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Original Article

Assessment of Sea Water Quality in some Limestone Island and Archipelagos Areas, Vietnam

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Abstract: With nearly 3,000 large and small islands, the islands and archipelagos of Vietnam have outstanding features in terms biodiversity and geology. The islands are mainly formed from carbonate (limestone), intrusive igneous rock, sedimentary and volcanic rocks, in which limestone islands predominate, distributed mainly in the Gulf of Tonkin. This paper presents the results of researches and assessments on sea water quality of Vietnam's typical limestone islands and archipelagoes through the 2017-2018 surveys. The research results show that although the water quality around of limestone and archipelago area of Vietnam is safety for the development of aquatic life, an increase in pollutants concentration in water has been recorded when compared to previous research results. The research results supplement the data set of sea water quality in limestone areas that defining the characteristics of marine biodiversity.

Keywords: limestone island, water quality, pollutants.

1. Introduction

Vietnam has about 2773 coastal islands with an area of 1721km^2 but they distribute very different in the coastal waters. The Northern Coast is home to the most islands (2321 islands), accounting for 83.7% of the total islands and 48.9% of the total area. The number of islands in the North Central Coast is at least, account for only 2% of the quantity and 0.83% of area. The South Central Coast and the South Coast have the same number of islands (about 7%) but in terms of area, the southern coastal islands are quite similar to the North, accounting for 40.3% of the total area of the islands, and the area of islands in the South Central Coast account for only 10% [1].

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The islands are mainly formed from carbonate (limestone), intrusive igneous rock, sedimentary and volcanic rocks, in which limestone islands is dominated, distributed mainly in the Gulf of Tonkin. According to Le Duc An, (2009) [1], among 5 island regions with 13 coastal island groups in Vietnam, there are only 3 island groups are formed by limestone and other sediments: Bai Tu Long-Ha Long Bay (including Cat Ba) (belong to the island group 2, region I) and Long Chau Island (belong to the island group 3, region I), located at the Tonkin Gulf; and Kien Hai island group (Kien Giang) (belong to the island group 12, region V). The islands located at central coast (region II and II) and at the southeastern coast (region IV) are formed by other sedimentary, volcanic rocks and granite, etc. Offshore islands such as the Spratly archipelagos are atolls that formed from coral reefs with time of millions of years.

The limestone islands are often coastal islands, therefore, the environment quality is greatly influenced by inland sources such as domestic and tourism, industrial and mining, aquaculture and livestock, etc. These are the main sources of waste generated from the socio-economic development activities of the Northeast islands (Ha Long Bay - Bai Tu Long Bay - Cat Ba - Long Chau). In the southern limestone islands (Kien Luong - Kien Giang), the main activities of tourism, aquaculture and livestock are the main waste sources of the area.

The water quality characteristics of limestone islands are the basic that defining species composition and biodiversity of ecosystems. Study of water quality of limestone island and archipelago areas of Vietnam has been conducted through a number of studies by Do Cong Thung et al (2003, 2005, 2012) [2-4], Tran Duc Thanh et al (2007, 2010) [5,6], and Cao Thi Thu Trang (2004) [7] etc., mainly concentrated in Ha Long Bay and Bai Tu Long bay. Within the framework of the national project "Study biodiversity of limestone islands and archipelagoes in Vietnamese waters; propose solutions and using patterns for conservation and sustainable development", coded KC09-11/16/20, field surveys have been carried out to assess the water quality characteristics of the limestone islands. This paper presents the updated data on characteristics of seawater quality of limestone islands and archipelagos of Vietnam through 2 surveys in 2017 and 2018.

2. Methods

Marine investigation methods

Survey times and locations

The surface water samples were collected in two main seasons: rainy season and dry season. The dry season lasts from November to next April in the Northeast island region and from December to next March in the Kien Luong area. The rainy season lasts from May to October in the Northeast island areas and from April to November in the Kien Luong area. therefore, two surveys were conducted, the first survey took place from December 2017 to January 2018 -representing for dry season, the second one took place from July to August 2018- representing for rainy season. Key study areas include: Ha Long Bay - Bai Tu Long Bay (the sites of Vu Ha, Cong Do, Dau Go, Hon Da Den, Hang Te, Cat Chuong To, Dau Be, Bo Nau, Hang Trai, Bu Xam, Hon Tay Hoi and Con Buom), Cat Ba - Long Chau area (Hang Sang, Van Boi, Gio Cung, Cat Dua, Long Chau islands) and Kien Luong area (Hon Tre, Hon Re Nho and Hon Da Lua, Hang Tuyen) - Figure 1. A total of 29 seawater samples were collected during the December 2017 survey and 43 water samples were collected during the August 2018 survey.

