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Influence of Commercial-Scale Seaweed Cultivation on Water Quality: A Case Study in a Typical Laver Culture Area of the Yellow Sea, North China

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Abstract: Seaweeds are important to marine ecosystems through biogeochemical processes. Laver are the most widely farmed seaweeds with the largest culture area in China. This study analyzes the water quality characteristics in a large-scale laver culture area (Taoluo) by comparing a small-scale laver culture area and non-culture areas, thereby assessing the changes in water quality due to large-scale laver cultivation. Particulate organic carbon and/or dissolved organic carbon decreased while the total suspended solid increased seasonally or with the distance from the coast. The concentrations of total nitrogen as well as dissolved inorganic nitrogen and phosphorus were generally higher near the shore and decreased seasonally in Taoluo. Substantial spatial variation in nutrient parameters between culture and non-culture sites was observed. Moreover, significant variations between culture and non-culture sites on a spatio-temporal scale were mostly observed in December compared with September and October. Furthermore, more clusters were found in December based on the water quality characteristics in various sampling sites using a hierarchical clustering analysis. These results suggested that more spatial deviation in water quality parameters between culture and non-culture sites were found in December; thus it can be hypothesized that the changes in water quality due to large-scale cultivation for laver was likely to occur in northern China in winter, i.e., the period of best growth status for the cold-temperate species of laver (e.g., *Neopyropia yezoensis*). We hope that this study can help to further understand the effects of seaweed farming on marine environments.

Keywords: *Neopyropia yezoensis*; *Neoporphyra haitanensis*; cultivation; hydrochemistry

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1. Introduction

The world aquaculture industry has grown dramatically over the last five decades. In China, the annul production of mariculture is about 3 times of that from fishing [1]. On one hand, aquaculture provides products with high quality proteins and other nutrients. As a result, the living standards of human beings have been greatly improved. On the other hand, the rapid growth of aquaculture has brought environmental problems [2]. Especially in coastal environments which is closely associated with intensive human economic activities except for aquaculture and receives most of the water runoff of organic and inorganic wastes from land through river or ground water systems [3]. One of the largest impacts of aquaculture is that the effluents, consisting mainly of nitrogen (N) and phosphorus (P), lead to an imbalance in nutrients dynamics and eutrophic conditions [4]. For example, the production of 1000 kg fish can lead to discharge of about 56.4 kg N and 7.0 kg P from the feed containing 45% protein and 1% P with the food conversion rate of 1.2 in a pen-based salmon aquaculture [5]. Eutrophication, with an excessive amount of N, P, and CO_2 and an insufficient amount of dissolved O_2 (DO), is becoming a serious problem in coastal marine environments in many parts of the world [3]. Marine plants play a vital role in maintaining the balance of marine environments. Microalgae and seaweeds have enormous potential for reducing global warming and climate change. Some cultivated seaweeds have very high productivity, absorb a large amounts of N, P, and CO_2, and produce a lot of O_2. They are periodically planted and harvested, thus uptake the nutrients from marine environments and assimilate N and P into nutritious chemical compounds for mankind. Cultivated seaweeds have been nominated as an essential part of sustainable aquaculture and applied for integrated multi-trophic aquaculture (IMTA) and bioremediation [3,5–9].

Within the world aquaculture sector, the cultivation of aquatic plants, dominated by seaweeds, is expanding by almost 8% per year over the past decade [10]. China is the biggest seaweed producer in the world, with the annual output of 2.35×10^6 tons in 2018. Nevertheless, the annual output of cultivated seaweeds accounted for only 6.04% of the total production from Chinese mariculture. The highest annual output (55.23%) came from fish and the second (37.18%) came from shellfish. The culture area of seaweeds accounted for 7.06% of Chinese marine aquaculture [1]. Farming seaweeds not only absorb nutrients, provide food, and raw material, but also have important ecological functions, such as serving as habitat and nursery grounds for juvenile marine animals and protection from predators [11–13]. There are still large requirements for seaweeds culture in China from both economical and ecological aspects.

Several seaweed species are cultured at industrial scales in China. Approximately 2/3 of the farmed seaweed production is from kelp (*Saccharina japonica*), and the second highest production is from *Gracilaria*. The annual production of laver (*Neopyropia yezoensis* and *Neoporphyra haitanensis*) is almost equal to that of *Undaria pinnatifida*, ranking 3rd of the total farmed seaweed production in China [1]. Laver contains high levels of nutrient components such as proteins, essential amino acids (EAA), and a high percent of eicosapenatenoic acid (EPA) in total fatty acids [14–16]. There is a long history of laver utilization for the Chinese, Korean, and Japanese [17]. Laver has become a more and more popular daily food and the most dominant cultivated seaweed in these countries. Although the annual production of laver is much lower than that of kelp, the culture area of laver has been expanding over the past decade in China and it is still increasing. *N. yezoensis* and *N. haitanensis* have become the most widely farmed seaweed species since 2010 in China, with a culture area accounting for over one half of all the farmed seaweeds. Since the 1980s, efforts have also been made to establish sea farming of laver in western countries, based on the increasing market for this food source [17,18].

Seaweeds have attracted much attention for bioremediation and to establish integrated aquaculture. Matos et al. [7] studied the uptake efficiency of nutrients (NUE, ammonium) in three red seaweeds, *Palmaria palmata*, *Chondrus crispus* and *Gracilaria bursa pastoris*, in northern Portugal by culturing the algae in a cascade tank system using the nutrient-rich effluents from a local turbot and sea bass farm. A seasonal and species specific NUE capacity was observed. The highest NUE (76.7% of NH_4-N) was obtained in *G. bursa pastoris*. Three estuarine macroalgae (*Ulva rotundata*, *Enteromorpha intestinalis*, and *Gracilaria gracilis*) were cultivated in the laboratory to assess their biofiltering capacities for ammonium N and phosphate P in waste effluents from a sea bass cultivation tank [6,19]. The net N and P uptake rate was found to be significantly affected by the water flow [6,19]. Both the maximum velocity of N and P uptake was found in *U. rotundata* [6]. Undoubtedly, all these seaweeds have a high NUE of N and P in the experiments under controlled inflow and outflow conditions. The uptake efficiency and/or rate differs among different species [6,7,19,20]. The uptake capacity of dissolved inorganic nitrogen and phosphorus in laver has been investigated in order for bioremediation and to develop a laver-fish integrated aquaculture [21–23]. It has been found that laver (*Pyropia tenera* and *Pyropia seriata*) has much higher uptake rates of ammonia than *Ulva pertusa*, *Hypnea charoides* and *Gracilaria* (*Gracilaria verrucosa* and *Gracilaria textorii*) [5]. Laver seems to be more efficient in removing dissolved inorganic nitrogen and phosphorus under the same experimental conditions [21].

The experiments were mostly conducted under controlled nutrients levels or controlled inflow and outflow conditions. The nutrients in the open sea change temporally and spatially. There are very limited reports regarding the influence of large-scale seaweeds cultivation on environmental factors in the open sea. Laver has been cultivated so widely and in such a large scale in China; it is necessary to investigate the impact of large-scale cultivation of laver on environment. He et al. [24] reported the efficiency of *N. yezoensis* in removing dissolved inorganic nutrients in the open sea in Lusi, the present largest laver farm with over 10^4 ha. Distinct removal of dissolved inorganic nitrogen and phosphorus was detected. However, the influence of laver farming on the other important environmental factors such as organic carbon, DO, and pH was not reported in the publication [24]. Indeed, it is difficult to quantify the exact NUE in an open-water system since there are many complex factors influencing the biogeochemical process as well as the influx and efflux. Further studies on the ecological effects of open sea seaweed farming is still needed. In this study, two open sea regions with different laver culture areas were chosen to monitor the changes in the environmental factors during laver culture. The correlation between laver culture and the change in some environmental factors was discussed. We hope that this study can help to further understand the effects of seaweed farming on water quality in an open-sea system.

