Phosphorus as a driver of nitrogen limitation and sustained eutrophic conditions in Bolinao and Anda, Philippines, a mariculture-impacted tropical coastal area

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Abstract

The dynamics of nitrogen (N) and phosphorus (P) was studied in mariculture areas around Bolinao and Anda, Philippines to examine its possible link to recurring algal blooms, hypoxia and fish kills. They occur despite regulation on number of fish farm structures in Bolinao to improve water quality after 2002, following a massive fish kill in the area. Based on spatiotemporal surveys, coastal waters remained eutrophic a decade after imposing regulation, primarily due to decomposition of uneaten and undigested feeds, and fish excretions. Relative to Redfield ratio (16), these materials are enriched in P, resulting in low N/P ratios (~6.6) of regenerated nutrients. Dissolved inorganic P (DIP) in the water reached 4 μM during the dry season, likely exacerbated by increase in fish farm structures in Anda. DIP enrichment created an N-limited condition that is highly susceptible to sporadic algal blooms whenever N is supplied from freshwater during the wet season.

Keywords mariculture, eutrophication, fish feed, phosphorus, nutrient ratios, Bolinao

1. Introduction

Fish farming in coastal areas (or mariculture) has become widespread in Southeast Asia including the Philippines, providing important food source and economic gains to the local people (FAO, 2009). However, intensified and unregulated fish farming produces exogenous materials such as wasted feeds that can lead to deterioration of water and sediment quality from excess nutrients and high turbidity; and frequent occurrence of algal blooms (Holmer and Kristensen, 1992; Folke et al., 1994; Wu, 1995; Kibria et al., 1996; Olsen et al., 2008; White, 2013). In tropical regions, mariculture areas are typically located adjacent to coral reefs and seagrass beds (e.g., Herbeck et al., 2013; Tanaka et al., 2014). It is therefore important to assess the effects of particulates and excess nutrients from mariculture effluents to these ecologically and economically important components of the coastal ecosystem.

In the towns of Bolinao and Anda in the province of Pangasinan located in Northern Luzon, Philippines, a shallow coastal embayment connected to the outer ocean through a few narrow channels has become the site for fish farming in cages or pens. The mariculture of milkfish (*Chanos chanos*) in the area started in the 1970s, and has intensified in 1995 when the number of fish farm structures reached >1000 units by early 2002 (Verceles et al., 2000). This number is twice the allowable maximum of 544 units set by the local government of Bolinao based on hydrodynamic constraints (San Diego-McGlone et al., 2008). The large increase in number of fish farm structures led to eutrophic waters, reduced water exchange rate and development of hypoxic bottom water in the area, and in 2002 resulted in a massive fish kill that coincided with the bloom of a harmful algae identified as *Prorocentrum minimum* (Azanza et al., 2005; 2006; San Diego-McGlone et al., 2008). Soon after the fish kill, Bolinao reduced the number of fish farm structures to comply with the allowable limit. Despite this effort, there has been recurrence of harmful algal blooms (HABs) and fish kills

resulting in economic losses which suggest that water quality in Bolinao and Anda have continued to deteriorate (Yap et al., 2004; Azanza et al., 2005; San Diego-McGlone et al., 2008; 2014; Azanza and Benico, 2013; Escobar et al., 2013). There is absence of macroinfaunal communities and presence of mats of the sulfide oxidizing bacteria *Beggiatoa* (Santander et al., 2008; Nacorda et al., 2012). There has been impact on the adjacent coral reef ecosystems leading to low survivorship of juvenile corals (Villanueva et al., 2006) and disappearance of seagrass species (Tanaka et al., 2014). Moreover, an accumulation of the indicators of non-cholera pathogens in the anoxic guts of milkfish and in the sediments was detected (Reichardt et al., 2013).

It is noteworthy that even if regulation on allowable number of fish farm structures has been enforced in Bolinao after 2002, the levels of nitrate (NO_3^-), ammonium (NH_4^+), and particularly phosphate (PO_4^{3-}) remained high (Fig. 2; San Diego-McGlone et al., 2008). The question is why these nutrients, especially P are persisting in these coastal waters.

Phosphorus (P) is a major nutrient in marine systems as well as nitrogen (N) and silicon (Si). Due to its highly reactive nature, P can be the ultimate limiting nutrient in some aquatic environments (reviews by Benitez-Nelson, 2000; Ruttenberg, 2003; Paytan and McLaughlin, 2007; MacKenzie et al., 2011; Slomp et al., 2011). Natural sources of P to coastal areas include atmospheric dusts, volcanic ashes, weathering products and upwelling; the inputs of which may gradually increase the concentrations in the water and enhance the supply of organic matter in the area, a process known as “natural eutrophication” (Nixon, 1995; Smith et al., 1999; Andersen et al., 2006). Major anthropogenic P sources include agriculture, livestock, chemical and human wastes from the watershed, and fish farming activities. Large loading of these sources to coastal environments due to human activities can result in accelerated buildup of nutrients and organic matter, otherwise known as “cultural eutrophication” (Hasler et al., 1969; Smith et al., 2009). Subsequently, the rate of organic

matter accumulation in the sediments can increase followed by development of hypoxia. Under reducing conditions, P which is closely linked to iron (Fe) and sulfur (S) cycle, is released to sediment porewater and overlying water (Kemp et al., 2009; Martin, 2009; Middleburg and Levin, 2009; Dale et al., 2013).

In this study, we analyzed the trophic status of the coastal waters of Bolinao and Anda a decade after the enforcement of fish farm structure regulation with emphasis on (i) the different historical and spatio-temporal behavior of N and P, which apparently resulted in relative enrichment of P in the water column, (ii) the role of wasted feeds, metabolic products of fish (feces and excretions), and sediment porewater in regulating the nutrient availability in the water column, and (iii) the potential importance of land-derived dissolved inorganic nitrogen (DIN) as a trigger of massive algal blooms in this area. Other factors such as hydrodynamics in the mariculture area, and the increase of fish structures in the adjacent waters of Anda that influenced the nutrient enrichment in Bolinao, are discussed with their implication on the management strategy for mariculture.

2. Materials and Methods

2.1 Study site

Bolinao and Anda (16.20-16.46°N, 119.76-120.05°E) are two coastal towns in Pangasinan, northwestern Philippines that share semi-enclosed coastal waters (Fig. 1). Their environment is affected by the country's two prevailing seasons – the dry season from November to March characterized by less precipitation (monthly average of 23 mm from 2003-2013) and strong northeasterly winds, and the wet season from June to September marked by heavy precipitation (monthly average of 736 mm) and southwesterly winds

(precipitation and wind data from the Philippine Atmospheric, Geophysical and Astronomical Services Administration ‘PAGASA’, 2013).

The Bolinao and Anda coastal waters are characterized by relatively deeper areas (>10 m) on the west side of Guiguiwanen Channel that connects to the South China Sea, at the passage between Siapar Island and Cabarruyan Island adjacent to the Lingayen Gulf, and at the Caquiputan Strait leading to Tambac Bay (Fig. 1a). The depth decreases to an average of 5 m between Bolinao and the northwest side of Cabarruyan Island, and further decreases to an average of 1.5 m in Tambac Bay. Fish cages are constructed at depths >5 m, while fish pens are situated at depths about 3 m (Fig. 1b). Bani and Alaminos Rivers are two major rivers that drain to Tambac Bay (Fig. 1a) creating an estuarine environment especially during the wet season.

2.2 Water sampling

Water sampling and hydrodynamic surveys were conducted at several stations in March (dry season) and September (wet season) from 2010-2014 to determine seasonal, spatial and temporal variation in water quality characteristics (Figs. 1a, 1c). A floating platform (Continuous and Comprehensive Monitoring System or CCMS) equipped with sensors was installed in September 2011 midway (16.3868°N, 119.9252°E) at Guiguiwanen Channel to collect high temporal resolution data (e.g. water level, water velocity, etc.; Fortes and Nadaoka, 2015). Surveys include large-scale bay-wide spatial investigation on the reef area, deep and shallow mariculture area, and the deep strait up to Tambac Bay (Sep 2010-2013; Mar 2011, 2013, 2014). Continuous (24-hr) observations were conducted at the West Station (Sta. 1), CCMS (Mid-channel), and East Station (Sta. 2) of the Guiguiwanen Channel (Sep 2011, Mar 2012). A small-scale grid survey around CCMS was performed as well (Sep 2012, Mar 2013). During the grid survey, 25 stations in the vicinity of the CCMS station

occupying a 550m x 550m area were sampled to determine the spatial heterogeneity of the water quality characteristics in the dense mariculture area.

