. Biosorbent dosage, initial adsorbate concentration, contact time, pH, agitation speed, and temperature are the variables that influence biosorption process (Crini and Badot, 2008). The biosorption experiments were conducted to investigate the effects of initial TC solution pH (2.0–10.0), biosorbent amount (0.25–4.0 g/L), initial TC concentration (2.5–400 mg/L) and contact time (2–180 min). 10 mL of TC solution in 15 mL centrifuging tube was agitated in a shaker at 80 rpm to achieve the equilibrium. In order to separate biosorbent from aqueous phase, 5 mL sample was centrifuged and filtered using 0.45 µm membrane filters. The residual concentration of TC was analyzed by a double beam UV–VIS spectrophotometer (UV-2401 PC) at a wavelength of λ_{max} = 357 nm. TC removal efficiency (%) was calculated according to Eq. (3) and biosorption capacity (mg/g) was calculated according to Eq. (4) as follows:

$$q_e = \frac{(C_I - C_F)}{m} V \tag{4}$$

where C_I and C_F are initial and final concentrations of TC, respectively, q_e (mg/g) is biosorption capacity of biosorbent, V (L) is the working volume of TC and m (g) is the weight of biosorbent. The experimental data was modeled with different isotherm models.

2.4.3. Isotherm modeling

The most widely used isotherm models, including Langmuir (Langmuir, 1918), Freundlich (Freundlich, 1907) and Sips (Sips, 1948) were studied to describe the biosorption equilibria. Equilibrium data of TC biosorption were fitted with non-linear forms of isotherm models as follows:

$$q_e = \frac{Q_0 \times b \times C_e}{1 + b \times C_e} \tag{5}$$

$$q_e = K_F \times C_e^{1/n} \tag{6}$$

$$q_e = \frac{K_S \times C_e^{\beta_S}}{1 + a_s \times C_e^{\beta_S}} \tag{7}$$

where q_e is the amount of adsorbate in the biosorbent at equilibrium (mg/g), Q_o is the maximum monolayer coverage capacities (mg/g), b is the Langmuir isotherm constant (dm^3/mg), C_e is the equilibrium concentration (mg/L), K_F is the Freundlich isotherm constant related to biosorption capacity (mg/g) $(dm^3/g)^n$, n is the biosorption intensity, K_S is the Sips isotherm model constant (L/g), β_S is the Sips isotherm model exponent and a_S is the Sips isotherm model constant (L/mg) (Foo and Hameed, 2010).

2.5. Statistical analysis and modeling

Mean ± SD values of replicate samples are reported for different treatments and control groups. Data were analyzed with an overall one-way analysis of variance (ANOVA) followed by Duncan's test ($p < 0.05$) to evaluate if there were significant differences among the obtained results. The IBM SPSS Statistics 21.0 software was used to carry out the statistical analyses. Biosorption isotherm models were fitted and plotted using Solver function in Excel 2016.

3. Results and discussion

3.1. Microalgal growth in DWW

Attainment of maximum microalgal growth and biomass production in wastewater is directly related to the efficient wastewater treatment and access to primary substance for lipid extraction and biodiesel production. Fig. 1 depicts the freshwater and marine water microalgal growth performance in terms of microalgal dry biomasses (g/L) during 12 days of cultivation time. *Sq* and *Ts* dry weights increased from 0.05 to 0.14 and 0.07 to 0.15 g/L, respectively after 24 h. Microalgal growth, after one day of cultivation reveals that both, *Sq* and *Ts* microalgae can strongly adapt to the new environmental conditions in DWW. As can be seen from Fig. 1, both microalgae grew gently from the beginning of cultivation until day 4. Then, the exponential growth phase began on day 4 and microalgal biomasses increased actively up to day 8. Maximum dry biomass of *Sq* was observed as 0.47 g/L on day 8 and subsequently, it decreased to 0.24 g/L on day 12. The growth of *Ts* continued after 8 days and reached the highest biomass as 0.61 g/L on day 12. Earlier decline phase was observed by *Sq* as compared to *Ts* can be related to depletion of nutrients. Phosphorous and nitrogen are considered as two limiting primary production nutrients in freshwater and marine water, respectively (Doering et al., 1995). Here, as presented in Table 1 and Fig. 2, the initial PO_4^{3-} concentration of 8.7 ± 0.1 mg/L, in *Sq* medium was decreased to 0.87 mg/L after 12 days of cultivation. In another study, Shen et al. (2015) reported that growth rate of *Scenedesmus obliquus* in sufficient N and P, sufficient N and limited P, and sufficient N and P deficiency media were 114.0, 99.4 and 37.8 mg/L.d,