2. Materials and Methods

2.1. Study Areas

Two laver farms in Shandong Province, China, were investigated in the present study, one of which was a typical laver farm covering approximately 40 ha at the coast of Taoluo (TL-LF) (between latitudes of 35°14.04′ N and 35°15.68′ N and longitudes of 119°24.44′ E and 119°26.44′ E), Rizhao city, the other was a small-scale experimental laver farm covering 0.067 ha at the coast of Muping (MP-LF) (between latitudes of 37°28.26′ N and 37°28.80′ N and longitudes of 121°47.65′ E and 121°47.82′ E), Yantai city (Figure 1). The sampling was carried out in 2019 for the study.

2.2. Sampling Design

This manuscript analyzes the water quality characteristics in a large-scale laver culture area by comparing with a small-scale laver culture area and non-culture areas, thereby assessing the changes in water quality due to large-scale laver cultivation. For this purpose, the current study included two different laver culture areas (TL-LF, MP-LF) and several non-culture areas (Figure 1). MP-LF had two non-culture control sites (i.e., nearshore and offshore control sites, represented as MP-NS and MP-OS, respectively) approximately 100 m outside it (Figure 1B). Similarly, TL-LF also had two non-culture control sites approximately 1 km outside it, represented as TL-NS and TL-OS, respectively (Figure 1C). In order to further ascertain whether the difference in water quality between the control sites and the culture area can be exclusively considered as an aftermath of laver cultivation, a reference site approximately 2 km away from TL-LF with no culture activities yet with a similar hydrographic setting was also included in the study, which was divided into three sampling sites from nearshore to offshore, represented by TL-CN, TL-CM, and TL-CO, respectively (Figure 1C).

The *N. haitanensis* cultivation began in August and ended in October in TL-LF and MP-LF. The *N. yezoensis* cultivation began in October and ended in December in MP-LF, and began in October and ended in March in TL-LF. Therefore, the sampling was also carried out in September, October, and December to take an account of temporal changes imposed on the water quality characteristics.

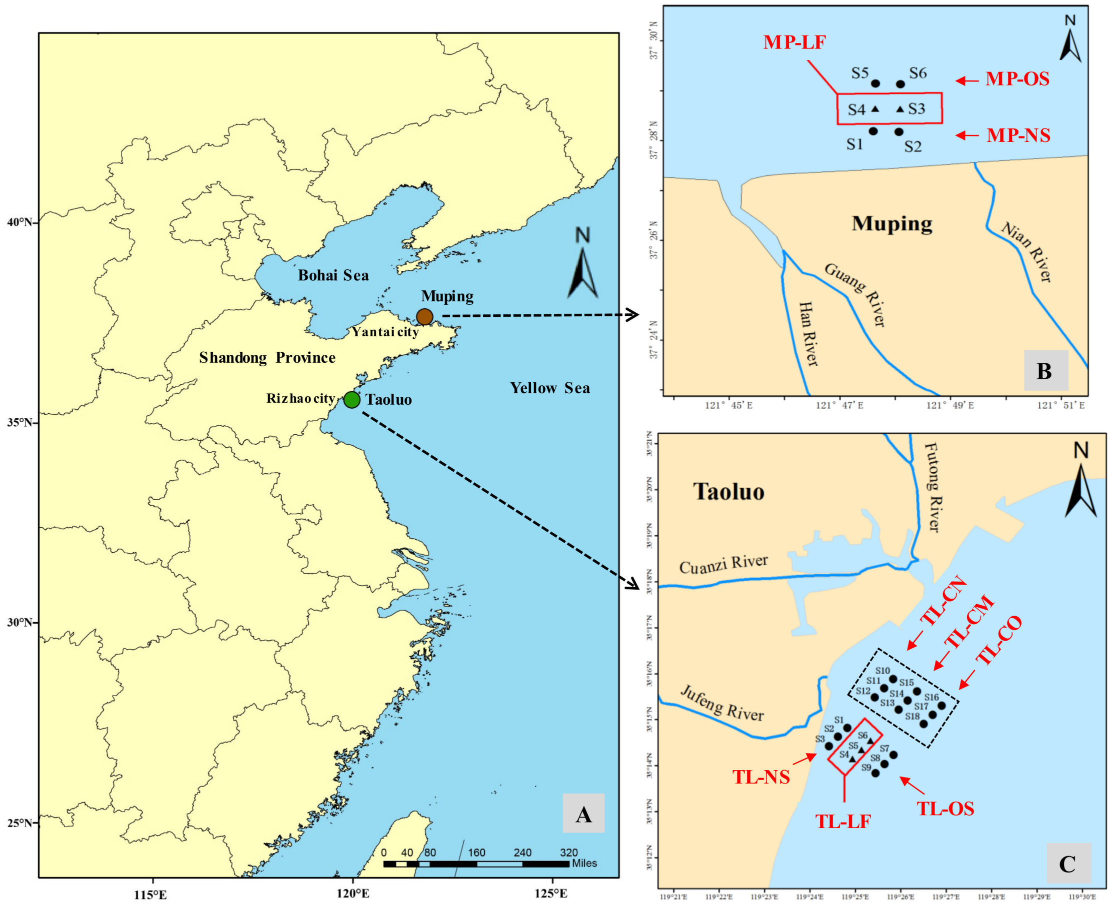

Figure 1. Map of the study areas. (**A**) Locations of the two study areas in Shandong Province, China. The brown and green dots indicate the sampling stations of Muping and Taoluo, respectively; (**B**) The sampling points in Muping, with two river estuaries (Han River and Guang River) in the west and Nian River estuary in the east. The red rectangle indicates a small-scale laver culture area (MP-LF, sampling points of S3–S4). The nearshore and offshore areas 100 m away from MP-LF were represented as MP-NS (sampling points of S1–S2) and MP-OS (sampling points of S5–S6), respectively. (**C**) The sampling points in Taoluo, with Jufeng River estuary in the west and Futong River estuary in the north. The red rectangle and black rectangle indicate a large-scale laver culture area (TL-LF, sampling points of S4–S6) and a reference site with no culture activities, respectively. The nearshore and offshore areas approximately 1 km away from TL-LF were represented as TL-NS (sampling points of S1–S3) and TL-OS (sampling points of S7–S9), respectively. In the reference site, three sampling sites were set parallel to TL-NS, TL-LF, and TL-OS, represented by TL-CN (sampling points of S10–S12), TL-CM (sampling points of S13–S15) and TL-CO (sampling points of S16–S18), respectively.

2.3. Water Quality Characteristics Determination

The hydrographic parameters (temperature, salinity, pH, total suspended solids (TSS), and water transparency (Tra)), hydrochemical parameters (ammonium nitrogen (NH_4^+-N), nitrate nitrogen (NO_3^--N), nitrite nitrogen (NO_2^--N), dissolved inorganic nitrogen (DIN, defined as sum of ammonia, nitrite and nitrate nitrogen), phosphate phosphorus (PO_4^{3-}-P), total nitrogen (TN), dissolved organic carbon (DOC), particulate organic carbon (POC), and dissolved oxygen (DO)), and chlorophyll-a (Chl-a) were measured following standard procedures in September (Sep), October (Oct), and December (Dec) from the culture, control, and reference sites.