In addition, water samples from offshore (South China Sea side), from along a longitudinal section on the west side of Lingayen Gulf (16.07-16.50°N, 120.00-120.19°E), from Bani and Alaminos Rivers and their tributaries, and other rivers (in Caquiputan Strait and Cabarruyan Island), and from groundwater sites in Bolinao and Santiago Island were collected to determine the end-member characteristics of potential sources of nutrients (Fig. 1a). The results of the surveys were compared with the time-series nutrient and chlorophyll-*a* data from the Bolinao Marine Laboratory (BML) monitoring station (16.3815°N, 119.9125°E; Fig. 1c).

Vertical measurements of temperature, salinity, and dissolved oxygen were acquired at every sampling station using an AAQ-RINKO water quality profiler (JFE-Advantech, Japan) prior to water sampling. Water samples were collected at the surface using a bucket, and at one meter above the bottom using either a Van Dorn water sampler (10 L; Rigo, Japan) or a Niskin sampler (5 L; General Oceanics, USA). Seawater samples for nutrients (dissolved inorganic nitrogen, DIN = NO_3^- , NO_2^- , NH_4^+ ; dissolved inorganic phosphorus, DIP = PO_4^{3-}) and total dissolved nitrogen and phosphorus (TDN, TDP) were filtered through 0.80 μm cellulose acetate DISMIC filters (Advantec, Japan) into duplicate 10 mL acrylic tubes and were kept frozen at -20°C until analysis. Freshwater samples were collected at the surface and stored in the same manner as seawater samples.

Water samples for chlorophyll-*a* (Chl-*a*) and particulate phosphorus (PP) were pre-filtered through a 200 μm sieve attached to a plastic funnel and collected into polypropylene containers. For Chl-*a* analysis, 100 ml of water was filtered onto pre-combusted (450°C, 3 hours) 25 mm glass fiber filters (GF/F; nominal pore size, 0.7 μm ; Whatman GE Healthcare Life Sciences, England), immersed in 6 mL *N, N*-dimethylformamide (DMF), and kept in the

dark at -20°C for Chl-*a* extraction. For PP, 200 ml of samples were filtered onto pre-combusted 25mm GF/F filters and kept in plastic tubes. All samples were stored at $\leq -20^{\circ}\text{C}$ until analysis.

2.3 Sediment sampling, incubation, and porewater extraction

Duplicate sediment cores (25-35 cm in length) were collected using acrylic pipes (i.d. = 5.0 cm) by SCUBA from 3 stations along the Guiguiwanen Channel (Fig. 1c) at the West Station (depth = 17 m), Mid-channel (CCMS; depth = 15 m), and East Station (depth = 10 m) in March 2013 (dry season, neap tide) and September 2013 (wet season, spring tide). Sediment core sampling stations were similar to the water sampling stations except for the East Station where the core samples were obtained ~360 m northwest of the water sampling station. Bottom water temperatures ranged from 29.3-29.8 $^{\circ}\text{C}$ during the sampling periods.

Core samples were immediately transferred to a temperature-controlled laboratory after sampling. Original overlying water was siphoned out and replaced with freshly collected bottom water (about 300 ml). Homogeneity of the overlying waters was maintained by moderate air circulation ($\sim 9.2 \text{ ml min}^{-1}$) using small tubes (i.d. = 3.18 mm) and a peristaltic pump. The cores were kept in the dark during incubation. Samples for nutrients were collected from the overlying water of each core every 12 hours for 48-60 hours. Sediment cores in March were black, emitted sulfidic odor and were covered with thick mats of *Beggiatoa*. In September, the *Beggiatoa* filaments were not clearly visible but appeared at the surface layer after 24 hours of incubation, albeit of lower density. Net nutrient flux from the sediment ($\text{mmol m}^{-2} \text{ d}^{-1}$) was calculated from the linear regression of the change in the amount of nutrients in the water phase with sampling time.

Sediment cores were also collected at the West Station, CCMS and East Station in September 2011 to measure porewater nutrient concentrations. The sediment cores were cut

into 5 cm sections and transferred into 150 ml plastic containers. Two 30 cm³ of sediment samples were collected from each section using a cut-off plastic syringe and the sediment section transferred onto pre-cleaned 50 ml centrifuge tubes. The sections were centrifuged (1274 g, 15 min) and the extracted porewater were transferred using a pipettor into acrylic tubes, which were kept frozen at -20°C for nutrient analysis.

2.4 Chemical analyses

Nutrient concentrations were determined colorimetrically using a QuAatro 2-HR (SEAL Analytical Ltd., Germany and BLTEC K.K., Japan) and AACS-III (BRAN+LUEBBE, Germany) segmented flow autoanalyzers. Accuracy of results was confirmed using reference materials provided by KANSO Japan (<http://www.kanso.co.jp/eng/index.html>) except for NH₄⁺. The same autoanalyzer was used for TDN and TDP analysis but with a D2BX-02 chamber (BLTEC K.K., Japan) that performs high-temperature (120 °C) wet oxidation of dissolved organic material to inorganic forms prior to colorimetric determination (Grasshoff et al., 1999). Dissolved organic nitrogen and phosphorus (DON, DOP) were calculated from the difference between TDN (TDP) and DIN (DIP) (DON data are not shown in this paper).

Chl-*a* concentration was measured using a 10-AU Fluorometer (Turner Designs, USA) using the method for samples extracted in DMF solvent (Suzuki and Ishimaru, 1990).

Samples for particulate phosphorus (PP) were processed following the persulfate wet oxidation method (Suzumura, 2008). Oxidized samples were analyzed using the same autoanalyzer for nutrients.

2.5 Water sampling and analysis of the long-term time series data

Water samples for nutrients (NO₃⁻, NO₂⁻, NH₄⁺, PO₄³⁻) and Chl-*a* were collected at the surface layer of the BML monitoring station (Fig. 1c) seasonally from 1995 to 2000, and

daily between 0700H and 0800H from 2002 to 2013. Details of sampling, sample processing and chemical analysis for nutrients and Chl-*a* concentrations are described in San Diego-McGlone et al. (2008). The annual DIN/DIP ratio was calculated using this nutrient data set.

2.6 Satellite image analysis

Satellite images were used to identify temporal change in the number of fish farm structures and their spatial distributions in Bolinao and Anda coastal waters. Fish cages and fish pens were manually counted from the high resolution images in Google Earth acquired by QuickBird (November 2003) and Pléiades (January 2014), and from WorldView-2 (March 2010; spatial resolution = 1.84 m) images.

3. Results

3.1 Historical and spatio-temporal behavior of N and P

After the massive fish kill event in 2002, the number of fish farm structures in Bolinao was reduced to comply with the allowable limit (Fig. 2c). Nutrient data at the BML monitoring station showed that average concentration of NO_3^- (Fig. 2e) and Chl-*a* (Fig. 2d) somewhat decreased by 2005. NH_4^+ concentrations (Fig. 2f) were not significantly different before and after implementation of the regulation. Unlike the N species, the concentration of PO_4^{3-} (Fig. 2g) consistently increased to almost twice, from 0.5 to $\sim 1.4 \mu\text{M}$, during the 10 years after regulation was implemented. The average DIN/DIP ratio (Fig. 2h) has decreased from near the Redfield ratio of 16 in 2002 to less than 5 in 2013, except for 2011 and 2012 when wet season precipitation was unusually high (Fig. 2a).