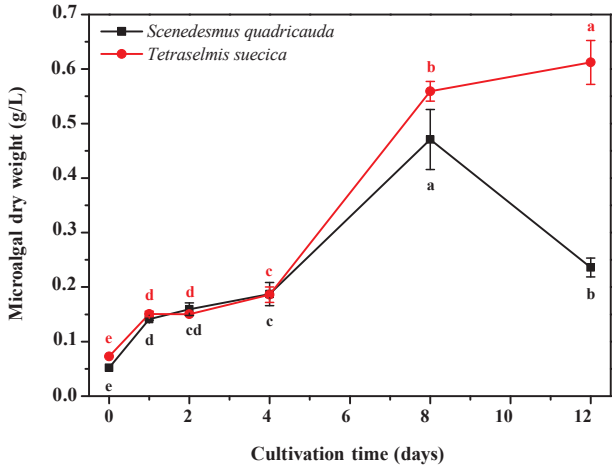

Fig. 1. Microalgal growth in dairy wastewater during 12 days of cultivation time.

Table 1
Characteristics of dairy wastewater for cultivation of freshwater and marine water microalgae.

Parameters	Values	
	DWW without sea salt	DWW with sea salt
NO_3-N (mg/L)	31.30 ± 3.3	31.60 ± 3.3
TN (mg/L)	85.99 ± 2.2	86.65 ± 2.2
PO_4^{3-} (mg/L)	8.70 ± 0.1	9.50 ± 0.1
SO_4^{2-} (mg/L)	13.00 ± 1.4	90.00 ± 1.4
TOC (mg/L)	170.11 ± 11.2	179.35 ± 11.2
[a] COD (mg/L)	654.45 ± 32.1	678.95 ± 42.5
[b] TDS (mg/L)	3830 ± 42.4	34650 ± 889.9
[c] EC (μs/cm)	7665 ± 91.9	68270 ± 3539.3
pH	6.45 ± 0.1	6.72 ± 0.3
Color	Whitish	Whitish

[a] Chemical Oxygen Demand.
[b] Total Dissolved Solids.
[c] Electrical conductivity.

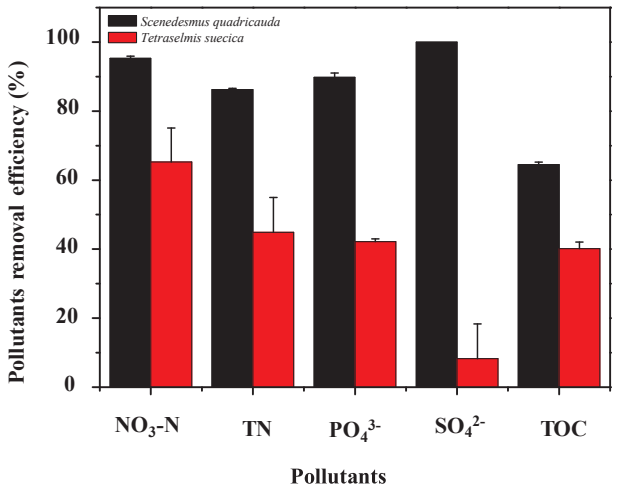

Fig. 2. Pollutants removal efficiency by microalgae during 12 days of cultivation time.

respectively (Shen et al., 2015). However, it seems that 44.92 ± 3.1% reduction in initial TN concentration (86.65 ± 2.23 mg/L) after 12 days of cultivation did not disturb the growth of marine water microalga, *Ts*. Reyimu and Özçimen (2017) cultivated marine water microalgae *Nannochloropsis oculata* and *Tetraselmis suecica* in different concentrations (25, 50, 75 and 100%) of treated municipal wastewater (Reyimu and Özçimen, 2017). They stated that depending on microalgae species and level of nutrients availability, growth curve shows different patterns in lag, log and stationary phases. The findings of this research revealed that DWW is a suitable medium for cultivation and production of freshwater and marine microalgae without using additional macro and micronutrients.

3.2. Wastewater treatment by microalgae

3.2.1. NO_3-N removal efficiency

The change of NO_3-N, TN, PO_4^{3-}, SO_4^{2-} and TOC concentrations in DWW was monitored along with the cultivation of *Sq* and *Ts* and removal efficiencies were depicted in Fig. 2. *Sq* and *Ts* removed 95.27 ± 0.6 and 65.28 ± 9.8% of NO_3-N, respectively from DWW after 12 days of cultivation. Microalgae are well-known microorganisms for the removal of inorganic (NO_2^-, NO_3^- and NH_4^+) and organic (Urea, CH_4N_2O) forms of nitrogen from wastewater. Microalgae utilize NO_3-N after its reduction to NO_2^- and then NH_4^+ by nitrate reductase and nitrite reductase enzymes (Salama et al., 2017). Many researchers have reported the uptake of NO_3-N form wastewater by microalgae. Nitrate removal efficiency from domestic wastewater by *Chlorella pyrenoidosa* and *Scenedesmus abundans* were found to be 80 and 83%, respectively (Lekshmi et al., 2015). In another study, 78.46% nitrate removal efficiency from secondary treated domestic sewage was observed by marine water microalga, *Tetraselmis indica* (Chandra et al., 2017). Results of this study showed that *Sq* is more efficient and fast for assimilation of NO_3-N as compared to T_S. However, higher nitrate removal efficiency by *Sq* can be dependent to the microalgal species; but, it can also be related to denitrification of nitrate by bacteria (in freshwater) and ammonia volatilization as well (Guldhe et al., 2017; Sabeti et al., 2018).