Temperature, salinity, pH, and DO were measured in situ using the YSI-multiparameter instrument (600 QS-M-O, USA) at a depth of 0.2 m. The seawater transparency was measured using a Secchi disc. The water samples were collected at a depth of 0.2 m at each sampling point by using an organic glass hydrophore. On board the ship, the sampled water was gently transferred into carboys and stored a cool and dark environment until filtration within 2 h. All the glassware used in this study was cleaned, acidified in 10% HCl, rinsed with deionized water, and combusted for 2 h at 500 °C in a muffle furnace before use to remove inorganic and organic carbon. For Chl-a measurements, 500 mL of sample were treated with 1% $MgCO_3$ powder suspension to prevent acidification and breakdown of chlorophylls, then filtered onto glass fiber filters (GF/F, pore size 0.45 μm, Whatman) and frozen in dry ice (frozen carbon dioxide). For determinations of DOC and TN, 100 mL of the samples were placed into glass bottles and were preserved with mercuric chloride ($HgCl_2$, 0.5 μM) to stop microbial activity. Moreover, 50 mL of sample were filtered through Whatman GF/F filters (pore size 0.45 μm) and frozen immediately for later, shore-based phosphate, ammonium, nitrate, and nitrite analyses. For POC determination, 200 mL of sample were filtered onto pre-combusted (450 °C, 5 h) Whatman GF/F filters (pore size 0.7 μm) and then frozen in dry ice.

The samples were transported to the laboratory and all the experiments were conducted within 24 h. The Chl-a concentration was determined according to the ISO standard [25] using a UV-Vis spectrophotometer (Metash UV-9000, Shanghai, China). TSS was determined by pouring 1 L of water through a pre-weighed filter of 0.45 μm pore size, then weighing the filter again after drying it at 105 °C for 2 h to remove all water. DOC and TN were measured using a plus nitrogen analyzer (TOC-VCSH, Shimadzu, Kyoto, Japan) according to Evangeliou and Florou [26] and the European standard [27], respectively. The POC analyses were conducted according to Ray et al. [28]. The concentrations of NH_4^+-N, NO_3^--N, NO_2^--N, and PO_4^{3-}-P were determined using standard colorimetric techniques on an auto-analyzer (Auto Analyzer AA-3, Bran-Luebbe, Norderstadt, Germany).

2.4. Statistical Analysis

To compare the significance of variations between groups, a one-way analysis of variance (ANOVA) followed by the post-hoc Tukey honestly significant difference (HSD) test was performed (ver. 22). In order to reveal the potential relationships between the environmental parameters and the distribution of the sampling stations, reduce the number of explanatory variables, and avoid collinearity effects in further analysis, a principal component analysis (PCA) was applied to the entire set of normalized environmental data. In order to look for groups of sampling stations with similar characteristics, we also used Ward's hierarchical clustering method by SPSS software as a complementary analysis to enrich the output of PCA. The Pearson correlation analysis was performed to assess the correlation between different environmental parameters. The difference was considered significant when $p < 0.05$.

3. Results

3.1. Spatio-Temporal Variations in Water Quality Parameters

The spatio-temporal variations in hydrographic parameter values and the hydrochemical parameter concentrations are shown in Tables 1 and 2. Water temperature varied from maxima of 24–25 °C in Sep to minima of 8–10 °C in Dec and showed a significant decrease with time. Temperature variability between sites in the same sampling station was small (Table 1). Seasonal variation in DO concentration was substantial. All minima in mean DO were recorded in Sep (ranging from 8.86 to 9.80 mg L^{-1}), as higher temperature depressed oxygen solubility (Table 2). Salinity varied geographically and monthly in both Taoluo (TL) and Muping (MP) zones, with an increasing trend from Sep to Dec and from nearshore to offshore. The pH value in TL-LF was significantly higher than that at TL-NS and TL-OS in Dec ($p < 0.05$). TSS was significantly lower in Muping than that in Taoluo in Sep ($p < 0.05$). Correspondingly, Tra was significantly higher in MP than that in TL in Sep ($p < 0.05$). TSS was significantly higher in TL-LF and TL-OS than in TL-NS ($p < 0.05$), and significantly higher in TL-CO than in TL-CS and TL-CM in Oct ($p < 0.05$) (Table 1).

Table 1. The values of hydrographic parameters for the water samples collected from nine sites in or around two laver farms in Shandong Province, China in September (Sep), October (Oct), and December (Dec) 2019. The abbreviations of the sites are the same in Figure 1. The different superscript letters in each column shows that the values are significantly different ($p < 0.05$). MP: Muping zone; TL: Taoluo zone; Temp: temperature; Sal: salinity; TSS: total suspended solids; Tra: transparency.

Month	Zone	Site		Temp (°C)	Sal	pH	TSS (mg L^{-1})	Tra (m)
Sep	TL	Control	TL-NS	24.95 ^{ab}	27.87 ^{h}	7.89 ^{fg}	47.30 ^{a}	0.65 ^{k}
			TL-OS	24.45 ^{c}	27.86 ^{h}	7.88 ^{fg}	46.30 ^{a}	0.65 ^{k}
		Culture	TL-LF	25.10 ^{a}	27.88 ^{h}	7.89 ^{efg}	53.40 ^{a}	0.70 ^{k}
		Reference	TL-CS	24.70 ^{b}	27.86 ^{h}	7.87 ^{fgh}	39.60 ^{ab}	1.23 ^{g}
			TL-CM	24.75 ^{b}	27.90 ^{h}	7.88 ^{fg}	48.40 ^{a}	1.25 ^{g}
			TL-CO	24.77 ^{b}	27.88 ^{h}	7.87 ^{fgh}	56.93 ^{a}	0.73 ^{jk}
	MP	Control	MP-NS	23.90 ^{d}	29.49 ^{e}	8.03 ^{b}	3.16 ^{c}	3.45 ^{ab}
			MP-OS	24.05 ^{d}	29.47 ^{e}	8.02 ^{b}	5.66 ^{c}	3.55 ^{a}
		Culture	MP-LF	24.00 ^{d}	29.47 ^{e}	8.01 ^{b}	2.68 ^{c}	3.35 ^{b}
Oct	TL	Control	TL-NS	18.23 ^{e}	28.91 ^{f}	7.99 ^{bc}	14.67 ^{c}	0.72 ^{jk}
			TL-OS	18.30 ^{e}	28.91 ^{f}	8.01 ^{b}	47.67 ^{a}	0.68 ^{k}
		Culture	TL-LF	18.20 ^{e}	28.92 ^{f}	7.96 ^{bcd}	53.33 ^{a}	0.70 ^{i}
		Reference	TL-CS	17.90 ^{f}	28.76 ^{g}	7.96 ^{bcde}	15.00 ^{c}	0.65 ^{k}
			TL-CM	18.20 ^{e}	28.91 ^{f}	8.00 ^{b}	21.00 ^{bc}	0.75 ^{jk}
			TL-CO	18.23 ^{e}	28.92 ^{f}	8.00 ^{b}	60.15 ^{a}	0.73 ^{jk}
	MP	Control	MP-NS	17.40 ^{g}	29.81 ^{d}	7.69 ^{k}	10.93 ^{c}	2.75 ^{c}
			MP-OS	17.60 ^{g}	29.78 ^{d}	7.84 ^{ghi}	7.30 ^{c}	2.20 ^{d}
		Culture	MP-LF	17.55 ^{g}	29.79 ^{d}	7.79 ^{ij}	9.90 ^{c}	2.65 ^{c}
Dec	TL	Control	TL-NS	8.63 ^{k}	30.05 ^{b}	7.90 ^{defg}	46.47 ^{a}	0.90 ^{ij}
			TL-OS	10.17 ^{i}	30.14 ^{b}	8.03 ^{b}	39.80 ^{ab}	0.85 ^{ijk}
		Culture	TL-LF	9.17 ^{j}	30.05 ^{b}	8.15 ^{a}	49.93 ^{a}	0.98 ^{hi}
		Reference	TL-CS	9.10 ^{j}	29.94 ^{c}	7.90 ^{defg}	42.07 ^{a}	1.15 ^{gh}
			TL-CM	10.03 ^{i}	30.08 ^{b}	7.93 ^{cdef}	42.53 ^{a}	1.10 ^{gh}
			TL-CO	10.73 ^{h}	30.09 ^{b}	8.03 ^{b}	41.67 ^{a}	0.97 ^{hi}
	MP	Control	MP-NS	7.85 ^{m}	30.83 ^{a}	7.80 ^{hij}	39.70 ^{ab}	1.90 ^{e}
			MP-OS	8.05 ^{m}	30.86 ^{a}	7.75 ^{jk}	42.70 ^{a}	1.50 ^{f}
		Culture	MP-LF	8.05 ^{m}	30.85 ^{a}	7.79 ^{ij}	44.80 ^{a}	1.45 ^{f}