Small-scale spatial nutrient distributions indicated high concentrations of NH_4^+ and DIP at the surface layer of the Guiguwanen Channel around the CCMS station (Fig. S1,

Supplementary Material). During the wet season, NH_4^+ (Fig. S1d) and DIP (Fig. S1e) were particularly high at the mid-section of the channel where fish cages are dense (Fig. S1a). Large-scale spatial nutrient distributions showed that DIN and DIP were generally higher in the mariculture areas than offshore and reef areas (Figs. 3a-d; Table 1). DIN at the mariculture areas are mostly composed of NH_4^+ except at the river mouth stations in Tambac Bay where NO_3^- input from rivers become apparent especially during the wet season (Fig. 3b). DIP was higher both at the surface and bottom layers of the mariculture areas in the dry season than in the wet season, especially at the relatively deep areas of the Guiguiwanen Channel and Caquiputan Strait (Figs. 3a-d); DIN/DIP ratios were generally lower than the Redfield ratio with slightly higher ratios at the surface layer during the wet season (seen as lower salinity, Fig. 3a). South China Sea and Lingayen Gulf waters adjacent to the mariculture areas were depleted in DIN relative to DIP, resulting in DIN/DIP ratios lower than the Redfield ratio, while the samples in reef area had near-Redfield ratio or higher. Rivers and groundwater were one- and two-orders of magnitude higher in DIN than the mariculture areas, respectively, and had very high DIN/DIP ratios (Table 1). The waters sampled at Bani and Alaminos River mouths at Tambac Bay also exhibited high DIN/DIP ratios particularly during the wet season (Fig. 3a).

Short-term continuous observation (24-h) at the CCMS station also indicated higher surface DIP concentrations during the dry season (Fig. 4b). DIP was usually the most dominant form of P representing 25 – 89%, and PP (11 – 71%) was usually more abundant than DOP (0 – 29%). Temporal change in bottom DIP was influenced by the tidal cycle. During the wet season sampling when tides were semidiurnal, two peaks in bottom DIP concentration were observed that corresponded with change in phase of tide (ebb to flood at 1300H; flood to ebb at 0300H; shaded areas; Figs. 4a, c). Flow velocities at the bottom layer were rather weak, but flow direction was into the channel (southeast direction) during flood

phase, and out of the channel (northwest) during ebb phase (Fig. 4e). Both the bottom DIP peaks were synchronized with decrease in bottom dissolved oxygen concentrations (Fig. 4g). In the dry season sampling when tides were diurnal, bottom DIP decreased and DO concentration increased as water flowed into the channel during flood tide (Figs. 4b, d, f, h). The highest DIP concentration was obtained after the peak of ebb flow when the water column was well mixed (salinity difference of 0.05 between surface and bottom).

Comparison of nutrient concentrations along the Guiguiwanen Channel during the short-term temporal observation (Table 2) showed highest DIP at the surface layer in the CCMS compared to the West Station and East Station. Bottom DIP was relatively depleted at the Mid-channel but enriched at the West and East station. The East station, which had the highest bottom DIP (1.5 μM) and DIN (11 μM), also had the lowest DO (2.5 mg L^{-1}). Surface DIN/DIP ratios were higher at the Mid-channel than the West and East stations. Bottom water DIN/DIP ratios were not significantly different among the stations (range of average values = 6.7-8.5).

3.2 Vertical profile of porewater nutrients and nutrient flux from sediment

Porewater DIN and DIP at the Guiguiwanen Channel were two- to three-orders of magnitude higher than water column concentrations (Figs. 5a, b). DIN is composed mainly of NH_4^+ (~99%); NO_2^- concentrations are typically <1-10 μM (1% of DIN); NO_3^- concentrations at times reach up to 16 μM but most data are below the detection limit of the instrument. At the CCMS station, DIN (Fig. 5a) generally increased from the upper sediment layer (~1000 μM) to deeper sections of the core (~3000 μM). At the East station, a sub-surface maximum of DIN was seen at 10-15 cm and decreased with depth. DIP (Fig. 5b) at upper sediment layer (~100 μM) increased to a sub-surface maximum at 10-15 cm, and decreased to values similar to the upper layer (at the CCMS station) or to lower values (at the

East station). Among the three sediment core stations, the West station had the lowest DIN and DIP porewater concentrations.

The average DIN and DIP fluxes from the sediments ranged from -0.16 to 1.90 mmol m⁻² d⁻¹ and -0.01 to 0.20 mmol m⁻² d⁻¹, respectively (Table S1). DIN fluxes are mostly constituted by NH₄⁺ fluxes and NO₃⁻ and NO₂⁻ fluxes are almost negligible except at the Mid-channel during the dry season when NO₃⁻ flux contributed 43% of the DIN flux. DIN fluxes were always higher at the West and CCMS stations compared to the East station. There was sediment uptake of both DIN and DIP at the East station during the wet season. While the DIN flux was not significantly different between the dry and wet seasons, DIP flux was lower during the wet season that resulted in higher DIN/DIP flux ratios.

4. Discussion

4.1 Nutrient dynamics in the Bolinao and Anda coastal waters: nutrient sources and reasons for sustained P enrichment

The urgency in establishing and maintaining sound water quality conditions in the coastal area of Bolinao and Anda is driven by substantial economic losses incurred due to repeated incidents of hypoxia-induced fish kills following collapse of algal blooms. Despite implementing regulations on allowable number of fish farm structures in Bolinao, nutrients such as DIP have increased to twice the levels in the 1990s. Long-term time series data (Fig. 2; San Diego-McGlone et al., 2008; 2014) and the results of our surveys show that the waters have continued to be eutrophic with values at times exceeding the ASEAN Water Quality Criteria for coastal waters (NH₄⁺ = 5.0 μM, PO₄³⁻ = 1.5 μM; ASEAN, 2004). Understanding the major sources and mechanisms of nutrient turnover, particularly of DIP, at various time scales is necessary for evaluating merits of the fish farm structure regulation imposed.

Seasonal variation of DIN and DIP showed accumulation of nutrients in bottom waters of the mariculture area with a constant DIN/DIP ratio (~ 6.6 ; Fig. 6a) during the wet season when water column was strongly stratified (Fig. 6c inset). The strong negative correlation of DIP with DO for bottom waters (Figs. 6c) suggests organic matter decomposition as the source of DIP enrichment. This accumulation is presumably due to recent regeneration of nutrients from detrital organic matter in the water column and/or the sediment. It is expected that, when the regenerated nutrients, whose N/P ratio is much lower than the Redfield ratio, become available for phytoplankton in the euphotic layer, DIN should be consumed and depleted faster than DIP, resulting in N-limited conditions in the phytoplankton populations. This study has seen the lower DIN/DIP ratio in surface waters, with DIN getting depleted in this layer (Fig. 6a). Such depletion of DIN during the wet season may be attributed to active uptake by phytoplankton, as evidenced by the concomitant conversion of DIP into PP during the 24-h survey (Fig. 4a; between 0700H to 1300H) and higher Chl-*a* during the grid survey (Fig. S1f). Seasonal trend showed higher concentrations of DIP both at the surface and bottom waters during the dry season than the wet season (Figs. 3, 6a-b). This may be due to enhanced accumulation of regenerated nutrients inside the embayment due to the direction of residual current during the dry season (Yoshikai et al., in preparation). The influence of vertical mixing of water column during the dry season (Fig. 6c inset) may be seen in the more scattered distribution of DIN and DIP (Fig. 6b).

Other factors contributing to DIP enrichment include hypoxic bottom water conditions that result in reductive dissolution of the DIP bound to Fe-hydroxides in the sediments (Martin, 2009; Middleburg and Levin, 2009). Bottom water hypoxia during neap tide was seen from continuous observations in the middle of Guiguiwanen Channel (Fortes and Nadaoka, 2015; Yoshikai et al., in preparation). Higher DIP was also seen during hypoxic bottom water conditions (Figs. 6c, 6d). The higher bottom DIP at the Caquiputan

Strait station during both wet and dry seasons (Figs. 3b, 3d, 6c, 6d) suggest that this area is a source of DIP for both seasons.

