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Changes in BOD, COD, Chlorophyll-a and Solids in Aquaculture Effluent with Various Chemical Treatments

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Four chemical treatments with hydrogen peroxide (H_2O_2), copper sulfate ($CuSO_4$), potassium permanganate ($KMnO_4$) and chlorine (Cl_2) were applied to the effluent pond water of a hybrid striped bass saltwater recirculating aquaculture system to compare their oxidation power. Four chemicals were applied at concentrations of 0 (control), 1, 5, 10 and 20 mg/l. An additional concentration of 40 mg/l was included in the chlorine treatment. Water samples from four hybrid striped bass ponds were tested with $KMnO_4$ and Cl_2. H_2O_2 did not reduce any of BOD, COD and chlorophyll-a, and copper sulfate was only effective on chlorophyll-a for the effluent pond. Removal efficiencies for chlorophyll-a by copper sulfate were 19.2%, 37.5%, 54.2% and 74.1% dose-dependently. Potassium permanganate effectively removed the BOD, COD and chlorophyll-a. The COD removal rates in four fish ponds varied from 15.9% to 31.6% at the concentration of 10 mg/l. Interestingly, Cl_2 did not reduce the BOD and COD at all, but the BOD and COD instead increased drastically with increasing the Cl_2 concentration. The pond water with the highest initial BOD and COD values among the fish ponds tested increased by 350% in the BOD and 150% in the COD at 20 mg/l. Furthermore, Cl_2 did not significantly reduce any types of solid matter in this study, while $KMnO_4$ seemed to reduce some extent volatile dissolved solid in the fish pond.

Keywords: Chemical treatment, Hybrid striped bass, Pond water, BOD, COD, Chl-a, Solids

Introduction

Since the fish farming of hybrid striped bass in the US started in the late 1980, it has greatly expanded to the industrial scale of approximately 30 million dollars in farm-gate value (Dasgupta and Thompson, 2013). The majority of hybrid striped bass are cultured in earthen ponds (Daniels, 2005). This is the most economical means of producing these fish, especially in rural areas of eastern North Carolina where the flat topography, high clay content of the soils and abundance of groundwater are ideally suited for this type of aquaculture production. However, cage culture is also practiced filling a small niche in southern Appalachian states with deep and watershed ponds that are not suitable for seining (Dasgupta and Thompson, 2013).

Feeds for hybrid striped bass are formulated to contain optimum nutrient levels, and fish are highly efficient at converting feed into nutrients while generating little waste product (Lochmann, 2015). However, some un-eaten feed or metabolites from the fish inevitably pass into the pond water. These waste products primarily contribute to the organic load, increasing biological and chemical oxygen demand, blooming algae, and turning the pond water green (Jayaram and Beamish, 1992). Occasionally, effluent is released from the ponds into drainage ditches and eventually enters public waterways. The most significant release of effluent occurs when ponds are drained between crops (Schwartz and Boyd, 1994). Since effluents from fish ponds are regulated under the National Pollutant Discharge Elimination System (NPDES) from 1973 in the US, all fish farms that discharge effluent into public waterways must be permitted (Schwartz and Boyd, 1994). Under the NPDES, the pond effluent with over 40 μg/l of chlorophyll-a, and measurable biochemical oxygen demand (BOD >5 mg/l) cannot be discharged. Based on past research, however, most pond effluent will not meet these standards, so that the appropriate effluent treatment methods are required.

Many chemicals are frequently used as the pond water treatment agents to improve soil and water quality and to control phytoplankton blooms and disease vectors (Boyd, 1995; Boyd and Tucker, 1998). Although the use of chemicals for pond water treatment often draws public concerns due to its potential impact on the culture animals, it would be still recognized as an effective method for water treatment between crops when no fish are present. Nevertheless, it is rare to find out the studies about the chemical oxidation efficiency on organics and phytoplankton removal. Therefore, the purpose of this study was to evaluate the oxidation efficiency of some chemicals which are known to reduce the organics or phytoplankton. The response of BOD, COD, chlorophyll-a and solids fraction with chemical treatment for the pond water were observed as indicators for oxidation efficiency of the chemicals.