Sampling and sample preservation

The method of seawater sampling complied with TCVN 5998-1995 (ISO 5667-9: 1992) - Water quality -Sampling - Guidance on seawater sampling [8]. Preservation of seawater samples complied with TCVN 6663-3:2016 (ISO 5667-3:2012)-Water quality-Sampling-Part 3: Preservation and handling of water samples [9]. The parameters of water temperature, pH, DO,

salinity and turbidity were measured directly in situ by the portable meters. Other water quality parameters (BOD_5, COD, inorganic dissolved nutrients of nitrogen and phosphorus, total suspended solids, oil and grease, heavy metals (Cu, Pb, Zn, Cd, Hg, As), and organochlorinated pesticides) were collected, fixed and then stored at 4°C until analysis. For water samples used to analysis of inorganic nitrogen and phosphate, the water was added chloroform (for dissolved nutrients) and H_2SO_4 1:1 (1ml/1 liter) (for total Nitrogen and total phosphorus) and stored at 4°C until analysis.

Analytical Methods

Analysis of water quality parameters follows by the published Vietnamese Standards and the US standard documents [10]. Biochemical oxygen demand (BOD_5) was determined by the difference of dissolved oxygen concentration (DO) in sample between the first day and the fifth day (DO was determined by Winkler method); Chemical oxygen demand (COD) was determined by the Potassium Permanganate (KMnO4) method in alkaline environment; Total suspended solids was determined by filtering through filter paper with pore size 1mm, then drying and weighing; The oil and grease in the water was determined by colorimetric method after extracted with n-hexane; Nutrients: phosphate ($P\text{-}PO_4^{3-}$), nitrite ($N\text{-}NO_2^-$), ammonium (NH_3 + NH_4^+), total Nitrogen, total Phosphorus were determined by colorimetric method; Heavy metals in water (Cu, Pb, Cd, Zn, Hg, As) were determined by ICP-MS equipment (Model ELAN DRC-e), the detection limit of method was 0.01 μg/l for each element; Organochlorinated pesticides including Lindane, Aldrine, Dieldrine, Endrine, 4,4'- DDD, 4,4'-DDE, 4,4'- were determined by the gas chromatography method with Electronic Capture Detector (GC/ECD-HP 6890). External calibration was used to calculate OCPs compounds concentration with standards Lindane, Aldrine, Endrine, Dieldrine, 4, 4- DDE, 4, 4- DDD and 4, 4- DDT. The recovery of the method for individual standard is about 70.6% - 97.8%. The detection limit of the method is 0.15 ng/l for Lindane and 4, 4- DDT and 0.10 ng/l for remaining compounds.