The extensive use of low quality feeds (fast sinking and disintegrating; Magdaong and Villanoy, 2008) may be the primary source of these regenerated nutrients resulting in the persistent eutrophication of Bolinao and Anda mariculture area. These feeds may contain P that exceeds the dietary phosphorus requirement ($\sim 0.85\%$ of dry diet; Borlongan and Satoh, 2001) for optimal growth (biomass) and calcification (bones and scales) of juvenile milkfish. These low-quality feeds may have $\sim 1.1\%$ of P, similar to a local commercial feed with comparable feed formulation (e.g., % of crude protein, crude fiber, crude fat and ash content; Sumagaysay-Chavoso, 2003). The N/P ratios of fish feeds used in the mariculture area (6.7 to 12.2, Table 1) are significantly lower than the ratio in the fish harvested (14.4; Holmer et al., 2002). This implies that fish feeds contain P in large excess of N relative to the fish requirements, which, in turn, makes the average fish feces (5.4 ± 0.2) and excreta (6.2 ± 0.3) enriched in P rather than N (Sumagaysay-Chavoso, 2003). In a study done on different sizes of milkfish fed by different diets including commercial feeds and reared in semi-indoor tanks, 47.2-58.5% of consumed P were lost as feces and 18.7-42.6% were excreted (Sumagaysay-Chavoso, 2003). This confirms that a smaller percentage of feed P is recovered in the harvested fish. Fish feeds used in Bolinao and Anda have a high feed conversion ratio (FCR ~ 2.5 ; the ratio of the amount of feed needed to grow 1 kg of fish; White et al., 2013; Fortes and Nadaoka, 2015) which translate to high feed wastage. Given an N/P ratio of 9 in fish feed, an N/P ratio of 14.4 in milkfish grown in fish pen (size of 2,886 m², depth of 3m), and harvest of 34,050 fish, the release rate of P in a 180-day grow-out period is $\sim 1.05 \mu\text{M d}^{-1}$, a value high enough to explain the high DIP concentrations in the mariculture area (see the Supplementary Material for detailed calculation).

Wasted feeds and particulate feces undergo decomposition in the water column and in the sediments. Relative to the other sources, sediment porewater has the highest concentration of DIN and DIP (Table 1). In this study, the DIN/DIP ratio in sediment pore waters (10.8 ± 4.7) overlapped the range of N/P ratio of fish feeds rather than that of fish feces, which suggests that porewater nutrients could be sourced mainly from remineralization of wasted unused feeds deposited in the sediments. The observed porewater DIP concentration ($> 100\mu\text{M}$; Fig. 5) is quite high and such concentrations have rarely been reported even in the porewater from embayments near big cities (Umezawa et al. 2015). This indicates that the area reached a hypertrophic state even after fish structure regulation. The porewater DIP in Bolinao reported by Holmer et al. (2002) (measured in Dec 2000) never exceeded $50\ \mu\text{M}$ except for one case. The porewater DIP observed in our study strongly suggest that P is retained in the system and there is sustained eutrophic condition with respect to P even after the fish farm structure regulation started.

The fluxes of DIN and DIP from the sediments appear to be controlled by the oxygenation status, as illustrated by the negative correlation in DO vs. DIN (mostly NH_4^+) and DIP in the stratified bottom water during the wet season (Figs. 6a, 6c). The sediment fluxes measured from cores collected underneath fish cages (Table S1) were actually in the lower limit of those reported by Holmer et al. (2003) (NH_4^+ flux = $1\text{-}22\ \text{mmol m}^{-2}\ \text{d}^{-1}$; PO_4^{3-} flux = $0.2\text{-}4.7\ \text{mmol m}^{-2}\ \text{d}^{-1}$) and lower than those seen under blue-fin tuna sea cages in Australia (maximum NH_4^+ flux = $2.4 \times 10^2\ \text{mmol m}^{-2}\ \text{d}^{-1}$; maximum PO_4^{3-} flux = $52\ \text{mmol m}^{-2}\ \text{d}^{-1}$; Lauer et al., 2009). Considering maximum DIN and DIP fluxes of 1.0 and $0.2\ \text{mmol m}^{-2}\ \text{d}^{-1}$ and a water depth of $10\ \text{m}$ for cages, the benthic flux contribution of 0.1 and $0.02\ \mu\text{M d}^{-1}$, respectively, may not be sufficient to explain the sustained eutrophic conditions in the mariculture area. The lower benthic fluxes could be an effect of fish cage regulation in Bolinao that may have improved water circulation and oxygenation status in the mariculture

area. Also, the fluxes calculated in this study may have been underestimated due to the oxic conditions during laboratory experiments. Lower *in situ* benthic PO_4^{3-} fluxes were seen by Holby and Hall (1991) underneath fish cages manipulated under oxic conditions. Moreover, the lower DIP fluxes, especially during the wet season when tidal mixing may have resuspended and oxygenated the sediments, may be due to the uptake by *Beggiatoa*, the sulfide-oxidizing bacteria. *Beggiatoa* have been reported to modify the DIP concentrations in hypoxic and sulfidic sediments (Brock and Schulz-Vogt, 2011; Dale et al., 2013). Nonetheless, the contribution of sediment fluxes to DIN and DIP enrichment through internal loading remains relatively minor compared to the direct impact by feed wastage and fish metabolites.

Based on satellite images, there has been an increase in fish farming structures in Anda over the years, especially for fish pens (Fig. 7). This is because the fish culture regulation in 2002 has been put in operation only in Bolinao area but not yet in the coastal area of Anda. Fish cages have also been installed in the relatively deep areas east of Siapar Island. This proliferation in fish farm structures is expected to increase wasted feeds, fish feces and excreta. Given that Bolinao and Anda share common waters, the increased mariculture activity in Anda is one of the likely reasons why waters in Bolinao have been persistently eutrophic. During the dry season, DIP from deteriorated waters and additional fish farm structures in Anda may reach Bolinao as residual current flows towards the Guiguiwanen Channel (Yoshikai et al., in preparation). Congestion of fish cages and pens in Anda along the channel will increase the residence time of deteriorated water and decrease flushing rates thereby increasing the likelihood of occurrences of hypoxia and its negative effects. This suggests that current fish farm structure regulations may have become ineffective in controlling and improving the water quality conditions in the mariculture areas of Bolinao and Anda, especially if only Bolinao has implemented the regulation.

4.2 Persistent algal blooms and other implications of sustained P pollution

Algal blooms in Bolinao and Anda happen during the onset of the wet season (e.g. May 2006, May 2010, Mar 2011), with fish kills likely occurring as dissolved oxygen is exhausted during collapse of algal blooms (San Diego-McGlone et al., 2008; Azanza and Benico, 2013; Escobar et al., 2013). Increased freshwater nutrient input from the watershed during the wet season could alleviate the N-limited conditions and consequently trigger phytoplankton blooms in the mariculture areas. The onset and the length of the rainy season have also been reported to be the key factors for the occurrence of algal blooms along the coasts of Hong Kong and Japan (Ho et al., 2010; Onitsuka et al., 2015).

Rivers are significant sources of DIN especially NO_3^- during the wet season. Fluctuations in NO_3^- concentrations in recent years (Fig. 2e) seem to depend on input from the watershed that was controlled by annual rainfall (Fig. 2a). The elevated annual average in DIN/DIP ratio in the water column when the annual rainfall was higher than normal years (Figs. 2a, 2h) implies input of additional DIN, especially NO_3^- , to coastal waters via rainfall. Nitrate levels in rainwater collected at the Bolinao Marine Laboratory was usually lower than $10\text{ }\mu\text{M}$, although concentrations higher than $20\text{ }\mu\text{M}$ were occasionally recorded during onset of the rainy season (Miyajima et al., unpublished data). However, when compared to rainwater, much higher concentrations of NO_3^- were seen in river waters and groundwater (Table 1). Therefore, NO_3^- originated from agricultural and domestic wastes disposed in the watershed and produced through nitrification in aquifers and river channels is considered to be the dominant source for river and groundwater NO_3^- (Mulholland, 1992; De Simone and Howes, 1998). Most of river water and groundwater sites sampled in this study contained DIN in large excess of DIP in terms of the Redfield ratio.

To better understand seasonal nutrient trends, the monthly average from historical data of nutrients (San Diego-McGlone et al. 2008; 2014) as well as DIN/DIP ratio were determined (Fig. 8). Data showed an increase in DIN/DIP ratio with rainfall during the wet season (June-September; Figs. 8b, 8h). The extent of NO_3^- carried by the rivers to the mariculture area may be seen in the northward flow of surface residual currents during the wet season based on numerical simulations (Yoshikai et al., in preparation). On the other hand, DIP in surface waters increased during the start of the dry season (Nov-Dec; Fig. 8g), which may be due to enhanced accumulation of regenerated nutrients, and vertical mixing of surface and bottom waters. The decrease in DIP during the latter part of the dry season (Mar-May) can be ascribed to uptake for primary production as solar radiation increased (Fig. 8a).