Materials and Methods

1. Pond water and chemical treatment

1) Oxidation efficiency of chemicals

Water samples were collected from the effluent pond of a hybrid striped bass saltwater recalculating system at the Lake Wheeler Fish Barn of North Carolina State University to evaluate the oxidation efficiency of the different commonly used chemicals. The four chemicals used were hydrogen peroxide (H_2O_2), copper sulfate ($CuSO_4$), potassium permanganate ($KMnO_4$) and chlorine (sodium hypochlorite; Cl_2). Each chemical was applied at 0 (control), 1, 5, 10 and 20 mg/l, and the concentration of 40 mg/l was added to chlorine treatment. The water samples for each chemical treatment were collected separately over a short period so that the water quality variable remained within similar ranges.

2) Saltwater fish pond water treatment with $KMnO_4$ and Cl_2

Water samples from four hybrid striped bass ponds were treated with $KMnO_4$ and Cl_2. The treatment concentrations were 0 (control), 1, 5, 10 and 20 mg/l. Four water samples used in this study were taken from a fish pond at the Tidewater Research Station of North Carolina Department of Agriculture (P1), two commercial fish ponds (P2, P3) at Pinetown, North Carolina and a commercial fish pond (P4) at Aurora, North Carolina, from February to March.

3) Chemical preparation and treatment procedure

All chemicals were reagent grade purchased from a commercial supplier (Fisher Scientific). Hydrogen peroxide was added at the required amount from a 50% H_2O_2 extra pure stabilized solution. Stock solutions of 5,000 mg/l were used for $CuSO_4$ and $KMnO_4$. A 10,000 mg/l stock solution of hypochlorite was used as the chlorine source. The hydrogen peroxide and chlorine concentration were quantified with permanganate titrimetric methods and DPD ferrous titrimetric method, respectively just before the experiment. Water temperature and pH of the sample waters were first adjusted at 20℃ and 8.5. The sample waters were divided into one-liter bottles, and the chemicals were added into the bottles at each concentration. The experiment was performed in triplicate for each treatment, and the contact times after adding the chemicals were 6 hours for $KMnO_4$, H_2O_2 and Cl_2, but 18 hours for $CuSO_4$. All the sample waters were vigorously shaken to vent out any residual gaseous chemicals before water quality analysis.

2. Water quality and statistical analyses

The BOD, COD and chlorophyll-a were measured for all water samples (APHA 1995), and the solid matters were measured for samples from three commercial fish ponds (P2, P3 and P4).

The 5 day BOD test was measured with 300 ml BOD bottles according to the APHA (1995). The water samples were agitated until the oxygen concentration increased up to 8.5 mg/l. At the end of contact time, the water samples including the control were seeded with the sample waters not treated, and the contribution of the seed was deducted in BOD calculation. The water sample were also diluted by 0~75% to with the distilled water, depending on the enrichment of the sample waters.

The COD was determined by the heat of dilution method with sulfuric acid and potassium dichromate. (Boyd, 1979). All glassware was washed with H_2SO_4-$Na_2Cr_2O_7$ combined cleaning solution, and rinsed with distilled water immediately prior to use.

The Acetone-Methanol extraction method was used to determine the chlorophyll-a concentration (Picharr, 1987). Chlorophyll-a was extracted in a hot mixture of acetone and methanol after filtering with glass fiber filters (Gelman Sciences AE, 47 mm). The extraction was acidified with 0.1 N HCl, and the difference between readings before and after acidification in absorbance at 665 nm were measured to calculate the concentration.

Total solids, total volatile solids, total suspended solids, total dissolved solids, volatile suspended solids and volatile dissolved