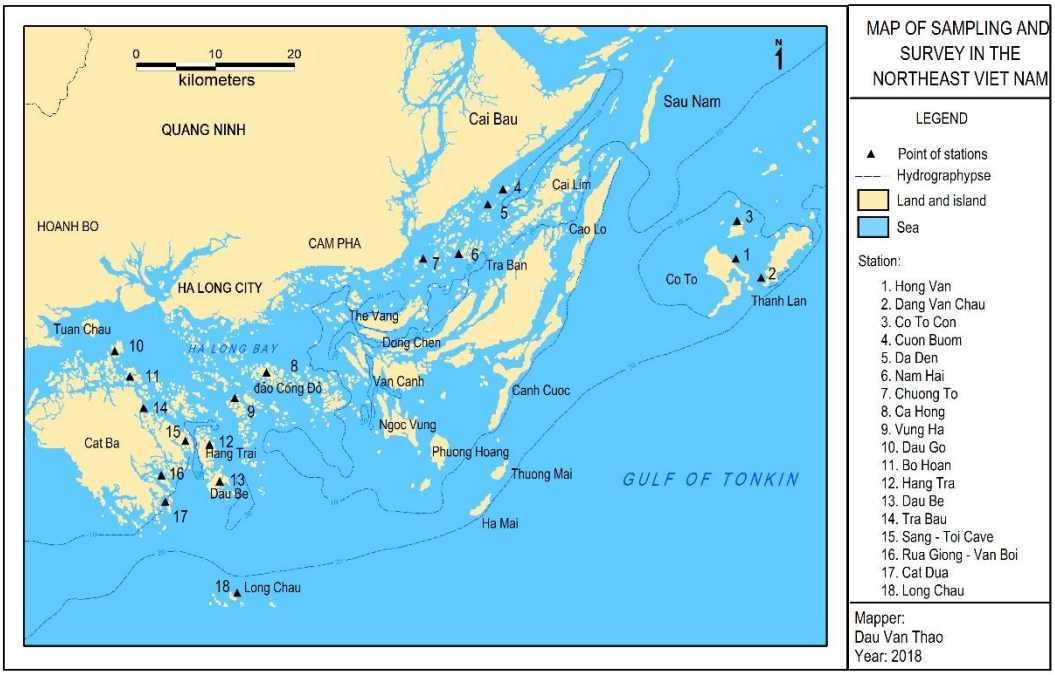

Figure 1-a. Sampling points of Northeast island and archipelagos areas.

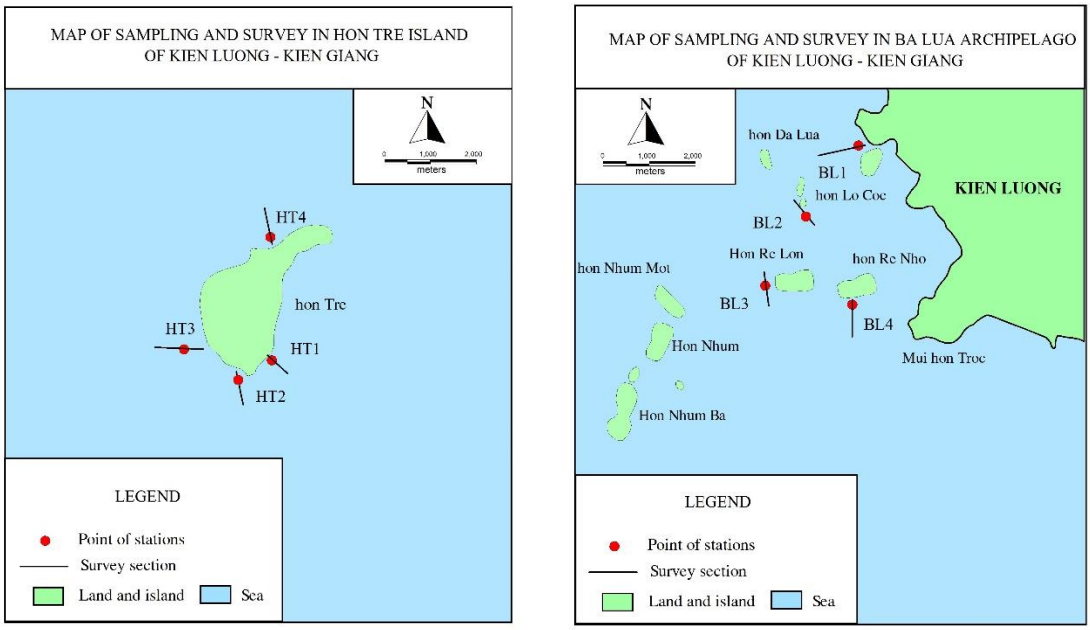

Figure 1-b. Sampling points of Kien Luong (Kien Giang) area.

Data treatment

To assess the seawater quality of limestone islands and archipelagos, the National Technical Regulations on Marine water quality QCVN 10-MT: 2015/BTNMT for protection of aquaculture and aquatic life [11] and the QCVN 8-MT:2015/BTNMT for surface water quality were used [12]. In addition, ASEAN criteria [13] was used to assessed water quality parameters that are not regulated in National Technical Regulation. Besides, using the risk quotient (RQ) for assessment of water quality. The risk quotient was calculated as follow [14]:

$$RQi = \frac{\text{Concentration of pollutant i in water}}{\text{regulated limitation value for parameter i}}$$

If RQ value <0.75: the water is not contaminated, 0.75 <RQ <1: water is at risk of contamination and RQ> 1: water is contaminated. As for DO, the RQ value of DO is calculated by dividing the regulated limitation value by the measured value.

3. Results and Discussion

Survey and analysis results of seawater quality around limestone islands and archipelagos of Vietnam in 2017 – 2018 are presented in Table 1 (December 2017) and Table 2 (August 2018).