Hydrodynamic simulations (Yoshikai et al., in preparation) show possible connectivity of the mariculture area with the adjacent seagrass and coral reef in Santiago Island. This means that mariculture effluents could reach the seagrass and reef area especially during the wet season. Thus, it is important to also consider the economic value of the adjacent reef area (Cruz-Trinidad et al., 2011) that may be compromised by fish farming activity. These various considerations must be in place in order to appropriately model the biogeochemical dynamics and consequences of fish farm regulation in such a complex coastal environment.

5. Summary and Conclusion

Sustainable mariculture operations require the balance of economic gain and ecosystem health. However, in the case of Bolinao and Anda coastal waters, water quality has deteriorated with eutrophic conditions persisting even a decade after enforcement of fish farm structure regulation in Bolinao. The results of this study and comparison with historical information underscore the importance of P dynamics in understanding the environmental

impact of mariculture activities and assessing the effectiveness of fish farm regulation on recovery of ecosystem integrity. The sustained high DIP concentrations in the mariculture area is primarily due to decomposition of organic matter from unconsumed feeds and fish by-products (feces and excretions) in the water column and sediments. DIP enrichment is from overfeeding and use of low-quality feeds that contain P in excess of the metabolic requirements of the fish. Internal loading from the sediments, which may be enhanced with the development of benthic hypoxia, also supply DIP to the overlying water although of lesser magnitude. Other factors that may have contributed to high DIP during the dry season include enhanced accumulation of regenerated nutrients inside the mariculture areas due to prevailing residual current, and vertical mixing of the water column which could cause resuspension and enhanced benthic efflux of DIP under strong bottom turbulence and shear stress conditions. In addition, the increase in density of fish farm structures in Anda contributes DIP through advection of deteriorated waters towards Bolinao during the dry season. All these factors have exacerbated DIP enrichment relative to DIN, and have rendered these coastal waters as N-limited. The DIN supplied by rivers at onset of the wet season is critical for the formation of algal blooms with consequent hypoxia and fish kills.

Although intrusion of adjacent offshore waters may be a source of oxic and lower nutrient waters to the mariculture area, obstruction of flow by the recent addition of fish farm structures at the channel connections may hinder a successful recovery from eutrophication and its adverse effects. Considering the uncontrollable effects of global environmental factors such as variation in precipitation and ocean warming, it is therefore of importance to implement measures to reduce local impacts that cause eutrophication. These include improving the FCR ratios of feeds and decrease in feed input through improvement of fish feed formulation, regulating DIN from the watershed, and participatory maintenance of allowable number of fish farm structures in the Bolinao and Anda coastal waters. There

should be a joint cooperative management of fish farm structures in the shared coastal waters of Bolinao and Anda, otherwise efforts only by Bolinao will remain unsuccessful and insufficient.

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References

- Andersen, J.H., Schluter, L., Aertebjerg, G. (2006) Coastal eutrophication: recent developments in definitions and implications for monitoring strategies. *Journal of Plankton Research* Vol. 28(7), pp.621-628
- ASEAN (2004) Marine water quality criteria for the ASEAN region. <<http://www.aseansec.org/cme/Marine%20Water%20Quality%20Criteria%20for%20the%20ASEAN%20Region.pdf>>/>.
- Azanza, R.V., Benico, G.A. (2013) Toxic *Alexandrium* blooms in Fish Farming Sites in Bolinao, Pangasinan. *Journal of Environmental Science and Management Special Issue 1-2013*, pp.44-49

- Azanza, R.V., Baula, I.U., Fukuyo, Y. (2006) Seasonal changes in phytoplankton composition in an extensive fish culture area in Bolinao, Pangasinan, northern Philippines. *Coastal Marine Science* Vol. 30, pp.85-87
- Azanza, R.V., Fukuyo, Y., Yap, L.G., Takayama, H. (2005) *Prorocentrum minimum* bloom and its possible link to a massive fish kill in Bolinao, Pangasinan, Northern Philippines. *Harmful Algae* Vol. 4, pp.519-524
- Benitez-Nelson, C.R. (2000) The biogeochemical cycling of phosphorus in marine systems. *Earth-Sci Rev* Vol. 51, pp.109-135
- Borlongan, I.G., Satoh, S. (2001) Dietary phosphorus requirement of juvenile milkfish, *Chanos chanos* (Forsskal). *Aquac Res* Vol. 32(Suppl. 1), pp. 26-32
- Brock, J., Schulz-Vogt, H.N. (2011) Sulfide induces phosphate release from polyphosphate in cultures of a marine *Beggiatoa* strain. *ISME J* Vol. 5, pp.497-506
- Cardenas MB, Zamora PB, Siringan FP, Lapus MR, Rodolfo RS, Jacinto GS, San Diego-McGlone ML, Villanoy CL, Cabrera O, Senal MS (2010) Linking regional sources and pathways for submarine groundwater discharge at a reef by electrical resistivity tomography, ^{222}Rn , and salinity measurements. *Geophys Res Lett* Vol. 37 [doi:10.1029/2010GL044066]
- Cruz-Trinidad, A., Geronimo, R., Cabral, R.B., Aliño, P.M. (2011) How much are the Bolinao-Anda coral reefs worth? *Ocean and Coastal Management* Vol. 54, pp.696-705
- Dale, A.W., Bertics, V.J., Treude, T., Sommer, S., Wallmann, K. (2013) Modeling benthic-pelagic nutrient exchange processes and porewater distributions in a seasonally hypoxic sediment: evidence for massive phosphate release by *Beggiatoa*? *Biogeosciences* Vol. 10, pp.629-651
- De Simone, L.A., Howes, B.L. (1998) Nitrogen transport and transformations in a shallow aquifer receiving wastewater discharge: a mass balance approach. *Water Resour Res* Vol. 34(2), pp.271-285
- Escobar, M.T.L., Sotto, L.P.A., Jacinto, G.S., Benico, G.A., San Diego-McGlone, M.L., Azanza, R.V. (2013) Eutrophic conditions during the 2010 fish kill in Bolinao and Anda, Pangasinan, Philippines. *Journal of Environmental Science and Management* Special Issue 1-2013, pp.29-35
- FAO (2009) Analysis of aquaculture development in Southeast Asia. FAO Fisheries and Aquaculture Technical Paper No. 509, Rome.
- Folke, C., Kautsky, N., Troell, M. (1994) The Costs of Eutrophication from Salmon Farming: Implications for Policy. *J Environ Manage* Vol. 40, pp.173-182
- Fortes, M.D., Nadaoka, K. (2015) *Guidebook: The Coastal Ecosystem Conservation and Adaptive Management (CECAM) Approach as an Innovation to Existing ICZM Frameworks*, Japan International Cooperation Agency, Japan Science and Technology Agency- Science and Technology Research Partnership for Sustainable Development Program, Commission on Higher Education (Philippines). http://www.cecamm-project.net/wp-content/uploads/2011/03/Updated-draft-of-guidebook_ver3-2_ready-for-printing_FINAL3edit1.pdf
- Grasshoff, K., Kremling, K., Ehrhardt, M. (1999) *Method of seawater analysis*, Wiley-VCH, Weinheim.
- Hasler, A.D. (1969) Cultural Eutrophication Is Reversible. *BioScience* Vol. 19, pp.425-431
- Herbeck, L.S., Unger, D., Wu, Y., Jennerjahn, T.C. (2013) Effluent, nutrient and organic matter export from shrimp and fish ponds causing eutrophication in coastal and back-reef waters of NE Hainan, tropical China. *Cont Shelf Res* Vol. 57, pp.92-104
- Ho, A.Y.T., Xu, J., Yin, K., Jiang, Y., Yuan, X., He, L., Anderson, D.M., Lee, J.H.W., Harrison, P.J. (2010) Phytoplankton Biomass and Production in Subtropical Hong