3.2.2. TN removal efficiency

Initial TN concentration in DWW was reduced from 85.99 ± 2.2 to 10.66 ± 2.2 mg/L (86.21 ± 0.4% removal efficiency) and 86.65 ± 2.2 to 47.61 ± 7.5 mg/L (44.92 ± 10.1% removal efficiency) by *Sq* and *Ts*, respectively. In both cases of freshwater and marine water microalgae, TN removal efficiency was less than NO_3-N. It shows that in DWW, there are some other forms of nitrogen (organic) which are removed in lower amount as compared to NO_3-N. In another study, the removal efficiency of TN and NH_4-N from municipal wastewater was investigated in hetero-photoautotrophic two-stage cultivation (Zhou et al., 2012). The authors reported that the removal efficiency of TN and NH_4-N was 59.7% and 69%, respectively in heterotrophic stage and 73.7% and 100%, respectively in autotrophic stage. The authors stated that reason of lower TN removal efficiency can be related to some organic sources of nitrogen that cannot be used by microalga.

3.2.3. PO_4^{3-} removal efficiency

As can be seen from Fig. 2, PO_4^{3-} removal efficiency was observed as 89.83 ± 1.2 and 42.18 ± 0.8% by *Sq* and *Ts*, respectively. Nayak et al. (2016) reported that cultivated *Scenedesmus* sp. in domestic wastewater could bioremediate 72.8 ± 2.6 to 81.9 ± 2.4% of phosphate with 9.24 ± 0.3 mg/L initial concentration by the end of growth period (Nayak et al., 2016). In the present study, uptake of phosphate from DWW by *Ts* was found remarkably lower than *Sq*. In agreement to our results, Serrano et al. (2017) reported 54.3% phosphate removal efficiency from table olive processing water effluent by *Nannochloropsis gaditana* (Serrano et al., 2017). They reported that this value is lower than phosphate removal from wastewater by some species like *Scenedesmus* sp. which can be achieved to > 80%. As depicted in Fig. 2, lower phosphate and TN removal efficiencies were observed by marine water microalga (*Ts*) as compared to freshwater microalga (*Sq*). Low removal efficiencies of phosphate (52.65%) and TN (62.25%) have been addressed by cultivation of marine water microalga (*Nannochloropsis oculata*) in municipal wastewater. Here, the final concentration of TN decreased to 10.66 mg/L, whereas the final concentration of phosphate decreased to 0.89 mg/L. These results revealed that freshwater microalga is more efficient for phosphate and nitrate removal from DWW. DWW treatment using *Sq* met European discharge standards for TN (< 15 mg/L) and phosphate (< 2 mg/L) (Novoveská et al., 2016).

3.2.4. SO_4^{2-} removal efficiency

Besides nitrogen and phosphorus, microalgae need sulfur to grow and maintain their metabolism. Removal efficiency of SO_4^{2-} as the main form of sulfur from DWW by *Sq* and *Ts* was also investigated in this work. After 12 days of cultivation, *Sq* displayed high removal efficiency of SO_4^{2-} (100%) (Fig. 2). Whereas, *Ts* showed 8.25 ± 1.1% removal efficiency of SO_4^{2-} during the same cultivation time. Significant difference of SO_4^{2-} uptake by *Sq* and *Ts* can be attributed to its initial concentration in their media. Initial SO_4^{2-} concentration in DWW (*Sq* medium) was 13 ± 1.4 mg/L and it was increased to 90 ± 1.4 mg/L by adding 34 g/L of sea salt to DWW (*Ts* medium). In a

study by Solovchenko et al. (2014), authors suggested that low removal of SO_4^{2-} (35%) from alcohol distillery wastewater by *Chlorella sorokiniana* might be related to limited rate of SO_4^{2-} involvement into microalgal metabolism (Solovchenko et al., 2014). Different metabolic pathways of *Sq* and *Ts* for SO_4^{2-} can be the other reason of lower SO_4^{2-} removal efficiency by *Ts*.