As Table 2 shows, the concentrations of NO_3^--N, DIN, and PO_4^{3-}-P in Sep, as well as NO_3^--N and DIN in Oct were both significantly lower in MP than that in TL ($p < 0.05$). Overall, DIN concentration decreased in TL and increased in MP with time (Table 2). The concentrations of NH_4^+-N, NO_3^--N, and DIN were the highest in TL-NS over the study period. In Dec, the concentrations of NH_4^+-N, NO_2^--N, DIN, and PO_4^{3-}-P were all significantly higher in TL-NS than that in TL-LF and TL-OS ($p < 0.05$), and TL-LF had the lowest concentrations of NO_3^--N, NO_2^--N, DIN, and PO_4^{3-}-P (Table 2). As a whole, TN concentration was the highest in Dec, and Chl-a concentration was the highest in Oct in both TL and MP. The DOC concentration was significantly lower in TL-NS than in TL-LF and TL-OS in Sep ($p < 0.05$). The concentration of POC in TL decreased while DOC increased from Oct to Dec. Both the concentrations of DOC and POC decreased from Sep to Dec in MP.

Table 2. The concentrations of hydrochemical parameters for the water samples collected from nine sites in or around two laver farms in Shandong Province, China in September, October, and December 2019. The abbreviations of the sites are the same in Figure 1. The different superscript letters in each column shows that the values are significantly different ($p < 0.05$). NH_4^+-N: ammonium nitrogen; NO_3^--N: nitrate nitrogen; NO_2^--N: nitrite nitrogen; DIN: dissolved inorganic nitrogen (sum of ammonia, nitrite and nitrate nitrogen); PO_4^{3-}-P: phosphate phosphorus; TN: total nitrogen; DOC: dissolved organic carbon; POC: particulate organic carbon; DO: dissolved oxygen; Chl-a: chlorophyll-a.