- Kong Waters: Influence of the Pearl River Outflow. *Estuaries and Coasts* Vol. 33, pp.170- 81
- Holby, O., Hall, P.O.J. (1991) Chemical fluxes and mass balances in a marine fish cage farm. II. Phosphorus. *Mar Ecol Prog Ser* Vol. 70, pp.263-272
- Holmer, M. Kristensenm, E. (1992) Impact of marine cage farming on sediment metabolism and sulfate reduction of underlying sediments. *Mar Ecol Prog Ser* Vol 80., pp.191-201
- Holmer, M., Marba, N., Terrados, J., Duarte, C.M., Fortes, M.D. (2002) Impacts of milkfish (*Chanos chanos*) aquaculture on carbon and nutrient fluxes in the Bolinao area, Philippines. *Mar Pollut Bull* Vol. 44, pp.685-696
- Holmer, M., Duarte, C.M., Heilskov, A., Olesen, B., Terrados, J. (2003) Biogeochemical conditions in sediments enriched by organic matter from net-pen fish farms in the Bolinao area, Philippines. *Mar Pollut Bull* Vol. 46, pp.1470-1479
- Kemp, W.M., Testa, J.M., Conley, D.J., Gilbert, D., Hagy, J.D. (2009) Temporal responses of coastal hypoxia to nutrient loading and physical controls. *Biogeosciences* Vol. 6, pp.2985-3008
- Kibria, G., Nugegoda, D., Lam, P., Fairclough, R. (1996) Aspects of Phosphorus Pollution from Aquaculture. *Naga, The ICLARM Quarterly* Vol. 19, pp.20-24
- Lauer, P.R., Fernandes, M., Fairweather, P.G., Tanner, J., Cheshire, A. (2009) Benthic fluxes of nitrogen and phosphorus at southern bluefin tuna *Thunnus maccoyii* sea-cages. *Mar Ecol Prog Ser* Vol. 390, pp.251-263
- Martin, W.R. (2009) Chemical processes in estuarine sediments, in Steele, J.H. et al (Eds.), *Encyclopedia of Ocean Sciences Online*, Academic Press, pp. 539-550.
- Mackenzie, F.T., De Carlo, E.H., Lerman, A. (2011) Coupled C, N, P, and O Biogeochemical Cycling at the Land-Ocean Interface, in Wolanski, E. and McLusky, D.S. (Eds), *Treatise on Estuarine and Coastal Science Vol 5*, Waltham: Academic Press, pp. 317-342.
- Magdaong, T., Villanoy, C.L. (2008) Determination of Properties of Milkfish Feed for Model Development. *UPV J Nat Sci* 13 Suppl, pp.116-122
- Middleburg, J.J., Levin, L.A. (2009) Coastal hypoxia and sediment biogeochemistry. *Biogeosciences* Vol. 6, pp.1273-1293
- Mulholland, P.J. (1992) Regulation of nutrient concentrations in a temperate forest stream: roles of upland, riparian, and instream processes. *Limnol Oceanogr* Vol. 37, pp.1512-1526
- Nacorda, H.M.E., Obliosca, J.M., Tentia, M.C.L., Jacinto, G.S., San Diego-McGlone, M.L.C. (2012) Deterioration of Soft Bottom Macroinfaunal Communities in a Milkfish Mariculture Zone off Bolinao-Anda, Pangasinan (NW Philippines), in Kawaguchi, M. et al. (eds), *Interdisciplinary Studies on Environmental Chemistry – Environmental Pollution and Ecotoxicology*, TERRAPUB, pp. 387-395.
- Nixon, S.W. (1995) Coastal marine eutrophication: A definition, social causes, and future concerns. *Ophelia* Vol. 41, pp.199-219
- Obliosca, J.M., Jacinto, G.S., San Diego-McGlone, M.L. (2003) Preliminary results on the development of marine environmental quality criteria for mariculture areas. *Philipp Scient* Vol. 40, pp.73-87
- Olsen, L.M., Holmer. M., Olsen, Y. (2008) Perspectives of nutrient emission from fish aquaculture in coastal waters: Literature review with evaluated state of knowledge. The Fishery and Aquaculture Industry Research Fund (FHF) project no. 542014: Final report.

- Onitsuka, G., Aoki, K., Shimizu, M. (2015) Meteorological conditions preceding *Chattonella* bloom events in the Yatsushiro Sea, Japan, and possible links with the East Asian monsoon. *Fish Sci* Vol. 81, pp.123-130
- Paytan, A., McLaughlin, K. (2007) The Oceanic Phosphorus Cycle. *Chem Rev* Vol. 107, pp.563-576
- Reichardt, W.T., Reyes, J.M., Pueblos, M.J., Lluisma, A.O. (2013) Impact of milk fish farming in the tropics on potentially pathogenic vibrios. *Mar Pollut Bull* Vol. 77, pp.325-332
- Ruttenberg, K.C. (2003) The Global Phosphorus Cycle, in Holland, H.D. and Turekian, K.K. (Eds) *Treatise on Geochemistry* Vol 8, Elsevier Ltd., pp. 585-643.
- San Diego-McGlone, M.L., Azanza, R.V., Villanoy, C.L., Jacinto, G.S. (2008) Eutrophic waters, algal bloom and fish kill in fish farming areas in Bolinao, Pangasinan, Philippines. *Mar Pollut Bull* Vol. 57, pp.295-301
- San Diego-McGlone, M.L., Lagumen, M.C., Escobar, M.T., Benico, G., Rivera, P.P., Azanza, R.V. (2014) Eutrophication, algal blooms, and fish kills: A continuing story (1995-2013). 2nd National Conference and Workshop of the Philippines-Japan Collaborative Project (NCW2), UP Marine Science Institute, Diliman, Quezon City, 17-18 June 2014.
- Santander-De Leon, S.M.S., San Diego-McGlone, M.L., Reichardt, W. (2008) Indicators of Diminished Organic Matter Degradation Potential of Polychaete Burrows in Philippine Mariculture Areas. *The Philippine Agricultural Scientist* Vol. 91, pp.295-300
- Smith, V.H., Schindler, D.W. (2009) Eutrophication science: where do we go from here? *Trends in Ecology and Evolution* Vol. 24(4), pp.201-205
- Smith, V.H., Tilman, G.D., Nekola, J.C. (1999) Eutrophication: impacts of excess nutrient inputs on freshwater, marine, and terrestrial ecosystems. *Environmental Pollution* Vol. 100, pp.179-196
- Slomp, C.P. (2011) Phosphorus Cycling in the Estuarine and Coastal Zones: Sources, Sinks, and Transformations, in Wolanski, E. and McLusky, D.S. (Eds) *Treatise on Estuarine and Coastal Science* Vol 5, Waltham: Academic Press, pp.201-229.
- Sumagaysay-Chavoso N.S. (2003) Nitrogen and phosphorus digestibility and excretion of different-sized groups of milkfish (*Chanos chanos* Forsskal) fed formulated and natural food-based diets. *Aquac Res* Vol. 34, 407-418
- Suzuki, R., Ishimaru, T. (1990) An improved method for the determination of phytoplankton chlorophyll using n,n-dimethylformamide. *J Oceanogr* Vol. 46, pp.190-194 [doi:10.1007/BF02125580]
- Suzumura, M. (2008) Persulfate chemical wet oxidation method for the determination of particulate phosphorus in comparison with a high-temperature dry combustion method. *Limnol Oceanogr-Meth* Vol. 6, pp.619-629
- Tanaka, Y., Go, G.A., Watanabe, A., Miyajima, T., Nakaoka, M., Uy, W.H., Nadaoka, K., Watanabe, S., Fortes, M.D. (2014) 17-year change in species composition of mixed seagrass beds around Santiago Island, Bolinao, the northwestern Philippines. *Mar Pollut Bull* Vol.88, pp.81-85
- Umezawa, Y., Suzumura, M., Tsukasaki, A., Ozaki, T., Muta, N., Yamaguchi, A. (2015) Biogeochemical phosphorus cycle in coastal environments. *Chikyu-Kankyo*, Vol.20(1), pp. 63-76 (in Japanese).
- Verceles, L.F., Talaue-McManus, L., Aliño, P.M. (2000) Participatory Monitoring and Feedback System: An Important Entry Towards Sustainable Aquaculture in Bolinao, Northern Philippines. *Science Diliman* Vol. 12(2), pp.78-87

- Villanueva, R.D., Yap, H.T., Montano, M.T.E. (2006) Intensive fish farming in the Philippines is detrimental to the reef-building coral *Pocillopora damicornis*. *Mar Ecol Prog Ser* Vol. 316, pp.165-174
- White, P. (2013) Environmental consequences of poor feed quality and feed management, in: Hasan, M. R., New, M. B. (Eds.) On-farm feeding and feed management in aquaculture. FAO Fisheries and Aquaculture Technical Paper No. 583, Rome, FAO, pp.553-564.
- Wu, R.S.S. (1995) The Environmental Impact of Marine Fish Culture: Towards a Sustainable Future. *Mar Pollut Bull* Vol. 31, pp.159-166
- Yap, L.G., Azanza, R.V., Talaue-McManus, L. (2004) The community composition and production of phytoplankton in fish pens of Cape Bolinao, Pangasinan: a field study. *Mar Pollut Bull* Vol. 49, pp.819-832

Figure Captions

Fig. 1 The Bolinao-Anda coastal waters – (a) water sampling stations for end member sources of nutrients to the mariculture area, (b) location of fish structures based on Google Earth image of January 2014, and (c) water and core sampling stations along the Guiguivanen Channel. Bathymetry data are from GEBCO 08. Depth contours are in meters.