3.2.5. TOC removal efficiency

One of the major contaminants in wastewaters is organic carbon that needs to be removed before discharging wastewater. TOC removal efficiency from DWW by *Sq* and *Ts* is illustrated in Fig. 2. As can be seen from this figure, 170.11 ± 11.2 and 179.35 ± 11.2 mg/L of TOC in *Sq* and *Ts* media reduced to 60.40 ± 2.7 (64.47 ± 0.7%) and 107.43 ± 10.7 mg/L (40.16 ± 1.9%), respectively after 12 days of cultivation. Carbon is an essential element for the growth of microalgae that forms approximately 50% of dry microalgal biomass (Sutherland et al., 2016). By cultivation of microalgae in wastewater, organic and inorganic carbon will be used to supply carbon. Subsequently, the concentration of carbon will be reduced as microalgae grow in wastewater (Gani et al., 2017). In this study, the percent removal of TOC from DWW was lower than other inorganic compounds (NO_3-N, TN and TP); but it was higher by the weight of pollutants (mg) per produced dry microalgal mass (g). Arora et al. (2016) reported the same range of TOC reduction in domestic, sewage, paper mill and dairy wastewaters from 144.21 ± 0.2, 120.12 ± 5.1, 127.12 ± 5.4 and 165 ± 7.8 to 41.2 ± 1.7, 7.32 ± 1.6, 9.12 ± 1.4 and 23.14 ± 3.5 mg/L, respectively (Arora et al., 2016).

3.3. Fatty acid methyl esters (FAMEs) composition

Extracted lipid from microalgal biomass after transesterification to biodiesel provides a potential alternative of fossil fuel and petroleum diesel (Likozar and Levec, 2014). Cultivation of microalgae in wastewater is a promising approach for providing biodiesel feedstock. In this research, lipids were extracted from microalgal biomasses after wastewater treatment. Since, the composition of fatty acids as the main component of biodiesel determines its properties, extracted FAMEs from cultivated *Sq* and *Ts* in BBM and F2 media and DWW were evaluated and presented in Table 2. In total, eight fatty acids namely, C14:0, C16:0, C16:1, C17:1, C18:2, C18:1n-7, C18:3n-3 and C18:3N-6 with different percentage were detected from microalgal lipid. C16 and C18 were the dominant fatty acids in *Sq*-BBM/DWW and *Ts*-F2/DWW.

Table 2
Fatty acid profile of *Scenedesmus quadricauda* and *Tetraselmis suecica* cultivated in synthetic media and dairy wastewater.

FAMEs composition (%)	*Sq*-BBM	*Sq*-DWW	*Ts*-F2	*Ts*-DWW
C14:0	n.d.[a]	1.65 ± 0.7	n.d.	2.41 ± 0.4
C16:0	46.43 ± 2.8	24.46 ± 0.7	14 ± 0.4	12.86 ± 1.4
C16:1	3.14 ± 0.4	5.31 ± 0.9	7.2 ± 0.1	5.99 ± 0.3
C17:1	8.76 ± 1.6	2.17 ± 0.2	21.18 ± 1.4	12.29 ± 0.5
C18:2	14.81 ± 5.6	16.99 ± 0.1	8 ± 0.7	25.15 ± 3.2
C18:1n-7	10.96 ± 1. 6	26.95 ± 0.4	n.d.	13.66 ± 1.5
C18:3n-3	15.89 ± 2.8	20.41 ± 0.4	49.61 ± 1.1	26.88 ± 1.5
C18:3n-6	n.d.	2.05 ± 0.1	n.d.	0.76 ± 1.1
Σ ω−3	15.89 ± 2.8	20.41 ± 0.4	49.61 ± 1.1	26.88 ± 1.5
Σ ω−6	14.81 ± 5.6	19.04 ± 0.1	8 ± 0.7	25.91 ± 2.6
Σ ω−3/Σ ω−6	1.1	1.1	6.2	1.1
Σ SFA[b]	46.43 ± 2.8	26.11 ± 1.4	14 ± 0.4	15.27 ± 1.8
Σ MUFA[c]	22.86 ± 3.6	34.43 ± 2.3	28.38 ± 1.4	31.94 ± 2.3
Σ PUFA[d]	30.7 ± 8.4	39.45 ± 0.5	57.61 ± 1.3	52.79 ± 4.1

[a] n.d. = not detected.
[b] SFAs = Saturated fatty acids.
[c] MUFAs = Monounsaturated fatty acids.
[d] PUFAs = Polyunsaturated fatty acids.

According to the literature, C16 and C18 are two major fatty acids that are produced by enzymatic procedure and carbon chain lengthening and desaturation reactions (Matos et al., 2017). Similar to the results of this study, Song et al. (2014) reported that C16-C18 with > 95% are the main portions of fatty acid profile in six studied microalgae strains (Song et al., 2014).