Mon	Site	NH_4^+-N ($\mu g\ L^{-1}$)	NO_3^--N ($\mu g\ L^{-1}$)	NO_2^--N ($\mu g\ L^{-1}$)	PO_4^{3-}-P ($\mu g\ L^{-1}$)	DIN ($\mu g\ L^{-1}$)	TN ($mg\ L^{-1}$)	DOC ($mg\ L^{-1}$)	POC ($mg\ L^{-1}$)	DO ($mg\ L^{-1}$)	Chl-a ($\mu g\ L^{-1}$)
Sep	TL-NS	160.08 ^{a}	738.28 ^{a}	19.06 ^{de}	28.42 ^{ab}	917.43 ^{a}	0.66 ^{defg}	3.06 ^{ef}	8.50 ^{bcd}	9.19 ^{hi}	1.63 ^{ef}
	TL-OS	106.67 ^{bc}	377.63 ^{c}	11.74 ^{ef}	29.91 ^{ab}	496.03 ^{bc}	0.70 ^{cde}	4.03 ^{a}	7.25 ^{cde}	8.96 ^{i}	1.00 ^{f}
	TL-LF	89.40 ^{cde}	307.67 ^{cde}	12.58 ^{ef}	38.03 ^{a}	409.64 ^{cde}	0.68 ^{def}	3.69 ^{abc}	9.40 ^{ab}	9.11 ^{i}	1.81 ^{ef}
	TL-CS	81.70 ^{cdef}	355.72 ^{cd}	28.85 ^{bc}	34.30 ^{ab}	466.26 ^{bcd}	0.85 ^{abc}	3.13 ^{def}	8.90 ^{abc}	9.65 ^{gh}	1.73 ^{ef}
	TL-CM	65.95 ^{cdef}	322.73 ^{cde}	17.48 ^{ef}	28.10 ^{ab}	406.16 ^{def}	0.77 ^{bcd}	3.00 ^{ef}	9.60 ^{ab}	9.18 ^{hi}	2.56 ^{def}
	TL-CO	67.23 ^{cdef}	540.79 ^{b}	8.54 ^{fg}	24.73 ^{abc}	616.55 ^{b}	0.61 ^{efgh}	3.00 ^{ef}	9.37 ^{abc}	8.86 ^{i}	1.81 ^{ef}
	MP-NS	35.30 ^{ef}	18.66 ^{k}	3.71 ^{g}	18.69 ^{def}	57.66 ^{k}	0.51 ^{fgh}	3.15 ^{def}	7.90 ^{bcd}	9.73 ^{g}	1.36 ^{f}
	MP-OS	31.54 ^{f}	17.62 ^{k}	3.71 ^{g}	16.34 ^{efg}	52.88 ^{k}	0.58 ^{fgh}	3.41 ^{bcd}	9.25 ^{abc}	9.80 ^{g}	1.51 ^{ef}
	MP-LF	30.18 ^{f}	18.09 ^{k}	4.96 ^{g}	18.30 ^{def}	53.23 ^{k}	0.49 ^{gh}	3.16 ^{def}	6.55 ^{def}	9.74 ^{g}	1.14 ^{f}
Oct	TL-NS	71.18 ^{cdef}	333.17 ^{cde}	28.86 ^{bc}	14.61 ^{efg}	433.21 ^{bcd}	0.63 ^{efgh}	3.08 ^{ef}	7.43 ^{cde}	12.53 ^{e}	7.56 ^{c}
	TL-OS	71.73 ^{cdef}	329.98 ^{cde}	28.06 ^{cd}	16.98 ^{def}	429.77 ^{bcd}	0.66 ^{defg}	3.08 ^{ef}	7.63 ^{bcd}	11.94 ^{f}	4.11 ^{cd}
	TL-LF	43.48 ^{def}	201.44 ^{fg}	16.35 ^{ef}	12.82 ^{fg}	261.28 ^{fgh}	0.74 ^{bcd}	3.15 ^{def}	6.77 ^{def}	13.49 ^{d}	16.45 ^{ab}
	TL-CS	54.76 ^{cdef}	235.83 ^{fg}	27.36 ^{cd}	20.46 ^{cde}	317.94 ^{efg}	0.68 ^{def}	3.27 ^{cde}	8.90 ^{abc}	12.04 ^{ef}	13.99 ^{b}
	TL-CM	97.89 ^{cd}	328.53 ^{cde}	40.52 ^{ab}	26.60 ^{abc}	466.94 ^{bcd}	0.77 ^{bcd}	3.21 ^{cde}	8.43 ^{bcd}	12.08 ^{ef}	19.07 ^{a}
	TL-CO	67.61 ^{cdef}	307.24 ^{cde}	23.02 ^{cd}	15.94 ^{efg}	397.87 ^{def}	0.48 ^{h}	2.91 ^{f}	7.70 ^{bcd}	12.11 ^{ef}	4.49 ^{cd}
	MP-NS	45.52 ^{def}	46.10 ^{j}	21.76 ^{de}	18.06 ^{def}	113.37 ^{ik}	0.59 ^{fgh}	3.07 ^{ef}	5.90 ^{def}	11.87 ^{f}	6.44 ^{cd}
	MP-OS	51.75 ^{cdef}	72.22 ^{ij}	15.69 ^{ef}	19.97 ^{def}	139.66 ^{ik}	0.62 ^{efgh}	2.97 ^{f}	7.15 ^{cde}	12.39 ^{ef}	5.31 ^{cd}
	MP-LF	59.12 ^{cdef}	72.64 ^{ij}	16.83 ^{ef}	19.62 ^{def}	148.59 ^{ghi}	0.70 ^{cde}	3.17 ^{def}	9.70 ^{a}	11.96 ^{f}	1.26 ^{f}
Dec	TL-NS	149.17 ^{ab}	267.91 ^{ef}	38.39 ^{ab}	36.40 ^{ab}	455.48 ^{bcd}	0.85 ^{abc}	3.75 ^{abc}	5.97 ^{def}	15.40 ^{a}	2.57 ^{def}
	TL-OS	60.98 ^{cdef}	237.51 ^{fg}	18.12 ^{de}	20.87 ^{cde}	316.61 ^{efg}	0.72 ^{cde}	3.58 ^{abc}	5.57 ^{def}	14.28 ^{bc}	2.54 ^{def}
	TL-LF	84.64 ^{cdef}	157.76 ^{gh}	17.05 ^{ef}	12.26 ^{g}	259.46 ^{fgh}	0.96 ^{a}	3.77 ^{ab}	5.97 ^{def}	14.69 ^{b}	1.32 ^{f}
	TL-CS	93.26 ^{cd}	241.77 ^{fg}	19.41 ^{de}	22.69 ^{bcd}	354.44 ^{def}	0.69 ^{def}	3.44 ^{bcd}	5.20 ^{ef}	14.31 ^{bc}	2.65 ^{def}
	TL-CM	54.60 ^{cdef}	260.79 ^{ef}	18.43 ^{de}	21.61 ^{cde}	333.82 ^{efg}	0.70 ^{cde}	3.34 ^{cde}	5.57 ^{def}	14.36 ^{bc}	3.28 ^{de}
	TL-CO	48.57 ^{def}	176.26 ^{gh}	17.04 ^{ef}	17.67 ^{def}	241.86 ^{fgh}	0.79 ^{abc}	3.49 ^{abc}	6.13 ^{def}	13.89 ^{cd}	3.20 ^{de}
	MP-NS	27.98 ^{f}	106.96 ^{hi}	38.91 ^{ab}	23.71 ^{bcd}	173.85 ^{ghi}	0.70 ^{cde}	2.88 ^{f}	4.75 ^{f}	14.11 ^{c}	0.91 ^{f}
	MP-OS	28.23 ^{f}	128.49 ^{hi}	41.35 ^{ab}	26.04 ^{abc}	198.07 ^{ghi}	0.88 ^{ab}	2.90 ^{f}	4.60 ^{f}	14.15 ^{c}	0.88 ^{f}
	MP-LF	50.10 ^{def}	123.33 ^{hi}	47.74 ^{a}	29.91 ^{ab}	221.16 ^{ghi}	0.93 ^{ab}	2.90 ^{f}	6.55 ^{def}	14.09 ^{c}	0.80 ^{f}

3.2. Spatio-Temporal Variations between Culture and Control Sites

An ANOVA of the environmental data revealed that some of the water quality parameters exhibited significant variations ($p < 0.05$) between the culture and control sites on a spatial and temporal scale, and these significant variations were mostly observed in Dec compared with Sep and Oct (Table 3). The NO_3^--N concentration and N:P in TL as well as TSS concentration in MP significantly varied ($p < 0.05$) between the culture and control sites in Sep. The concentrations of NO_3^--N, NO_2^--N, and DIN in TL, and pH value in TL, as well as POC concentration in MP exhibited significant variation ($p < 0.05$) between the culture and control sites in Oct. Significant variations ($p < 0.05$) of the values of temperature, salinity, pH, and N:P, as well as the concentrations of TSS, NH_4^+-N, NO_3^--N, NO_2^--N, PO_4^{3-}-P, DIN, DO, and Chl-a between the culture and control sites in Dec were observed.

3.3. Spatio-Temporal Variations in Culture and Control Sites in Comparison to the Reference Sites

In order to further ascertain whether the spatial variation (between the culture and control sites) in nutrient parameters can be exclusively considered as an aftermath of laver cultivation, the water quality parameters under investigation were compared with the values obtained at the reference sites with no culture activities yet with a similar hydrographic setting (situated 2 km away from the culture sites). Significant variations

($p < 0.05$) between culture and reference sites and between control and reference sites were also mostly observed in Dec compared with Sep and Oct (Table 4). The Tra value significantly varied ($p < 0.05$) between the control and reference sites in TL in Sep. The concentrations of DIN and DO exhibited significant variation ($p < 0.05$) between the culture and reference sites in Oct. Significant variations ($p < 0.05$) of pH and N:P values as well as the concentrations of TN and Chl-a between the culture and reference sites in Dec were observed. Moreover, the concentrations of Chl-a and DO as well as the Tra value exhibited a significant variation ($p < 0.05$) between the control and reference sites in Dec. There was no significant difference in nutrient parameters between the control site and the reference site ($p > 0.05$).

Table 3. Significant variations between culture and control sites on a spatial and temporal scale. Numbers in bold indicate significant differences. MP: Muping zone; TL: Taoluo zone; Temp: temperature; Sal: salinity; TSS: total suspended solids; Tra: transparency; NH_4^+-N: ammonium nitrogen; NO_3^--N: nitrate nitrogen; NO_2^--N: nitrite nitrogen; DIN: dissolved inorganic nitrogen; PO_4^{3-}-P: phosphate phosphorus; TN: total nitrogen; DOC: dissolved organic carbon; POC: particulate organic carbon; DO: dissolved oxygen; Chl-a: chlorophyll-a.