Fig. 2 Long-term data on environmental parameters (rainfall, wind), nutrients, chlorophyll-*a* and fish farm structures in Bolinao. Rainfall and wind data are from a station ~170km southeast of Bolinao (Subic monitoring site of the Philippine Atmospheric, Geophysical and Astronomical Services Administration-PAGASA). Nutrients and chlorophyll-*a* are from San Diego-McGlone et al. (2008, 2014). The numbers of fish structures in Bolinao are from San Diego-McGlone et al. (2008, 2014) and based on issued permits and ocular surveys by the Bolinao Municipal Agricultural Office. Rainfall is expressed as cumulative data per season and nutrients and chlorophyll-*a* as average values. Nutrients and chlorophyll-*a* were collected seasonally in 1995-2001, and daily from 2002-2013. Error bars represent standard error from mean values.

Fig. 3 Spatial distribution of DIN, DIP, DIN/DIP and salinity at the Bolinao-Anda coastal waters. Data are average values from the spatial and 24-hr surveys in Sep 2010-2014, and in Mar 2011, 2013, and 2014. Parameters in a station data set without available information are represented by the section symbol (§). Filled bars for DIP, DIN/DIP and salinity indicate values greater than the reference scale. Actual values of parameters are given in Tables S2 and S3 of the Supplementary Material. Site locations within the mariculture areas are indicated by specific station names (B3, B4, B5, T1, ALA1).

Fig. 4 Diurnal variation of DIP, DOP, PP, tide, flow velocity at the bottom layer and DO at CCMS station during the wet and dry seasons. Black (gray) lines in (e) and (f) represent the magnitude of the north-south (east-west) component of horizontal velocity. Positive horizontal velocity values represent north (east) direction of the flow while negative values

correspond to south (west) direction of the flow. Shaded areas indicate the times when peaks in bottom water DIP were observed.

Fig. 5 Sediment porewater DIN and DIP at the West Station, Mid-channel and East Station in the Guiguivanen Channel for cores collected in 2011. Data from CCMS Sep 15 are from two cores.

Fig. 6 DIN/DIP ratios and relationship of DIP with DO in the mariculture area. Data are from the spatial, grid and 24-hr surveys in Sep 2010-2014, and in Mar 2011, 2013, and 2014. Slopes of the lines in (a) and (b) indicate DIN/DIP ratios (16 = Redfield, 10.8 = porewater, 6.6 = bottom water, 4.4 = ratio calculated from nutrient release rates during decomposition of feeds). Shaded areas in (c) and (d) are hypoxic ($<62.5 \mu\text{mol kg}^{-1}$) to anoxic conditions. Data denoted by arrows in (c) and enclosed by broken line in (d) are from the station at Caquiputan Strait. Inset plots in (c) are the vertical profiles of σ_t for wet (Sep 2012) and dry (Mar 2013) season.

Fig. 7 Temporal change in fish structures in the Bolinao-Anda coastal waters. Fish cages and fish pens were manually counted from Google Earth (Nov 2003 and Jan 2014) and World View-2 (Mar 2010). The numbers of fish farm structures counted for Bolinao are similar to those in Fig. 2c.

Fig. 8 Calculated monthly variation of long-term data on environmental parameters (solar radiation, rainfall), nutrients and chlorophyll-*a* in Bolinao. Solar radiation data are from a station ~220km southeast of Bolinao (PAGASA's National Solar Radiation Center in Quezon City). Rainfall and wind data are from a station ~170 km southeast of Bolinao (PAGASA's Subic monitoring site). Nutrients and chlorophyll-*a* are from the monitoring data by San Diego-McGlone et al. (2008; 2014). Solar radiation is expressed as monthly average of the total hourly average from 2011-2014. Rainfall and wind data are expressed as monthly average from 2002-2013. Nutrient and chlorophyll-*a* data are calculated as monthly average from 2002-2013. Error bars represent standard deviation from mean values.

Tables

Table 1 End member values of DIN, DIP, N/P ratio, and salinity of the sources of nutrients to the mariculture area. In parentheses are number of samples unless otherwise stated.

End Member	DIN, μM	DIP, μM	N/P	Salinity
Station, Season	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.
Offshore water				
South China Sea (surface)				
<i>Wet</i>	0.27 $\pm$ 0.06 (2)	0.17 $\pm$ 0.14 (2)	7.72 $\pm$ 7.05 (2)	30.8 $\pm$ 0.3 (2)
<i>Dry</i>	0.25 $\pm$ 0.07 (2)	0.07 $\pm$ 0.02 (2)	4.36 $\pm$ 2.34 (2)	33.0 $\pm$ 0.0 (2)
Lingayen Gulf (surface)				
<i>Wet</i>	0.48 $\pm$ 0.90 (6)	0.12 $\pm$ 0.19 (6)	2.81 $\pm$ 1.49 (6)	28.9 $\pm$ 5.5 (6)
<i>Dry</i>	0.09 $\pm$ 0.03 (7)	0.05 $\pm$ 0.04 (6)	4.20 $\pm$ 4.12 (7)	33.2 $\pm$ 0.3 (7)
Rivers				
Bani				
<i>Wet</i>	16.8 $\pm$ 5.1 (6)	0.94 $\pm$ 0.50 (6)	28.2 $\pm$ 25.4 (6)	0.15 $\pm$ 0.03 (5)
<i>Dry</i>	9.62 $\pm$ 8.47 (2)	2.68 $\pm$ 2.24 (2)	3.13 $\pm$ 0.54 (2)	16.3 $\pm$ 11.6 (3)
Alaminos				
<i>Wet</i>	12.5 $\pm$ 2.6 (3)	1.32 $\pm$ 0.56 (3)	11.1 $\pm$ 3.9 (3)	0.07 $\pm$ 0.00 (2)
<i>Dry</i>	21.5 $\pm$ 5.0 (2)	0.10 $\pm$ 0.04 (2)	235 $\pm$ 33 (2)	14.4 (1)
Other rivers				
<i>Wet</i>	16.2 $\pm$ 10.0 (2)	0.43 $\pm$ 0.07 (2)	41.9 $\pm$ 29.6 (2)	0.43 (1)
Groundwater				
<i>Wet</i>	245 (159-417)**	0.28 (0.19-0.63)**	656 (466-1105)**	0.54 $\pm$ 0.32 (12)
<i>Dry</i>	194 (121-358)**	0.24 (0.16-0.44)**	772 (289-1192)**	0.62 $\pm$ 0.35 (12)
Sediment porewater				
upper 10cm	1.33 $\times 10^3 \pm 5.40 \times 10^2$ (10)	128 $\pm$ 33 (10)	10.8 $\pm$ 4.7 (10)	---
Fish feed				
particulate	---	---	6.68-12.2 ^a	---
dissolved	---	---	4.41 ^b	---
Fish excreta				
particulate (feces)	---	---	5.40 $\pm$ 0.21 ^c	---
dissolved (excreta)	---	---	6.22 $\pm$ 0.32 ^c	---

** DIN, DIP and N/P ratios for groundwater are median values instead of mean. Values in parentheses are the interquartile range. Number of samples for these parameters is same as the number of samples for salinity (12).