Table 2 presents the fatty acids percentage of freshwater and marine water microalgae. The fatty acid compositions of *Sq*-BBM, *Sq*-DWW, *Ts*-F2 and *Ts*-DWW had different amounts of saturated fatty acids (SFA), mono- and polyunsaturated fatty acids (MUFA and PUFA). The percentage of SFA (mainly C16) in both *Sq*-BBM and *Sq*-DWW was significantly higher than in *Ts*-F2 and *Ts*-DWW. On the contrary, higher percentage of PUFA was observed in *Ts* (F2 and DWW) as compared to *Sq* (BBM and DWW). The main reason of higher PUFAs in Ts (F2 and DWW) is referred to higher concentrations of C18:2 and C18:3n-3. In marine microalgae, PUFAs is synthesized by desaturation of saturated fatty acids mainly C18 and C16 to C18:2n-6 by Δ9 and Δ12 desaturases enzymes. Furthermore, C18:2n-6 can also be desaturated to C18:3n-3 by Δ15 desaturases enzymes (Monroig et al., 2013). In another study, Patil et al. (2007) investigated the fatty acid composition of nine marine and three freshwater microalgae (Patil et al., 2007). They reported that ω−3-PUFAs are the main fatty acids in marine microalgae while SFA and MUFA are dominant ones in freshwater species. Jaiswar et al. (2017) stated that different taxa, rigorous taxon and strains of microalgae are diverse in fatty acid compositions (Jaiswar et al., 2017).

Table 2 demonstrates the fatty acid profiles of cultivated microalgae in synthetic media (BBM and F2) and DWW. The results show significant changes in fatty acids proportions of each freshwater and marine water microalgae cultivated in wastewater as compared to synthetic media (BBM and F2). The ω−3 percentage of *Sq* cultivated in BBM was 15.89 ± 2.8%, while it was 20.41 ± 0.4% in DWW. However, high concentration of ω−3 (49.61 ± 1.1%) in *Ts*-F2 decreased to 26.88 ± 1.5% in *Ts*-DWW. Drastic differences in SFAs and MUFAs percentages of *Sq*, cultivated in BBM and DWW were noticed in this study. Higher SFAs and MUFAs percentages as 46.43 ± 2.8 and 34.43 ± 2.3% were observed in *Sq*-BBM and *Sq*-DWW, respectively. These results show that the composition of fatty acids of one species of microalga can be changed under different cultivation conditions. In the current study, the characteristics of BBM and DWW, such as the concentrations and sources of nutrients, organic carbon amount, color, microbial/bacterial loading and conductivity, were different. For example, the concentration of NO_3-N in BBM and DWW was 41.25 and 31.30 mg/L, respectively. Lower concentration of MUFAs (mainly C18:1n-7) in DWW as compared to BBM can be related to lower concentration of nitrate. Shen et al. explained that nitrogen limitation activates stearoyl-ACP desaturase, a gene responsible for synthesis of C18:1 fatty acid (Shen et al., 2016).

According to Table 2, *Sq* and *Ts* were cultivated in the same kind of wastewater, however, their fatty acid profiles were different. The amounts of ω−3, ω−6 and PUFA of *Ts* were higher than *Sq*; but the concentration of SFA in *Ts* was lower than *Sq*. Activity of different enzymes in synthesis pathway of fatty acids change the lipid composition of freshwater and marine water microalgae (Srinuanpan et al., 2018).

3.4. Tetracycline biosorption by $Sq_{biomass}$ and $Ts_{biomass}$ after lipid extraction

3.4.1. Effect of initial solution pH on TC removal efficiency

Fig. 3 depicts the effect of varying initial solution pH from 2 to 10 on TC biosorption from water by microalgal biomasses ($Sq_{biomass}$ and $Ts_{biomass}$) after lipid extraction. TC removal efficiency was increased dramatically from 0 to 48.84 ± 1.4% by $Sq_{biomass}$ and 0 to 36.71 ± 2.1% by $Ts_{biomass}$ as initial solution pH increased from 2 to 8. Then, removal efficiency of TC decreased as initial solution pH increased from 8 to 10.

Usually, biosorption process is a pH-dependent process. Initial

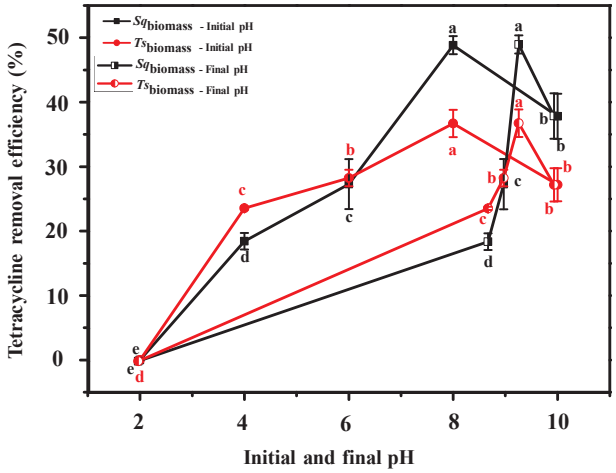

Fig. 3. Effect of initial and final solution pH on TC removal efficiency from water by microalgal biomasses after lipid extraction [Biosorbent dosage: 1 g/L, TC concentration: 10 mg/L, contact time: 180 min].