Hydrochemical and water quality characteristics

Hydrochemical factors in seawater around of limestone islands and archipelagos

- Temperature: seawater temperature of limestone islands and archipelagos ranged from 20.4 to 32.5^{o}C, in which the difference in water temperature between rainy and dry seasons was quite clear in the Northern islands and archipelagoes. In the North, during the rainy season, due to coinciding with the summer months, the rising water temperature can affect the growth of ecosystems of coral reef, fish, etc.

- pH: The pH value ranged from 7.45 to 8.35 units in which the low pH was found in the areas of Ha Long Bay and Cat Ba island during the rainy season. The pH of seawater was quite stable, fluctuating around 8, showing the great role of the carbonate - bicarbonate buffer system in seawater, balancing ions in the water body, helping to stabilize water quality.

- Salinity: Salinity of water around of limestone islands and archipelagos ranged from

17 to 32 ‰ in which in the dry season, the salinity was high and stable, the average value of all areas was 29.5 ‰. In rainy season, due to the influence of fresh water from inland, salinity of water decreased, especially in the areas of Ha Long Bay and Cat Ba island, the average value of all areas in rainy season was 28 ‰.

- Turbidity: Turbidity of seawater was quite low, ranging from 1 to 6 NTU, which was very favorable for the development of coral reefs and photosynthesis of aquatic plants. In rainy season, water tended to be more turbid compared to that in dry season due to the stirring of rainwater and river water.

Organic substances consuming oxygen

To assess the pollution of organic matters in water, the parameters of dissolved oxygen, biochemical oxygen demand and chemical oxygen demand are used.

- Dissolved oxygen (DO) concentration: DO in seawater around of limestone islands and archipelagos was quite high, the value was in the range of 5.67 - 7.02 mg/l, with an average value of 6.64 mg/l in the dry season and 6.49 mg/l in the rainy season. These values were within the limitation value regulated in the QCVN 10: 2015/BTNMT for coastal water. According to Thung et al (2018) [15], water in the aquaculture areas in the Northeast islands was insufficient dissolved oxygen, with many samples had DO concentrations in water less than 5 mg/l. Thus, the seawater of the limestone island and archipelago area in the 2017-2018 surveys had a higher concentration of dissolved oxygen, related to the sampling site that far from the aquaculture areas. The high concentration of dissolved oxygen in water is favorable to the growth and development of organisms.

Table 1. Average value of water quality parameters in limestone islands of Vietnam (Dry season-December 2017)

Parameters	Ha Long Bay (n=11)	Bai Tu Long Bay (n=4)	Cat Ba (n=8)	Kien Luong (n=6)
Temperature, ^oC	21.15±0.69	20.63±0.25	21.2±0.35	27.3±0.39
pH	7.96±0.15	7.92±0.06	8.07±0.10	8.07±1.87
Salinity (‰)	28.5±5.5	29.3±0.58	30.1±0.35	29.8±0.98
Turbidity (NTU)	2.8±2.0	2.3±3.7	2.5±1.6	2.8±1.0
DO, mg/l	6.65±0.21	6.51±0.08	6.54±0.35	6.81±1.87
BOD_5, mg/l	1.96±0.42	2.25±0.41	1.73±0.19	2.23±0.49
COD, mg/l	2.64±0.64	2.92±0.36	2.40±0.18	3.18±0.71
$N\text{-}NO_2^-$, µg/l	27.89±10.55	8.33±1.26	38.03±8.49	8.09±2.43
$N\text{-}NO_3^-$, µg/l	43.28±12.96	39.20±2.73	44.67±7.14	28.11±2.67
$N\text{-}NH_4^+$, µg/l	37.61±19.27	22.83±2.37	38.15±14.48	17.62±1.06
$P\text{-}PO_4^{3-}$, µg/l	38.07±7.73	36.14±4.40	129.75±10.94	21.11±7.11
N-T (mg/l)	0.76±0.17	0.70±0.15	1.92±0.11	0.49±0.05
P-T (mg/l)	0.37±0.33	0.60±0.03	0.64±0.33	0.14±0.02
Oil and grease, mg/l	0.19±0.06	0.26±0.05	0.23±0.03	0.14±0.07
Cu, µg/l	59.50±21.65	73.63±5.48	74.77±9.28	58.18±1.14
Pb, µg/l	1.56±1.47	0.27±0.06	0.31±0.23	1.88±0.22
Zn, µg/l	18.35±8.43	15.24±2.89	11.35±0.56	34.17±2.32
Hg, µg/l	0.34±0.17	0.25±0.06	0.19±0.05	0.14±0.07
Total OCP, ng/l	4.33±4.74	2.68±4.64	4.69±4.43	2.99±2.83

Note: OCP - Organochlorinated pesticides; n = number of samples; NA-data not available; ND-not detected.