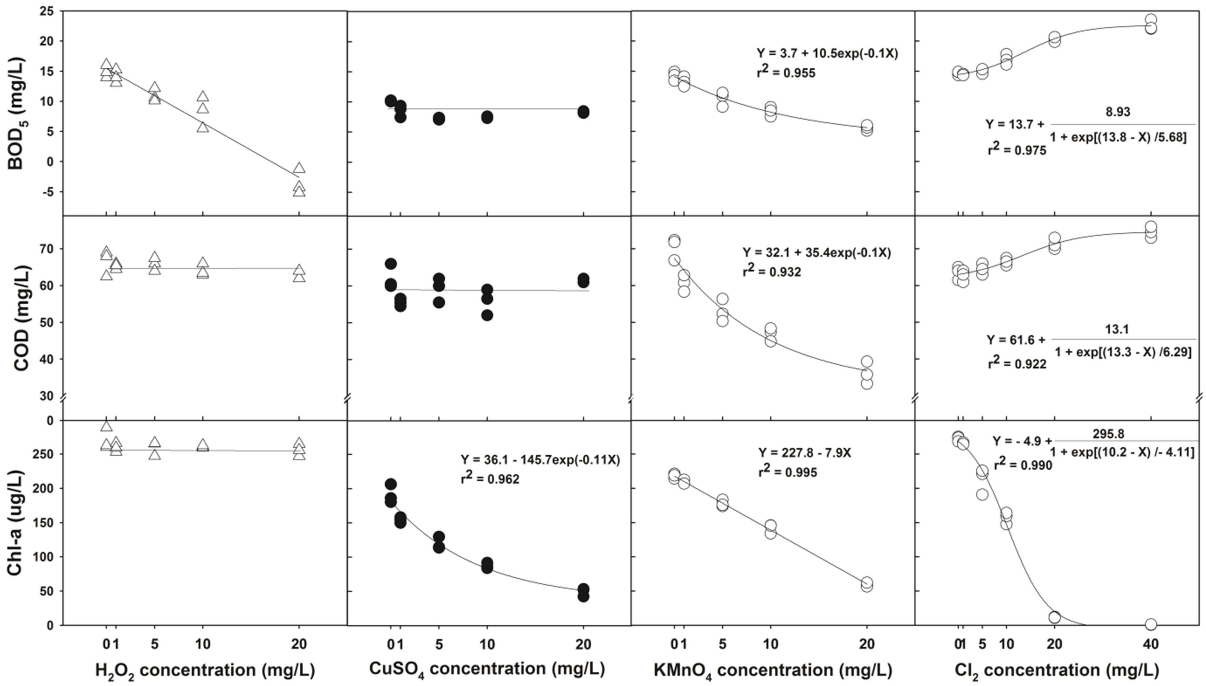

Fig. 1. Changes of BOD, COD and chlorophyll-a concentrations in the effluent pond water of a hybrid striped bass recirculating aquaculture system treated with H_2O_2, $CuSO_4$, $KMnO_4$ and Cl_2.