solution pH affects the surface charge of biosorbents ($Sq_{biomass}$ and $Ts_{biomass}$) and it affects the dissociation of TC. TC molecule is an amphiprotic compound that has different cationic (TCH_3^+), zwitterionic (TCH_2^0) and anionic (TC^{2-}) forms in pK_{a1} < 3.3, pK_{a2} = 3.3–7.7 and pK_{a3} > 7.7, respectively (Pi et al., 2017). As can be seen from Fig. 3, the initial solution pH increased from 2.00 to 2.17 ± 0.1 (< 3.3). In acidic pH, surface charge of biosorbents and TC are positive and electrostatic repulsion occurs. At pH higher than 7.7, TC removal efficiency decreases due to negatively charged biosorbent's surface and anionic form of TC. In this study, the initial TC solution pHs of 4 to 10 were shifted to the final pHs of 6.86 to 7.75 after 3 h of contact time. The lowest electrostatic repulsion is expected in this range of final pHs as zwitterionic (TCH_2^0) is the dominant form of TC and the surface charge of biosorbents is neutral. In agreement to the result of this study, Zhou et al. (2017) reported the highest TC removal efficiency by iron and zinc doped sawdust biochar at pH 6–7 as TCH_2^0 was the dominant form of TC (Zhou et al., 2017).

3.4.2. Effect of $Sq_{biomass}$ and $Ts_{biomass}$ amount on TC removal efficiency

Fig. 4 illustrates the removal efficiency of TC onto $Sq_{biomass}$ and $Ts_{biomass}$ by varying amounts from 0.25 to 4.00 g/L. TC removal efficiency by $Sq_{biomass}$ increased sharply from 21.50 ± 2.4 to 43.32 ± 3.6% as the biosorbent amount increased from 0.25 to 0.50 g/L. The highest TC removal efficiency was observed as 53.31 ± 3.8% by 2 g/L of $Sq_{biomass}$. However, removal efficiency decreased by increasing the $Sq_{biomass}$ from 2 to 4 g/L. Removal efficiency of TC onto $Ts_{biomass}$ gradually increased from 8.82 ± 1.7 to 54.90%, as $Ts_{biomass}$ amount increased from 0.25 to 2.00 g/L. Then, TC removal efficiency decreased to 52.13 ± 4.31% as biomass dosage increased to 4 g/L; which was not significantly different from 54.90% with 2 g/L biosorbent dose. By increasing the amount of $Sq_{biomass}$ *and* $Ts_{biomass}$, more adsorption sites and surface area will be available for TC biosorption. However, by further increasing the biosorbent amount (higher than optimum value) can consequently lead to aggregation and overlapping of biosorption sites and as a result, can decrease TC removal efficiency. Pouretedal and Sadegh (2014) reported similar trend of antibiotics removal from water using activated carbon nanoparticles (Pouretedal and Sadegh, 2014).

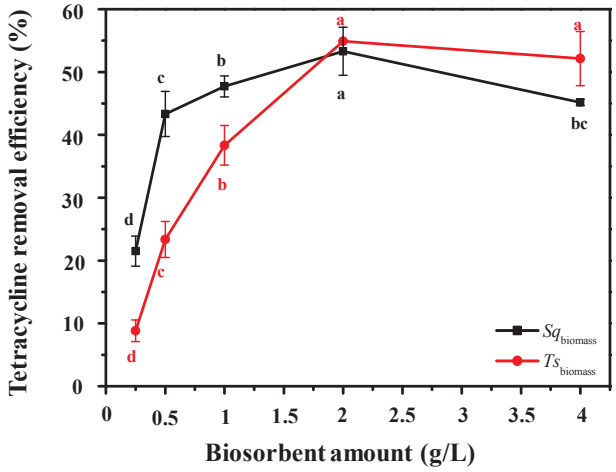

Fig. 4. Effect of biosorbent amount on TC removal efficiency from water by microalgal biomasses after lipid extraction [pH: 8, TC concentration: 10 mg/L, contact time: 180 min].