Parameters	September		October		December	
	MP	TL	MP	TL	MP	TL
Temp	0.074	0.349	0.221	0.252	0.103	**0.001**
Sal	0.081	0.380	0.207	0.228	0.704	**0.003**
pH	0.596	0.426	0.054	**0.009**	0.154	**0.001**
TSS	**0.023**	0.363	0.076	0.134	0.737	**0.046**
Tra	0.441	0.945	0.095	0.644	0.062	0.189
NH_4^+-N	0.291	0.486	0.472	0.208	0.104	**0.019**
NO_3^--N	0.974	**0.020**	0.228	**0.040**	0.712	**0.047**
NO_2^--N	0.385	0.425	0.751	**0.003**	0.297	**0.030**
PO_4^{3-}-P	0.759	0.477	0.730	0.406	0.182	**0.023**
DIN	0.792	0.060	0.502	**0.024**	0.529	**0.030**
N:P	0.347	**0.033**	0.588	0.223	0.953	**0.049**
TN	0.170	0.918	0.749	0.187	0.138	0.064
DOC	0.609	0.420	0.592	0.919	0.977	0.799
POC	0.533	0.664	**0.030**	0.653	0.407	0.880
DO	0.675	0.367	0.533	**0.008**	0.928	**0.002**
Chl-a	0.894	0.603	0.090	**0.014**	0.979	**0.007**

Table 4. Significant variations between culture and reference sites as well as control and reference sites on a monthly scale in Taoluo zone. Numbers in bold indicate significant differences. The abbreviations are the same in Table 3.

Parameters	September		October		December	
	Culture Site	Control Site	Culture Site	Control Site	Culture Site	Control Site
Temp	0.066	0.939	0.643	0.143	0.270	0.347
Sal	0.591	0.903	0.615	0.645	0.974	0.229
pH	0.228	0.365	0.512	0.632	**0.003**	0.927
TSS	0.692	0.947	0.359	0.998	0.094	0.924
Tra	0.163	**0.034**	0.952	0.924	0.339	**0.003**
NH_4^+-N	0.858	0.076	0.986	0.158	0.628	0.093
NO_3^--N	0.514	0.780	0.127	0.353	0.147	0.802
NO_2^--N	0.668	0.834	0.176	0.947	0.902	0.203
PO_4^{3-}-P	0.163	1.000	0.353	0.501	0.199	0.314
DIN	0.846	0.346	**0.043**	0.634	0.584	0.276
N:P	0.611	0.426	0.994	0.165	**0.003**	0.981
TN	0.792	0.654	0.558	1.000	**0.048**	0.668
DOC	0.122	0.129	0.991	0.916	0.248	0.338
POC	0.990	0.285	0.683	0.419	0.888	0.970
DO	0.870	0.677	**0.000**	0.696	0.207	**0.025**
Chl-a	0.952	0.309	0.936	0.173	**0.000**	**0.020**

3.4. Multivariate Analysis (Principal Component Analysis and Hierarchical Clustering Analysis)

A principal component analysis was conducted to reveal the relationships between the environmental parameters and the distribution of the sampling stations in a biplot (Figure 2). In September (Figure 2A), the first and second principal components (PC1 and PC2) explained 47.5% of the total variation in environmental data, and showed clear partitioning of TL-CS from TL-CO and TL-NS samples along the PC1, and partitioning of TL-CS and TL-NS from TL-CO, TL-OS, TL-LF, and TL-CM samples along PC2. PC1 strongly correlated to variables of N:P, nitrate, TN, DIN, and Tra. PC2 strongly correlated to variables of TSS, nitrite, and DO.

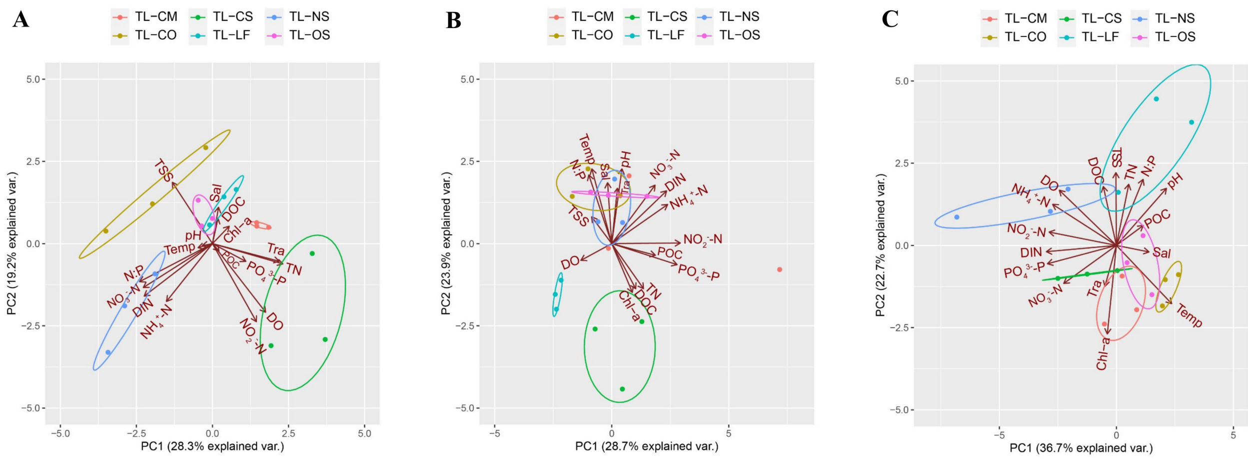

Figure 2. Biplot of the principal component analysis (PCA) for environmental parameters in various sampling stations in September (**A**), October (**B**) and December (**C**). Red arrows represent environmental variables, the directions of arrows represent correlation between variable and principle components, lengths represent devotion of environmental parameters data to principle components, and circles in color represents sampling points. Ellipses represent 68% confidence intervals of core regions. The abbreviations of the sites are the same in Figure 1. The abbreviations of the environmental parameters are the same in Table 3.

In October (Figure 2B), PC1 and PC2 explained 52.6% of the total variation in the environmental data, and showed clear partitioning of TL-CS and TL-LF from TL-NS, TL-CO, and TL-OS samples along the PC2. PC1 strongly correlated to variables of phosphorus, nitrite, ammonium, and DIN. PC2 strongly correlated to variables of N:P, temperature, and pH.

The variability of the environmental parameters was explained reasonably well (59.4% of variance) by PC1 and PC2 in December (Figure 2C). Clear partitioning of TL-CO and TL-OS from TL-NS samples along the PC1 was observed, and partitioning of TL-LF and TL-NS from TL-CO, TL-CS, and TL-CM samples along the PC2 was found. PC1 strongly correlated to variables of DO, ammonium, nitrate, nitrite, DIN, and phosphorus. PC2 strongly correlated to variables of Chl-a, N:P, and TSS.

A cluster analysis was conducted for various sampling sites based on the water quality characteristics in Sep, Oct, and Dec, respectively (Figure 3). The three clusters were formed based on the water quality characteristics in Sep and Oct. TL-NS was a cluster by itself in Sep. TL-CS and TL-CM were merged into a cluster in Oct. The sampling sites in MP were clustered into a cluster in Sep, Oct, and Dec. Four clusters were formed in Dec, with TL-LF and TL-NS clustered separately.