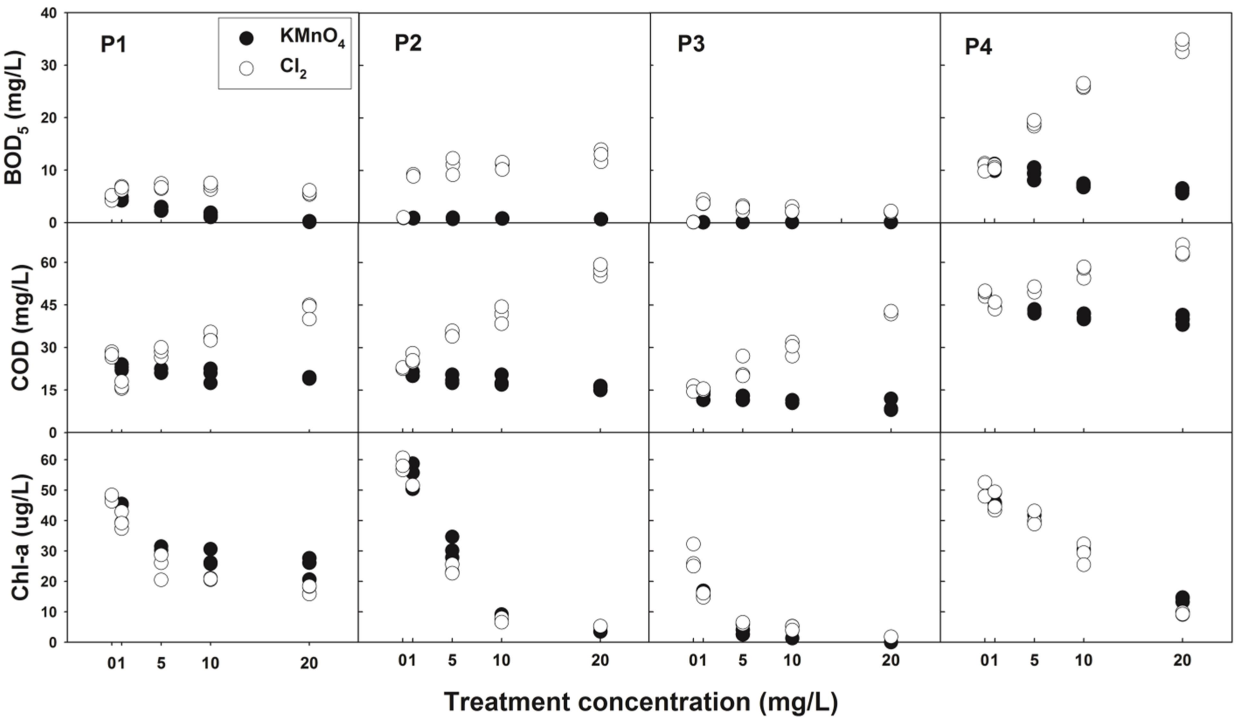

Fig. 2. Changes of the BOD, COD and chlorophyll-a concentration in four hybrid striped bass pond waters treated with $KMnO_4$ and Cl_2.

solids were determined according to the APHA (1995).

One-way ANOVA tests were performed to evaluate the effects of $KMnO_4$ and Cl_2 treatments on solids fractions in the commercial fish pond waters using SPSS 12.0 for Windows (SPSS Inc., Chicago, IL, USA). Duncan's multiple range test was used for detecting significant differences among the averages (p < 0.05).

Results and Discussion

Hydrogen peroxide has been used to reduce the BOD, COD and offensive odor in domestic or industrial wastewater for many years (Ksibi, 2006). There has been also some interest on H_2O_2 as an oxidizing agent in aquaculture to reduce the BOD and COD in the pond water. However, H_2O_2 removed neither the BOD, COD, nor chlorophyll-a within the treatment concentration used in this study (Fig. 1). Although the BOD concentration greatly decreased with H_2O_2 treatment, apparently the residual oxygen atom released from the H_2O_2 and captured in BOD bottles increased the oxygen concentration. This was supported by the fact that the BOD concentration in the highest treatment concentration greatly exceeded the initial value, and the COD concentration did not change with the treatment. The bottles with H_2O_2 treatment were vigorously shaken after 6 hours and left open to the air for an hour before the BOD setup. Nevertheless, increases in DO concentration in the BOD bottles with the treatment were still observed in an additional experiment for validation. Ksibi (2006) reported reduced COD in domestic wastewater with H_2O_2 treatment. However, the treatment concentrations showing more than 30~80% reduction were 150~450 mg H_2O_2/l, which were much higher concentrations compared to those in the present study. The concentration of 75 mg H_2O_2/l that was the lowest in the previous study, but was much higher than the highest value in this study, showed only 5% COD reduction. Furthermore, Brik et al. (2004) also reported that the maximum reduction in color material of textile wastewater was only less than 10% at 250 mg H_2O_2/l. Vänninen and Koskula (1998) demonstrated that a single application of 125 mg H_2O_2/l to one week old cucumber seedling or daily applications over three weeks of 100 mg H_2O_2/l reduced algal growth by 40~60%. After algae were already grown up intensively, however, a concentration of 1,500 mg H_2O_2/l could not eliminate the algae or reduce its further growth in the study. Based on the previous study and this study, the effective concentration to reduce the organic matter or control the algae with H_2O_2 treatment would be much higher than the concentration used in this study, which is unlikely to be acceptable for fish pond or effluent water treatment.