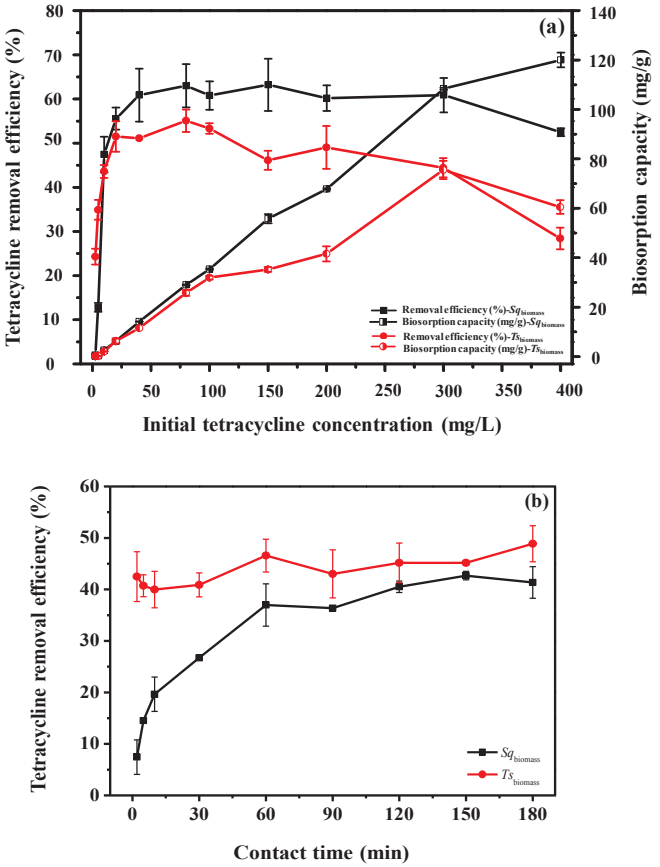

Fig. 5. Effect of TC concentration (a) and contact time (b) on TC removal efficiency from water by microalgal biomasses after lipid extraction [pH: 8, Biosorbent dosage: 2 g/L, TC concentration: 10 mg/L (in effect of contact time experiment), contact time: 180 min (in effect of initial tetracycline concertation experiment)].

3.4.3. Effect of initial TC concentration and contact time on TC removal efficiency

Fig. 5(a) shows the effect of initial concentration of TC on the TC removal efficiency and biosorption capacity (mg/g) by microalgal biomasses after lipid extraction. TC removal efficiency by $Sq_{biomass}$ increased sharply from 2.00 ± 0.2 to 62.97 ± 4.9% as initial TC concentration increased from 2.5 to 80 mg/L. The value of TC removal efficiency was almost constant by increasing TC initial concentration from 80 to 300 mg/L (62.97 ± 4.9–60.85 ± 3.9%). As can be seen from Fig. 5(a), TC removal efficiency decreased from 60.85 ± 3.9 to 52.46 ± 0.9% as initial concentration increased from 300 to 400 mg/L. As compared to $Sq_{biomass}$, $Ts_{biomass}$ showed higher removal efficiency of TC (24.29 ± 1.8%) at lower initial concentration (2.5 mg/L).

Maximum TC removal efficiency by $Ts_{biomass}$ was observed as 55.11 ± 2.5% at 80 mg/L of initial TC concentration. TC removal efficiency by $Ts_{biomass}$ decreased gradually as the initial concertation increased from 80 to 300 mg/L. Then, it sharply decreased from 44.44 ± 2.2 to 28.41 ± 2.4% as TC concentration increased from 300 to 400 mg/L. According to the results, it can be concluded that 80 mg/L of TC is the optimum concentration to attain the highest removal efficiency by $Sq_{biomass}$ and $Ts_{biomass}$. The reason of increasing TC removal efficiency by increasing of initial concentration (up to 80 mg/L) might be related to driving force which is the concentration gradient (Esmaeli et al., 2013). However, the availability of TC biosorption sites will be decreased due to saturation at very higher initial concentration. In another study, the removal efficiency of pharmaceutical and personal care products (PPCPs) was not affected by the lower concentration of PPCPs but it decreased at higher concentration of the same (Attia et al., 2013).

The effect of initial TC concentration on biosorption capacity of $Sq_{biomass}$ and $Ts_{biomass}$ was investigated (Fig. 5(a)). Biosorption capacity of both biosorbents increased linearly as initial TC concentration increased from 2.5 to 300 mg/L. These results revealed that for the higher concentrations of initial TC (> 80 mg/L) with lower removal efficiency, there is higher biosorption capacity. It can be due to driving force and more efficient collision of adsorbate/biosorbent at higher initial TC concentration. At higher concentration of TC (300 to 400 mg/L), biosorption capacity of $Ts_{biomass}$ decreased from 75.52 ± 3.7 to 60.54 ± 2.8 mg/g due to saturation of biosorbent.