Table 2. Average value of water quality parameters in limestone islands of Vietnam (Rainy season -August 2018)

Parameters	Ha Long Bay (n=12)	Bai Tu Long Bay (n=10)	Cat Ba (n=8)	Long Chau (n=7)	Kien Luong (n=6)
Temperature, ^oC	31.83±0.68	31.25±0.43	31.0±0.37	29.44±2.49	28.1±0.74
pH	7.85±0.25	7.99±0.07	8.00±0.09	8.23±0.09	8.07±1.87
Salinity (‰)	23.8±5.9	23.7±1.75	27.3.1±1.52	32±0.8	29.3±0.82
Turbidity (NTU)	2.9±1.7	3.3±1.7	2.4±0.6	4.8±0.4	3.2±1.3
DO, mg/l	6.47±0.47	6.10±0.55	6.29±0.66	6.78±0.89	6.70±
BOD_5, mg/l	1.66±0.09	1.62±0.10	1.69±0.09	1.64±0.24	2.31±0.05
COD, mg/l	2.53±0.21	2.33±0.19	2.32±0.14	2.52±0.17	3.06±0.11
$N\text{-}NO_2^-$, µg/l	23.19±5.94	10.85±1.66	15.22±7.78	24.66±6.64	22.29±12.38
$N\text{-}NO_3^-$, µg/l	35.70±7.56	52.63±13.68	54.09±7.03	NA	93.94±13.10
$N\text{-}NH_4^+$, µg/l	49.69±5.36	33.81±8.25	41.69±6.85	48.54±5.11	248.14±189.0
$P\text{-}PO_4^{3-}$, µg/l	18.70±3.21	20.83±3.58	21.89±5.55	66.39±15.77	27.00±4.30
N-T (mg/l)	1.03±0.32	0.81±0.24	1.93±0.36	NA	1.31±0.59
P-T (mg/l)	0.11±0.03	0.07±0.02	0.61±0.18	NA	0.27±0.18
Oil and grease, mg/l	0.19±0.05	0.16±0.07	0.19±0.03	0.29±0.13	0.09±0.05
Cu, µg/l	25.38±9.82	25.04±7.08	22.95±2.49	21.70±8.75	28.33±0.94
Pb, µg/l	0.89±0.39	4.73±5.38	0.61±0.61	8.97±8.29	2.00±0.83
Zn, µg/l	29.51±5.48	25.86±6.37	22.59±9.86	53.24±16.78	11.33±0.47
Hg, µg/l	0.23±0.17	0.16±0.12	0.12±0.03	0.07±0.01	ND
Total OCP, ng/l	1.75±2.05	0.35±0.68	2.56±1.72	23.12±51.32	0.75±0.70

Note: OCP - Organochlorinated pesticides; n = number of samples; NA-data not available; ND-not detected.

- Biochemical oxygen demand (BOD): BOD was in the range of 1.42-3.18 mgO_2/l. Comparison of biochemical oxygen demand among areas, it was found that water in Long Chau had the lowest biochemical oxygen demand, followed by Cat Ba island, Ha Long Bay - Bai Tu Long Bay, and Kien Luong. These are tourist areas so the quality of water here is greatly influenced by tourism activities, especially in Kien Luong area. Compared to the limitation value regulated in QCVN 08: 2015/BTNMT for surface water (6mg/l), the water in the limestone islands and archipelagos of Vietnam did not show polluted by easy - decomposed organic matters.

- Chemical oxygen demand (COD): COD was in the range of 2.0 - 4.42 mg/l, of which high values were recorded at Cong Do (Ha Long Bay) in December 2017. Comparison of chemical oxygen demand among areas, it was found that water in Cat Ba and Long Chau islands had the lowest chemical oxygen demand, followed by Ha Long Bay - Bai Tu Long Bay area and Kien Luong. Compared to the limitation value regulated in the QCVN 08:2015/BTNMT for surface water quality (15mg/l), the water in the limestone islands and archipelagos of Vietnam did not polluted by organic matters. However, attention should be paid to the water in Kien Luong area that has a higher chemical oxygen demand than other areas.