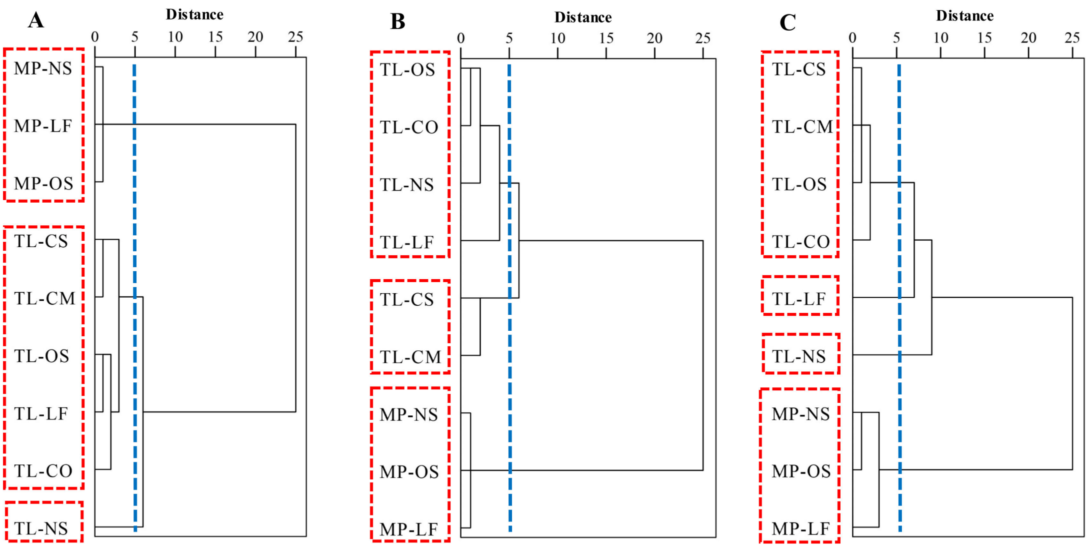

Figure 3. Hierarchical clustering using Ward's method based on the water quality characteristics in various sampling sites in September (**A**), October (**B**), and December (**C**). The horizontal vertical axis shows the distance between sampling sites based on Euclidean distance. The abbreviations of the sites are the same in Figure 1.

3.5. Correlation Analysis between the Water Quality Parameters

Great differences between the TL and MP sampling stations were observed using the Pearson correlation analysis (Tables 5 and 6). A significant correlation between DO and temperature was present in TL (Table 5) but was absent in MP (Table 6). TN seemed to have no strong correlation with the other environmental factors including DIN in TL, while TN was significantly correlative to the other factors such as DIN and PO_4^{3-}-P in MP. DIN and PO_4^{3-}-P had a significant negative correlation with temperature and a positive correlation with salinity in MP (Table 6). In TL, DIN had a weak correlation with temperature and salinity (Table 5). The Pearson correlation coefficients were below 0.5 between DOC and the other environmental factors in TL while DOC had a significant correlation with temperature, salinity, and transparency in MP. Significant correlation was present between POC and temperature, as well as DO and salinity in TL, while POC was weakly correlative to these factors in MP.

Table 5. Pearson correlation coefficient between the water quality parameters based on the data from the stations at Taoluo. The bold numbers indicate a statistically significant correlation ($p < 0.05$). The abbreviations are the same in Table 3.

Taoluo	TN	DIN	NO_3^--N	NH_4^+-N	NO_2^--N	PO_4^{3-}-P	DOC	POC	TSS	DO	Chl-a	Temp	Sal	pH	Tra
TN	1.00														
DIN	−0.24	1.00													
NO_3^--N	−0.34	**0.98**	1.00												
NH_4^+-N	0.20	**0.70**	**0.54**	1.00											
NO_2^--N	0.18	−0.03	−0.16	0.26	1.00										
PO_4^{3-}-P	0.21	**0.42**	**0.34**	**0.58**	0.13	1.00									
DOC	**0.49**	−0.27	−0.36	0.29	−0.17	0.29	1.00								
POC	−0.24	**0.45**	**0.51**	0.02	−0.01	**0.39**	**−0.47**	1.00							
TSS	0.08	0.19	0.22	0.14	**−0.58**	0.21	0.16	0.00	1.00						
DO	0.30	**−0.61**	**−0.68**	−0.15	0.32	−0.47	0.27	**−0.86**	−0.20	1.00					
Chl-a	−0.07	−0.19	−0.20	−0.24	**0.56**	−0.26	−0.29	0.17	**−0.62**	0.16	1.00				
Temp	−0.33	**0.58**	**0.65**	0.10	−0.22	0.40	**−0.37**	**0.89**	0.14	**−0.97**	−0.01	1.00			
Sal	0.30	**−0.60**	**−0.66**	−0.16	0.20	**−0.44**	**0.33**	**−0.89**	−0.15	**0.97**	0.03	**−0.99**	1.00		
pH	0.24	**−0.51**	**−0.52**	−0.30	0.10	**−0.72**	0.13	−0.45	−0.22	**0.61**	0.22	**−0.57**	**0.61**	1.00	
Tra	**0.49**	−0.31	−0.32	−0.13	0.01	0.15	0.00	−0.23	0.10	0.20	−0.35	−0.29	0.26	−0.12	1.00

Table 6. Pearson correlation coefficient between the water quality parameters based on the data from the stations at Muping. The bold numbers indicate a statistically significant correlation ($p < 0.05$). The abbreviations are the same in Table 3.

Muping	TN	DIN	NO_3^--N	NH_4^+-N	NO_2^--N	PO_4^{3-}-P	DOC	POC	TSS	DO	Chl-a	Temp	Sal	pH	Tra
TN	1.00														
DIN	**0.93**	1.00													
NO_3^--N	**0.93**	**0.98**	1.00												
NH_4^+-N	0.19	0.29	0.13	1.00											
NO_2^--N	**0.90**	**0.95**	**0.95**	0.04	1.00										
PO_4^{3-}-P	**0.91**	**0.89**	**0.90**	0.05	**0.92**	1.00									
DOC	**−0.62**	**−0.82**	**−0.83**	−0.05	**−0.83**	**−0.79**	1.00								
POC	−0.35	**−0.48**	**−0.55**	**0.47**	**−0.66**	**−0.54**	**0.79**	1.00							
TSS	**0.89**	**0.88**	**0.92**	−0.16	**0.97**	**0.93**	**−0.76**	**−0.65**	1.00						
DO	0.09	0.31	0.20	**0.78**	0.10	−0.10	−0.22	0.10	−0.13	1.00					
Chl-a	−0.31	−0.13	−0.21	0.39	−0.18	−0.38	−0.04	−0.06	−0.38	**0.71**	1.00				
Temp	**−0.88**	**−0.96**	**−0.97**	−0.02	**−0.99**	**−0.88**	**0.85**	**0.66**	**−0.96**	−0.15	0.18	1.00			
Sal	**0.87**	**0.91**	**0.94**	−0.11	**0.98**	**0.91**	**−0.82**	**−0.70**	**0.99**	−0.02	−0.28	**−0.98**	1.00		
pH	**−0.72**	**−0.79**	**−0.78**	−0.17	**−0.76**	**−0.56**	**0.66**	**0.57**	**−0.66**	−0.49	−0.20	**0.78**	**−0.72**	1.00	
Tra	**−0.89**	**−0.98**	**−0.98**	−0.16	**−0.95**	**−0.90**	**0.89**	**0.61**	**−0.89**	−0.25	0.08	**0.96**	**−0.93**	**0.79**	1.00

4. Discussion

Although the biofiltering capacity of seaweeds have been testified and the application for bioremediation and integrative aquaculture has attracted much attention [5–9,19–23], there are very few reports regarding the investigation of the effect of seaweeds cultivation on the environmental factors in open sea systems, where changes are difficult to be monitored and controlled. He et al. [24] reported the capability and efficiency of large-scale *N. yezoensis* cultivation in the removal of dissolved inorganic nitrogen and phosphorus from the open sea area at Lusi coast, where the largest laver farm is located on the northern bank of the mouth of the Yangtze River in China.