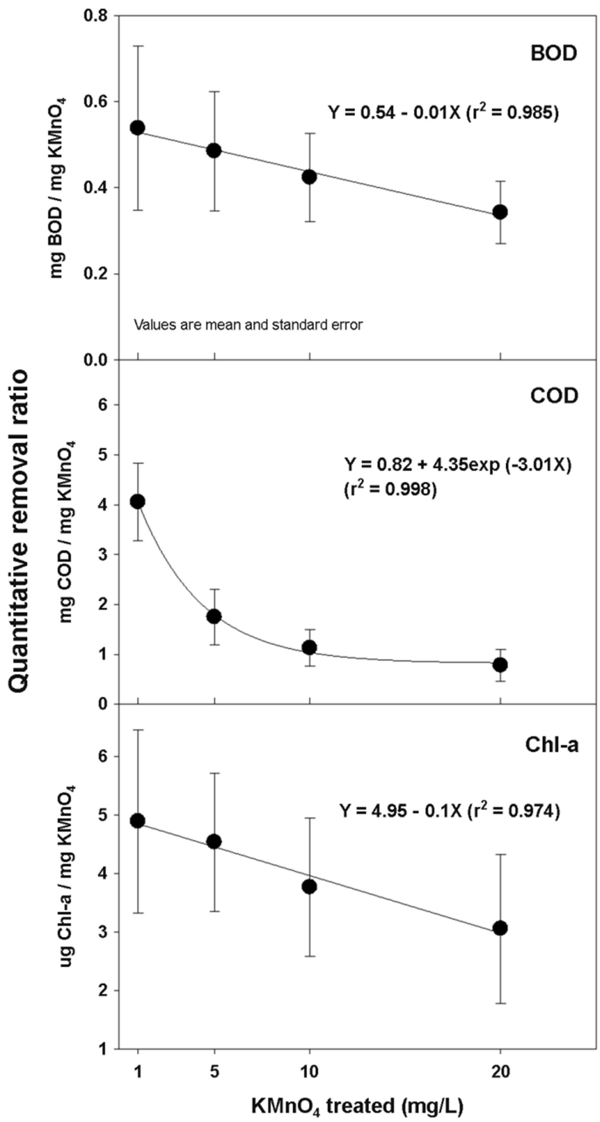

Fig. 3. Quantitative removal ratios of BOD, COD and Chlorophyll-a with $KMnO_4$ treatment.

Copper sulfate did not reduce either the BOD or COD, but reduced chlorophyll-a. Copper sulfate has been recognized as a strong algaecide, and many authors have demonstrated its toxicity to algae (Bartlett et al., 1974; Levy et al., 2009; Mischke et al., 2009). The removal efficiencies of $CuSO_4$ were 19.2%, 37.5%, 54.2% and 74.1% at the treatment concentrations of 1, 5, 10 and 20 mg $CuSO_4$/l. The values were relatively much lower than the values 13%, 37% and 53% at 0.5, 1 and 2 mg $CuSO_4$/l, reported by Palmer (1956). Many studies have addressed different effects of $CuSO_4$ on algae as a function of pH, alkalinity, temperature, and exposure opportunity (Bartsch, 1954; Fitzgerald and Faust, 1963; Takamura et al., 1990; Mischke et al., 2009). In addition, the toxicity of $CuSO_4$ may vary even at the same treatment concentration within same genus or species, as copper absorption into the algae is governed by competitive binding at the cell surface (Webster and Gadd, 1996; Wilde et al., 2006; Mischke et al., 2009).

Potassium permanganate was most effective in reducing the BOD, COD and chlorophyll-a of the effluent pond and fish pond waters in this study (Fig. 2). Potassium permanganate is known as a strong oxidizer that reacts with organic matter, and even reduces inorganic substances in water (Boyd, 1979), consequently reducing the rate of oxygen consumption by biological and chemical processes (Lay, 1971). Tucker and Boyd (1977) reported slight reduction of BOD and COD levels with $KMnO_4$ treatment. The dissolved oxygen levels after four days in pond water with the COD level under 100 mg/l remained high after treatment with $KMnO_4$ concentrations of 4 and 8 mg/l. In addition, the COD level was reduced by 15% at a $KMnO_4$ concentration of 8 mg/l. The removal efficiency with $KMnO_4$ for the effluent pond in this study was 33.2% at 10 mg/l, which is higher than the value reported by Tucker and Boyd (1977). In the meantime, the COD removal rates in four fish ponds greatly varied from 15.9% to 31.6% at the treatment concentration of 10 mg/l. The effectiveness of chemicals can vary with the other water qualities such as pH, water temperature and dissolved oxygen, and the amount of consumption of the organic matter in water. Furthermore, the sample water used in the experiment was previously treated with the geo-textile bag and ozone treatment before being released to the pond, and the bio-chemical degradability would be enhanced to show the better removal efficiency for the BOD and COD. As many authors have

Table 1. Solids fractions in the commercial fish pond waters treated with $KMnO_4$ and Cl_2