However, according to the research of Thanh et al (2007) [5] in Ha Long Bay area, the average values of BOD and COD in seawater were 0.52 mg/l and 2.1 mg/l, respectively. Thus, there is an increase in the concentration of organic matter in the seawater of Ha Long Bay, especially for easily decomposed organic matter from 2007 to present.

Nutrients in water

The nitrite concentration ranged from 4.35 to 48.87µg/l, with the highest average value in Cat Ba island. Vietnam does not regulate the limitation value of nitrite concentration for

coastal water, however, this value in the ASEAN criteria is 55μg/l. Compared to this criteria, the water in the limestone islands and archipelagos of Vietnam was not contaminated by nitrite.

In contrast, the concentration of nitrate in water of limestone islands and archipelagos was quite high compared to the ASEAN criteria (60μg/l). The nitrate concentration ranged from 22.46 -119.17 μg/l of which water was polluted by nitrate in Cong Do (Ha Long Bay), Cat Chuong To (Bai Tu Long Bay), Cat Dua (Cat Ba). Especially, in Kien Luong area (Kien Giang province), the water was polluted by nitrate in August 2018 in all sampling points.

The concentration of ammonium in water varied strongly from 15.83 to 553.18 μg/l, in which the water in Northern limestone islands and archipelagos were not contaminated with ammonium if compared to the regulated value of QVN 10:2015/BTNM for coastal water (100μg/l), however, nitrate concentration in water tended to increase in the rainy season. While in Kien Luong, water was polluted by ammonium in the rainy season at most of the sampling points, especially at Hang Tuyen, the ammonium value was 5.5 times higher than the limitation value.

The increase of nitrogen mineral nutrients in water in rainy season at Kien Luong area may influence to the survival and growth of coral reefs, so it is necessary to have timely coral reef monitoring to prevent it from their degradation.

The phosphate concentration ranged from 9.8 to 175.73 μg/l, lower than the limitation value regulated in QCVN 10: 2018/BTNMT for coastal water (200 μg/l). However, compared with ASEAN criteria (45 μg/l) for protection of aquatic life, the area of Vung Ha (Ha Long Bay) and Cat Ba was polluted by phosphate, especially in the dry season.

The total nitrogen concentration in water ranged from 0.42 mg/l to 2.7 mg/l, of which the high concentration was found in Cat Ba in both rainy and dry seasons. The total phosphorous concentration in water ranged from 0.04 mg/l to 1.26 mg/l, of which in the dry season (December 2017), the total phosphorus concentration was much higher than that in the rainy season in Ha Long Bay and Bai Tu Long bay. In particular, in Cat Ba limestone island area, the total phosphorus concentration in water was high in both dry and rainy seasons. This demonstrates that the pollution sources of N and P from the mainland have impacted the nutrient level in seawater in the Northeast islands.

The increasing in the nutrient concentration of nitrogen and phosphorus in water was recorded when compared to the study results of Thanh et al 2007 in Ha Long Bay area. The survey results of in 2007 with 20 sampling points in Ha Long Bay showed that the average values of N-T and P-T were 0.45 mg/l and 0.09 mg/l, respectively [5]. Compared with the results in this study, the nutrient concentration in water of Ha Long Bay has increased nearly 2 times for N and 2.7 times for P.

Pollutants

- Oil and grease: oil and grease content in water around of limestone islands archipelagos ranged from 0.05 to 0.5mg/l, at most of the sampling points, oil contents were less than the limitation value regulated in the QCVN 10: 2015/BTNMT for coastal water (0.5mg/l). However, if compared with the ASEAN criteria (0.14 mg/l), the water in all areas is contaminated with oil and grease, except for the Kien Luong area.

- Heavy metals: among the 4 heavy metals (Cu, Pb, Zn and Hg) monitored, there was no pollution of these heavy metals in seawater around of limestone islands and archipelagos observed.

- Organochlorinated pesticides (OCPs): the analysis results of OCPs in water around of limestone islands and archipelagos showed that the total OCPs residue ranged from trace amount to 139.48 ng/l in which the highest concentration was detected in the Long Chau island. Except for the unusual high residue in one sample at Long Chau area, the average value of total OCPs fluctuates quite similarly between areas (from 0.7 to 4.7 ng/l), the water was not contaminated by these compounds.