4.1. Hydrographic Parameters

Surface seawater temperature is affected by meteorological and hydrological conditions such as rainfall, evaporation, air temperature, wind speed, intensity of solar radiation, and current [29]. A typical seasonal and spatial pattern in the change in surface water temperature was observed at all sampling stations in this study (Table 1). At the end of Sep, the water temperature was higher at nearshore than at offshore. As air temperature decreased, the surface water temperature dropped more at nearshore than at offshore. The salinity level varies geographically and seasonally and might increase through evaporation and sea ice freezing and decrease through precipitation and freshwater input from rivers [30]. The salinity showed a significant increase over the study period in both Taoluo and Muping zones, and was generally lower at the nearshore sampling points and higher at the offshore ones (Table 1), likely owing to the cumulative influence of rainfall, discharge of estuary, and temperature. There was a sharp decrease in rainfall and runoff from the rivers around the study sites in autumn and winter, which may contribute to the increase in salinity from late Sep to early Dec. The pH values generally increase from the estuary to the ocean, caused by the input from land (rivers) [29]. Consistently, the pH at the reference site without a laver farm increased with the distance from the coast increasing in Taoluo in Dec (Table 1). However, the pH was significantly higher in the laver farm than that in the control site in Dec, likely owing to larger cultivation. It has been found that seaweeds greatly improve the pH and DO of the culture system through assimilation of HCO_3^- into photosynthetic carbon fixation and photosynthetic oxygen evolution [31–33].

4.2. Hydrochemical Parameters

The discharge of estuary had a positive effect on all nutrients. In general, nutrient concentrations were highly correlated with hydrographic variables in the estuary in the study (Tables 5 and 6). The composition of DIN varies among different coastal regions.

In the Lusi *N. yezoensis* farming area, the dominant DIN was ammonium at the most study stations and in most seasons [24]. In Taoluo, nitrate was dominant and nitrite was the lowest at all sampling points and sampling times (Table 2). The concentration and form of the nutrient may affect the nutrient uptake rate [34]. Many seaweeds seem to preferentially uptake NH_4^+-N in laboratory experiments, such as *N. yezoensis*, *Gracilaria tenuistipitata*, and *U. lactuca* [35–40]. When both nitrate and ammonium N sources were available, *Porphyra dioica* preferentially removed NH_4^+-N [41]. *N. yezoensis* assimilated more NH_4^+-N (50–94%) than NO_3^--N (21–38%) in the ocean-based laver farm in Lusi where NH_4^+-N was the principal nutrient [24]. The preference to the form of DIN depends on the conditions such as N/P ratio [42]. In Taoluo, the concentrations of all the three forms of DIN were the lowest at the laver farm in Sep and Oct. Especially in Sep, the nitrate together with the DIN concentrations decreased sharply from the nearshore to the laver farm. Similarly, although the DIN concentrations were similar at the laver farm and the offshore area in Dec, they dropped sharply from the nearshore area to the nearby laver farm (Table 2). Furthermore, we found that spatial variation (between the culture and control sites) in nutrient parameters, including NH_4^+-N, NO_3^--N, NO_2^--N, PO_4-P, DIN, and N:P, was substantial in the study, especially in Dec (Table 3). In addition, there was no significant difference in these nutrient parameters (i.e., NH_4^+-N, NO_3^--N, NO_2^--N, PO_4^{3-}-P, DIN, and N:P) between the control site and the reference site (Table 4), indicating that the spatial variation in these nutrient parameters at the culture sites compared with the control sites was likely to be considered as an aftermath of laver cultivation. Moreover, significant variations in DIN concentration in Oct and N:P value in Dec were observed between the culture and reference sites (Table 4), which further ascertained the difference in DIN concentration in Oct and N:P value in Dec between culture and control sites were probably due to laver cultivation.

NUE by seaweeds are relatively low in open-water systems due to the 3-D hydrographic nature of the water flow [43,44]. This is affected by many factors, such as seaweed species, light, and the concentration and composition of nutrients [6,7,21,45]. The NUE of laver in open sea culture systems should be highly dependent on the culture areas. Based on the 300 ha culture area, *N. yezoensis* farming can result in the reduction of 21–94% of the varying dissolved inorganic nutrients [24]. At the culture scale of 40 ha in Taoluo, there were significant promotions of DO and pH by laver farming, and a significant reduction in DIN was also observed. At a smaller culture scale (0.067 ha in Muping), all the above changes by the laver cultivation were not detected.

Organic substances are important components of the marine environment as they determine the properties of seawater and the key biogeochemical processes. The coastal regions are important zones for transport of terrestrial and riverine organic matter to the open ocean [46]. Both DOC and POC play a major role in the carbon cycle, especially in shelf seas. DOC concentrations are several times greater than POC concentrations in most marine systems, except riverine and estuarine systems [47–49]. In this study, both the concentrations of POC from the surface seawater and the fluctuation of POC concentrations were higher than those of DOC at the study sites. POC and/or DOC concentrations fluctuate seasonally [50] and change vertically [51], depending on many environmental factors [49,52–55]. The organic carbon concentration usually depends on distance from the land, i.e., are more abundant in coastal and estuarine areas than in the open sea [56]. It was found that the POC concentrations exhibited a typical seasonal change in this study, suggesting that the terrestrial resource was the main resource of POC at the sampling points. TSS are composed of a wide variety of organic and mineral particles, such as silt, sand, microorganism, decaying plant and animal matter, industrial wastes, and sewage [57]. TSS was significantly higher at the laver farm in Taoluo in Sep. This may be caused by the release of reproductive spores and/or slight shedding at the tip and edge of the blade due to maturity or decaying. The Chl-a concentration was low in the laver farm in Taoluo in Dec, which was probably due to the competition of nutrients.

Significant variations ($p < 0.05$) between culture and control sites and between culture and reference sites on a spatio-temporal scale were mostly observed in Dec compared with Sep and Oct (Tables 3 and 4). Furthermore, based on the water quality characteristics in various sampling sites, more clusters were observed in Dec using a hierarchical clustering analysis, with TL-LF and TL-NS being clustered separately (Figure 3). In addition, the percentage variability explained (59.4% of variance) by PC1 and PC2 was higher in Dec compared with Sep and Oct (Figure 2). These results together support that more spatial deviation in monitored parameters between culture and non-culture sites were found in Dec.

5. Conclusions

Salinity increased from Sep to Dec and generally increased with the distance from the coast in both regions, corresponding to the reducing terrestrial inputs. POC and/or DOC decreased while TSS increased seasonally or with the distance from the coast. The concentrations of TN, DIN, and phosphorus were generally higher nearshore and decreased seasonally in Taoluo. Salinity correlated negatively to POC in both regions and DIN in Taoluo, and positively to TSS in both regions and DIN in Muping. The results indicate different origins dominating these spatial or seasonal changes, by terrestrial inputs or re-suspension of bottom sediment. The laver farms occasionally had higher POC and TSS and lower Chl-a levels. Abundant spatial variation in nutrient parameters was found. More spatial deviation in water quality parameters between culture and non-culture sites were found in Dec. The changes in water quality due to large-scale laver cultivation probably occur in northern China in winter.

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