Pond	Solid	Control (mg/l)	$KMnO_4$ (mg/l)				p	Cl_2 (mg/l)				p
			1	5	10	20		1	5	10	20	
P2	TS	1170.0±124.4	1041.1±137.7	1062.2±132.4	1045.6±94.5	1035.6±100.8	0.628	1161.1±57.4	1138.9±106.6	1136.7±82.1	1118.9±106.3	0.968
	TDS	932.2±22.7^{a}	902.2±24.1^{a}	912.2±53.5^{ab}	807.8±17.1^{bc}	796.7±99.1^{c}	0.029	862.2±20.4	843.3±95.6	832.2±22.2	824.4±23.4	0.105
	TSS	66.7±5.1^{c}	72.3±4.7^{bc}	76.7±2.3^{b}	78.0±4.6^{b}	87.7±6.4^{a}	0.004	65.0±7.0	65.7±7.8	63.0±4.0	67.3±5.8	0.917
	TVS	315.6±23.6^{a}	285.6±28.3^{ab}	264.4±36.7^{ab}	265.6±47.3^{ab}	234.4±13.9^{b}	0.099	300.0±26.5	289.9±40.4	272.2±39.1	268.9±40.3	0.479
	VDS	232.2±13.5^{a}	221.1±1.9^{a}	205.6±38.5^{ab}	175.6±13.9^{bc}	158.9±11.7^{c}	0.006	230.0±15.3	223.3±24.0	204.4±12.6	196.7±24.0	0.146
	VSS	43.4±5.9^{a}	43.3±3.3^{ab}	45.6±13.5^{b}	50.0±3.3^{b}	60.0±6.7^{b}	0.100	44.4±4.7	45.6±5.7	44.4±4.7	47.8±5.8	0.994
P3	TS	625.6±57.5	603.3±86.9	584.4±70.7	578.9±61.9	583.3±70.6	0.915	621.1±48.6	607.8±15.0	596.9±8.4	583.3±21.9	0.607
	TDS	476.7±14.5^{a}	424.4±50.9^{ab}	415.6±35.6^{ab}	402.2±31.0^{b}	381.1±31.0^{b}	0.063	466.7±32.1	457.8±21.7	431.1±19.0	433.3±35.1	0.199
	TSS	99.0±5.2^{c}	105.0±10.5^{bc}	112.3±7.5^{abc}	119.7±9.9^{ab}	123.7±5.5^{a}	0.020	111.0±6.2	114.3±12.5	113.0±10.8	110.0±9.8	0.342
	TVS	107.8±12.6	98.9±6.9	91.1±23.6	80.0±15.3	76.7±6.7	0.119	104.4±15.9	101.1±6.9	97.8±5.1	91.1±8.4	0.653
	VDS	93.3±6.7^{a}	83.3±14.5^{ab}	73.3±3.3^{b}	74.4±5.1^{b}	70.0±5.8^{b}	0.031	92.2±10.7	90.0±3.3	87.8±6.9	84.4±9.6	0.648
	VSS	44.4±6.9^{c}	53.3±17.6^{bc}	64.4±7.7ab^{c}	71.1±6.9^{ab}	75.0±11.7^{a}	0.035	45.6±5.1	48.9±9.6	50.0±5.8	52.2±5.1	0.628
P4	TS	1412.2±13.5	1405.6±74.0	1397.8±93.8	1402.2±76.9	1390.0±83.7	0.997	1393.3±24.0	1422.2±80.4	1394.4±85.3	1417.8±17.1	0.944
	TDS	1374.7±34.9^{a}	1344.0±34.9^{ab}	1300.0±18.3^{abc}	1306.7±58.3^{bc}	1253.3±30.3^{c}	0.025	1368.0±17.4	1334.7±82.4	1265.3±26.0	1361.3±58.0	0.115
	TSS	55.0±1.0^{d}	60.7±3.1^{cd}	66.0±3.5^{bc}	71.3±2.5^{b}	88.3±8.1^{a}	0.000	53.3±2.3	56.7±3.2	56.7±3.1	57.3±2.1	0.332
	TVS	375.6±17.1^{a}	363.3±34.6^{a}	337.8±35.0^{ab}	325.6±25.5^{ab}	298.9±26.7^{b}	0.053	365.6±21.4	334.4±20.1	340.0±31.8	332.2±28.0	0.231
	VDS	344.0±28.8^{a}	332.0±34.9^{ab}	298.7±10.1^{b}	294.7±6.1^{b}	252.0±12.0^{c}	0.003	348.0±28.0	330.7±34.5	314.7±47.7	305.3±40.1	0.544
	VSS	26.7±1.2^{b}	27.3±2.9^{b}	30.7±1.5^{ab}	32.3±5.7^{ab}	34.3±3.1^{a}	0.074	23.3±4.6	24.7±2.5	25.3±3.2	29.3±4.6	0.339

Values are means ± SD (cm, g) of triplicate groups. Means ± SD within the same columns for each of potassium permanganate or chlorine treatment with the same superscript are not significantly different at p < 0.1, based on Duncan's multiple range test reported the effectiveness of $KMnO_4$ to remove algae from water (Fitzgerald, 1964; Kemp et al., 1966; Ma et al., 1997; Drikas et al., 2001; Knappe et al., 2004), the chlorophyll-a levels greatly decreased with $KMnO_4$ treatment for both of effluent and fish ponds in this study.