^a calculated from reported N and P concentration of the different types of fish feeds used in Bolinao (Holmer et al., 2002); N:P ratio varies according to feed type

^b calculated from release rates of NH_4^+ and PO_4^{3-} upon decomposition of milkfish feeds used in Bolinao (Obliosca et al., 2003)

^c calculated from reported fecal N and P excreted by different sizes of milkfish fed with natural food-based, formulated, and commercial feeds (Sumagaysay-Chavoso, 2003)

Table 2 Average values of DIN, DIP, DOP, PP, DIN/DIP, salinity and DO during the short-term (24-h) temporal survey at the Guiguiwanen Channel. In parentheses are number of samples. (“*h*” indicates sampling depth)

Station	DIN, μM	DIP, μM	DOP, μM	PP, μM	DIN/DIP	Salinity	DO, mg L^{-1}
Season, Depth	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.	Mean $\pm$ S.D.
West Station (Sta 1)							
Wet (2011/09)							
surface (<i>h</i> =0 m)	0.84 ± 0.94 (9)	0.74 ± 0.09 (9)	0.55 ± 0.14 (9)	1.74 ± 0.36 (4)	1.04 ± 1.05 (9)	19.6 ± 0.3 (9)	7.28 ± 1.00 (9)
middle (<i>h</i> =10.5 m)	2.75 ± 1.00 (9)	0.31 ± 0.13 (9)	0.10 ± 0.04 (9)	0.12 ± 0.02 (4)	9.22 ± 1.80 (9)	31.0 ± 0.1 (9)	5.36 ± 0.44 (9)
bottom (<i>h</i> =20 m)	9.23 ± 1.11 (9)	1.09 ± 0.14 (9)	0.19 ± 0.09 (6)	0.22 ± 0.04 (4)	8.46 ± 0.67 (9)	31.9 ± 0.1 (9)	3.24 ± 0.71 (9)
CCMS (Mid-channel)							
Wet (2011/09)							
surface (<i>h</i> =0m)	8.00 ± 1.61 (13)	1.49 ± 0.11 (13)	0.30 ± 0.10 (13)	1.27 ± 0.31 (9)	5.36 ± 0.96 (13)	19.2 ± 0.7 (10)	6.14 ± 0.25 (10)
bottom (<i>h</i> =13 m)	6.46 ± 1.71 (13)	0.85 ± 0.24 (13)	0.06 ± 0.02 (6)	0.24 ± 0.08 (9)	7.63 ± 0.55 (13)	31.4 ± 0.1 (11)	3.53 ± 0.66 (11)
Dry (2012/03)							
surface (<i>h</i> =0 m)	5.82 ± 1.42 (13)	1.75 ± 0.18 (13)	0.51 ± 0.11 (13)	0.75 ± 0.25 (9)	3.32 ± 0.75 (13)	33.3 ± 0.0 (12)	5.19 ± 0.58 (12)
bottom (<i>h</i> =13.5 m)	5.53 ± 2.04 (13)	0.89 ± 0.44 (13)	0.22 ± 0.21 (12)	0.46 ± 0.23 (9)	6.69 ± 1.14 (13)	33.1 ± 0.1 (12)	4.73 ± 0.67 (12)
East Station (Sta 2)							
Wet (2011/09)							
surface (<i>h</i> =0 m)	0.54 ± 0.82 (9)	0.68 ± 0.15 (9)	0.47 ± 0.08 (9)	1.56 ± 0.17 (4)	0.65 ± 0.78 (9)	18.7 ± 0.7 (9)	7.84 ± 1.12 (9)
middle (<i>h</i> =8 m)	9.43 ± 1.04 (9)	1.29 ± 0.17 (9)	0.11 ± 0.06 (5)	0.22 ± 0.01 (4)	7.36 ± 0.29 (9)	31.0 ± 0.2 (9)	2.87 ± 0.53 (9)
bottom (<i>h</i> =13 m)	10.7 ± 1.9 (9)	1.49 ± 0.19 (9)	0.08 ± 0.04 (4)	0.22 ± 0.04 (4)	7.20 ± 0.50 (9)	31.2 ± 0.2 (9)	2.46 ± 0.51 (9)

Figures

Fig. 1 The Bolinao-Anda coastal waters – (a) water sampling stations for end member sources of nutrients to the mariculture area, (b) location of fish structures based on Google Earth image of January 2014, and (c) water and core sampling stations along the Guiguivanen Channel. Bathymetry data are from GEBCO 08. Depth contours are in meters.

Fig. 2 Long-term data on environmental parameters (rainfall, wind), nutrients, chlorophyll-*a* and fish farm structures in Bolinao. Rainfall and wind data are from a station ~170km southeast of Bolinao (Subic monitoring site of the Philippine Atmospheric, Geophysical and Astronomical Services Administration-PAGASA). Nutrients and chlorophyll-*a* are from San Diego-McGlone et al. (2008, 2014). The numbers of fish structures in Bolinao are from San Diego-McGlone et al. (2008, 2014) and based on issued permits and ocular surveys by the Bolinao Municipal Agricultural Office. Rainfall is expressed as cumulative data per season and nutrients and chlorophyll-*a* as average values. Nutrients and chlorophyll-*a* were collected seasonally in 1995-2001, and daily from 2002-2013. Error bars represent standard error from mean values.

Fig. 3 Spatial distribution of DIN, DIP, DIN/DIP and salinity at the Bolinao-Anda coastal waters. Data are average values from the spatial and 24-hr surveys in Sep 2010-2014, and in Mar 2011, 2013, and 2014. Parameters in a station data set without available information are represented by the section symbol (§). Filled bars for DIP, DIN/DIP and salinity indicate values greater than the reference scale. Actual values of parameters are given in Tables S2 and S3 of the Supplementary Material. Site locations within the mariculture areas are indicated by specific station names (B3, B4, B5, T1, ALA1).

Fig. 4 Diurnal variation of DIP, DOP, PP, tide, flow velocity at the bottom layer and DO at CCMS station during the wet and dry seasons. Black (gray) lines in (e) and (f) represent the magnitude of the north-south (east-west) component of horizontal velocity. Positive horizontal velocity values represent north (east) direction of the flow while negative values correspond to south (west) direction of the flow. Shaded areas indicate the times when peaks in bottom water DIP were observed.

Fig. 5 Sediment porewater DIN and DIP at the West Station, Mid-channel and East Station in the Guiguiwanen Channel for cores collected in 2011. Data from CCMS Sep 15 are from two cores.

Fig. 6 DIN/DIP ratios and relationship of DIP with DO in the mariculture area. Data are from the spatial, grid and 24-hr surveys in Sep 2010-2014, and in Mar 2011, 2013, and 2014. Slopes of the lines in (a) and (b) indicate DIN/DIP ratios (16 = Redfield, 10.8 = porewater, 6.6 = bottom water, 4.4 = ratio calculated from nutrient release rates during decomposition of feeds). Shaded areas in (c) and (d) are hypoxic (<62.5 $\mu\text{mol kg}^{-1}$) to anoxic conditions. Data denoted by arrows in (c) and enclosed by broken line in (d) are from the station at Caquiputan Strait. Inset plots in (c) are the vertical profiles of σ_t for wet (Sep 2012) and dry (Mar 2013) season.

Fig. 7 Temporal change in fish structures in the Bolinao-Anda coastal waters. Fish cages and fish pens were manually counted from Google Earth (Nov 2003 and Jan 2014) and World View-2 (Mar 2010). The numbers of fish farm structures counted for Bolinao are similar to those in Fig. 2c.

Fig. 8 Calculated monthly variation of long-term data on environmental parameters (solar radiation, rainfall), nutrients and chlorophyll-*a* in Bolinao. Solar radiation data are from a station ~220km southeast of Bolinao (PAGASA's National Solar Radiation Center in Quezon City). Rainfall and wind data are from a station ~170 km southeast of Bolinao (PAGASA's Subic monitoring site). Nutrients and chlorophyll-*a* are from the monitoring data by San Diego-McGlone et al. (2008; 2014). Solar radiation is expressed as monthly average of the total hourly average from 2011-2014. Rainfall and wind data are expressed as monthly average from 2002-2013. Nutrient and chlorophyll-*a* data are calculated as monthly average from 2002-2013. Error bars represent standard deviation from mean values.