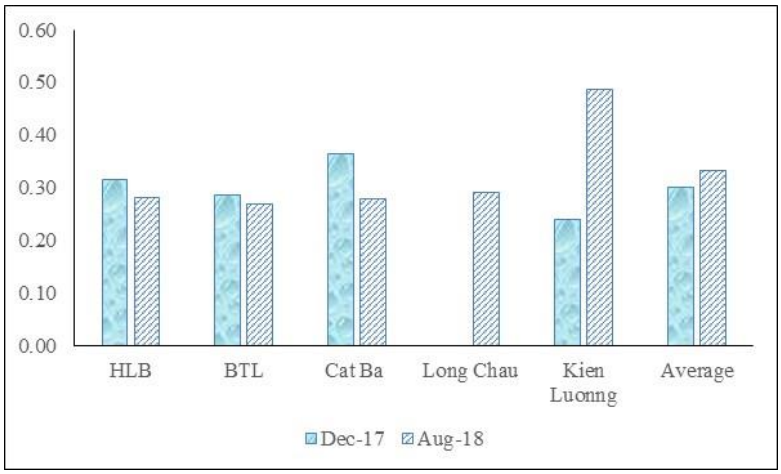

Figure 2. Risk quotient for water quality in limestone islands and archipelagos of Vietnam.

Risk quotient for water quality in limestone islands and archipelagos

Survey data shows that, in the dry season (December 2017), the water around of limestone islands archipelagos of Vietnam was quite good, the average risk quotient ranges from 0.24 to 0.36. Considering for all areas, the water quality of Kien Luong area was the best ($RQ_{average}$= 0.24), followed by Bai Tu Long Bay ($RQ_{average}$ = 0.29), Ha Long Bay ($RQ_{average}$ = 0.31) and finally Cat Ba island ($RQ_{average}$ = 0.36). The $RQ_{average}$ for all area is 0.30 (Figure 2).

In rainy season, there is a change in order, the best water quality was in Bai Tu Long bay ($RQ_{average}$ = 0.27), followed by Cat Ba island and Ha Long Bay ($RQ_{average}$ = 0.28), Long Chau island ($RQ_{average}$ = 0.29) and finally Kien Luong area ($RQ_{average}$ = 0.49). The $RQ_{average}$ for all areas was 0.33. Especially, in rainy season, the water in Bai Tu Long bay and Cat Ba island was at risk of nitrate pollution; the water in Kien Luong area was contaminated by nitrate and ammonium. The $RQ_{average}$ of all areas was higher in rainy season - Figure 2.

4. Conclusion

Investigation results on water quality in limestone islands and archipelagos of Vietnam in November 2017 and August 2018 showed that the water have low turbidity, high dissolved oxygen concentration, favorable for the development of organisms, especially the coral reefs. Water has a high and stable pH, the value fluctuating around 8 pH units. The salinity of water samples ranged from brackish to salty. On spatial distribution, the hydrochemical characteristics of water are different between areas, in which the areas of Ha Long Bay - Bai Tu Long Bay and Cat Ba islands are strongly influenced by the inland sources, so the value of dissolved oxygen and pH is low in rainy season. Meanwhile, Long Chau area is less affected by inland sources. Long Chau area can represent for the typical seawater area with high and stable salinity, high dissolved oxygen, and high pH.

The water was polluted by nitrate when compared to the ASEAN criteria (60μg/l) in some sampling points such as Cong Do, Cat Chuong To (in Ha Long Bay) and Kien Luong (Kien Giang), especially in rainy season. In Kien Luong area, in rainy season, the water was polluted by ammonium with the average concentration was 2.5 times higher than limitation value. Water in Cat Ba island was polluted by phosphate during the dry season if compared to the ASEAN criteria (45μg/l - for estuary water); however, if compared to the limitation value in QCVN 10: 2015/BTNMT for coastal water (200 μg/l), the water was not

polluted by phosphate. Water in the observed limestone islands and archipelagos of Vietnam was not polluted by oil grease, heavy metals and organochlorinated pesticides. Although there were several samples showing contaminated by some pollutants, but these results were still within the limitation values.

In general, the water quality of the observed limestone islands and archipelagos of Vietnam is quite good and safe for the development of aquatic ecosystems. The research results have contributed to building a database of Vietnam's seawater quality, establishing a scientific basis to protect the marine environment in some island areas in Vietnam.

Acknowledgements

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