The chlorination effects on the organic matter in wastewater have been assessed by measuring the BOD and COD in last decades. It was generally accepted that chlorination slightly reduced the parameters presenting some benefits for removal of organic matters (El-Rehaili, 1995). In this study, Cl_2 reduced the chlorophyll-a above the treatment concentration of 20 mg Cl_2/l. However, interestingly, Cl_2 could not reduce the BOD and COD at all, but the BOD and COD instead increased drastically with increasing treatment concentration. The increases in the BOD and COD after Cl_2 treatment were also observed in all of the hybrid striped bass pond waters. Although the BOD levels at P1 and P3 did not greatly increase with increasing Cl_2 concentration, the levels after treatment were still higher than the values in the control. Specifically, the P4, with the highest initial BOD and COD values among the fish ponds tested, presented increases of 350% in BOD and 150% in COD at 20 mg Cl_2/l. El-Rehalili (1995) has reported that chlorination increased the BOD and COD together on secondary effluent wastewater with increasing Cl_2 concentration. The BOD and COD values in the raw wastewater were 29 mg/l of BOD and 74 mg/l of COD which were similar with this study, and those values increased to 35 mg/l and 85 mg/l after 10 mg/l of chlorine treatment. At the higher chlorine concentration of 20 mg/l, the values increased to 40 mg/l and 118 mg/l respectively, so that the tendency was coincident with this study (Fig. 3). Chlorine which is one of the strongest oxidizing agents and commonly used for water disinfection seemed to change the biological and chemical properties of the water, specifically its organic characteristics. Basically, the BOD and COD used for measuring the organic content in water is an indirect measure which does not represent any chemical identity or status of organic matter by themselves (Jolley, 1975; Michael et al., 1991; El-Rehalili, 1995). For these increases in the BOD and COD, El-Rehalili (1995) has presumed that Cl_2 treatment might result in modifying the organic matrix to render it more biodegradable and amenable to oxidation in the BOD and COD tests.

The changes of the solid fraction in the commercial fish pond waters treated with $KMnO_4$ and Cl_2 would help understanding the changes of the BOD and COD. In this study, although the total solids (TS) were not significantly changed with either chemical treatment ($p > 0.1$, Table 1), Potassium permanganate seemed to reduce to some extent volatile dissolved solids (VDS). Furthermore, $KMnO_4$ treatment was likely to increase the suspended solids, so that VDS could be reduced with $KMnO_4$ treatment by both oxidation and coagulation. Thus, the reduction in BOD and COD with $KMnO_4$ treatment could be attributed to the oxidation of VDS, which is a major component of the organic part of solid matters. Unlike $KMnO_4$, although the solids measuring method would be somewhat rough to precisely identify the differences derived from the chemical treatment and there was decreasing tendency in VDS at high treatment concentration, Cl_2 could not significantly reduce any types of solid matters in this study, which had not been considered ($p > 0.1$). El-Rehalili (1995) also mentioned that total organic carbon (TOC) concentration in the secondary wastewater did not change after chlorination even in the high dose of 30 mg/l, which has a thread of connection with the solids fraction result in this study. Therefore, it is quite acceptable that Cl_2 may not reduce the organic part of the solid, but change the property of the organic matters more to a bio-chemically degradable form to increase the apparent BOD and COD levels.

During this study, $CuSO_4$ and Cl_2 could reduce only chlorophyll-a, while $KMnO_4$ reduced both organic matters and chlorophyll-a. However, the removal ratios of the BOD, COD and chlorophyll-a to 1 mg of $KMnO_4$ (quantitative removal ratio) greatly decreased with increasing treatment concentration. Thus, the $KMnO_4$ was more required to remove further BOD, COD and chlorophyll-a. These changes in the quantitative removal ratio may be caused by a relative increase of non-degradable organic matter resulting in more $KMnO_4$ demands.

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