Fig. 5(b) demonstrates the effect of contact time on the TC removal efficiency by $Sq_{biomass}$ and $Ts_{biomass}$. TC biosorption by $Sq_{biomass}$ dramatically increased during 60 min. After 60 min, TC removal efficiency slowly enhanced up to 120 min. Thereafter, the biosorption attained the equilibrium within 180 min. Higher TC removal efficiency in the beginning of biosorption process can be expected due to the availability and accessibility of more vacant biosorption sites on surface of $Sq_{biomass}$. After occupying the surface sites, biosorption of TC to inner layer of biosorbent is a slow process. $Ts_{biomass}$ revealed a different pattern of biosorption behavior in response to contact time in comparison with $Sq_{biomass}$. As Fig. 5(b) represents, TC removal by $Ts_{biomass}$ was not time-dependent at the investigated experimental condition. TC removal efficiency reached to 42.50 ± 2.1% after 2 min. Then, removal efficiency increased to 45.16 ± 0.4 and 48.87 ± 2.6% up to 150 and 180 min, respectively. Attia et al. (2013) also reported that contact time did not affect diclofenac-Na removal from water by magnetic nanoparticles coated zeolite (Attia et al., 2013). The reason of different patterns of biosorption between $Sq_{biomass}$ and $Ts_{biomass}$ can be related to different morphology and functional groups of freshwater and marine water microalgal biomasses.

3.4.4. Isotherm modeling

Isotherm models describe the biosorption process and interaction between adsorbate (TC) and biosorbents ($Sq_{biomass}$ and $Ts_{biomass}$). Here, three well-known isotherm models namely, Langmuir, Freundlich and Sips were used to analyze TC biosorption by microalgal biomasses. Table 3 presents the calculated values of the parameters and constants of these models. The applicability of the models is judged by the higher value of R^2, lower value of RMSE, and good fitting of calculated and experimental data (Daneshvar et al., 2017). As can be seen from Table 3 and Fig. 6, biosorption isotherm of TC onto microalgal biomasses is better described by both Sips and Langmuir models than Freundlich model. It can be concluded that the dominant process of TC biosorption is monolayer with contribution of heterogeneous surface of microalgal biomasses. According to the Langmuir model, maximum biosorption capacity of $Sq_{biomass}$ and $Ts_{biomass}$ towards TC was found to be 295.34 and 56.25 mg/g, respectively. These values are comparable with the other adsorbents used for TC removal, such as high surface area porous carbon material derived from human hair and ferric-activated sludge-based adsorbent with the maximum adsorption capacity of 162.20 and 87.87 mg/g, respectively (Ahmed et al., 2017; Yang et al., 2016).

Table 3
Langmuir, Freundlich and Sips isotherm models for biosorption of TC by microalgal biomasses after lipid extraction.

Isotherm models	Parameters	$Sq_{biomass}$	$Ts_{biomass}$
Langmuir	Q_0 (mg/g)	295.34	56.25
	b (dm^3/mg)	0.003	0.020
	R^2	0.989	0.980
	*RMSE	4.508	2.870
Freundlich	K_F (mg/g) (dm^3/g)$^{1/n}$	1.97	1.51
	n (−)	1.283	1.335
	R^2	0.987	0.993
	RMSE	6.196	2.446
Sips	K_S (L/g)	0.001	0.001
	q_m	163.97	42.42
	n	0.676	0.510
	R^2	0.996	0.991
	RMSE	3.100	1.765

* RMSE: Root Mean Square Error.

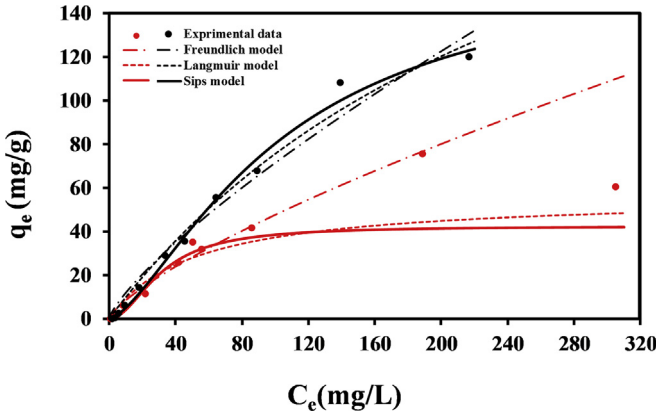

Fig. 6. Isotherm modeling for the biosorption of TC by microalgal biomasses after lipid extraction: $Sq_{biomass}$ (Black) and $Ts_{biomass}$ (Red) [pH: 8, Biosorbent dosage: 2 g/L, contact time: 180 min]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Conclusions

The results of this study demonstrated that microalgae grew well in DWW without adding nutrients. Maximum biomass weight of *Sq* (0.47 g/L) was observed after 8 days, but *Ts* growth was dynamic after 12 days. TN, PO_4^{3-}, SO_4^{2-} and TOC were efficiently removed from DWW by studied microalgae. The percentage of FAMEs as the main sources of biodiesel, were different in synthetic media and DWW. This research also showed the feasibility of using miocroalgal biomasses, after lipid extraction, for biosorption of TC. It can be concluded that microalgae as renewable resources can address environmental issues in a sustainable way.

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