Water Policy 20 (2018) 855–870

Historical changes in primary production in the Seto Inland Sea, Japan, after implementing regulations to control the pollutant loads

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Abstract

A total pollutant load control system (TPLCS) was implemented in the Seto Inland Sea in 1979 to reduce the water pollution and the frequency of red tides. We estimated primary production from 1981 to 2010 to determine the effects of reducing the nutrient loadings from the surrounding land. While primary production has decreased overall in the Seto Inland Sea in response to the TPLCS and the associated reductions in the total nitrogen (T-N) and phosphorus (T-P) loads from land since 1981, the reductions were limited to 4 of its 11 subareas. Primary production has increased in the Harima Nada but has been stable in the Bingo Nada subarea, reflecting the fact that the T-N and T-P stocks have not decreased in these subareas over the study years. The inconsistent responses of the 11 subareas suggest that the characteristics of each subarea should be considered when environmental management measures are established and implemented in the Seto Inland Sea. The controls on the nutrient loadings according to the TPLCS should be modified to permit better management of this semi-enclosed sea.

Keywords: Control of nutrient loadings; Historical change; Primary production; Seto Inland Sea and its subareas; Total pollutant load control system

Introduction

The Seto Inland Sea is in the western part of Japan. It is the 11th largest semi-enclosed sea in the world and, as shown in Figure 1, is divided into 11 subareas. Human activities in the area, such as

doi: 10.2166/wp.2018.093

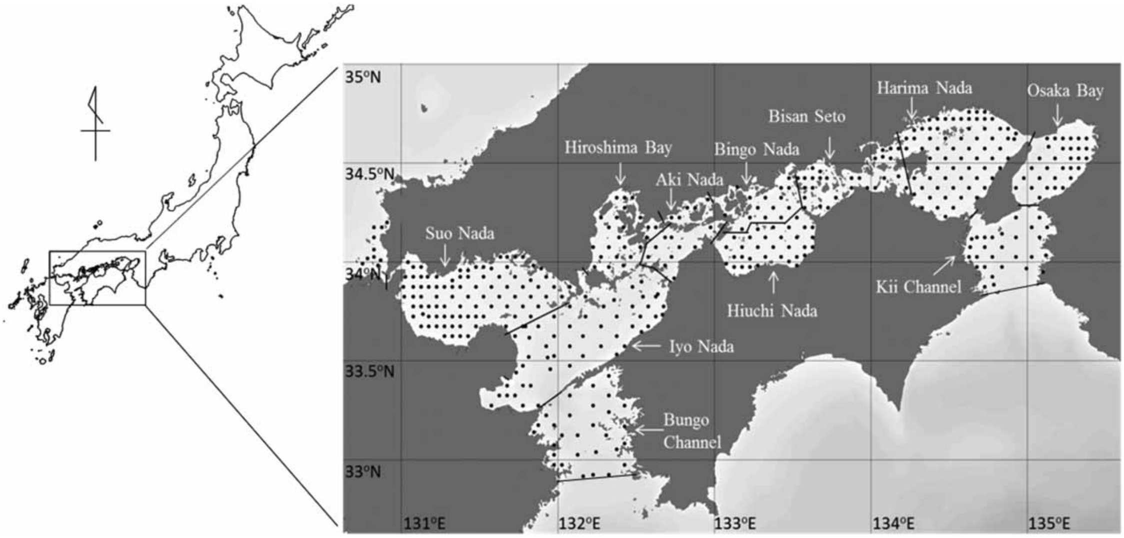

Fig. 1. Seto Inland Sea and its 11 subareas. The border lines were defined by the Voronoi tessellation. The dots in the subareas indicate the CWQS monitoring stations of the MOE, Japan.

industrialization and urbanization, resulted in severe eutrophication, water pollution, and outbreaks of algae including red tides during the 1950s–1970s. In response, the Water Pollution Control Law (1970) and the Law Concerning Special Measures for Conservation of the Environment of the Seto Inland Sea (1973) were enacted, and a total pollutant load control system (TPLCS) has been employed seven times since 1979. Under this system, the loadings of chemical oxygen demand and nutrients such as nitrogen and phosphorus from the land surrounding the Seto Inland Sea must decrease by scheduled amounts every five years (Tezuka, 2009).

At the same time as the TPLCS was implemented, the Environment Agency of Japan (the former Ministry of Environment (MOE)) initiated a comprehensive water quality survey (CWQS) in the Seto Inland Sea in 1972. Results from the TPLCS show that the total nitrogen (T-N) and phosphorus (T-P) loadings have decreased, and over the decade from 1999 the T-N and T-P loadings into the Seto Inland Sea were 73% and 69% of the loads in 1999, respectively (Fujiwara, 2013). Also, red tides in the Seto Inland Sea, which were observed 299 times/y in 1967, were only observed about 100 times/y in the second half of the 1980s (Imai *et al.*, 2006). On the other hand, other problems have emerged. For example, discoloration of Nori and Wakame seaweed has been reported, possibly because of oligotrophication (Yamamoto, 2003; Fujiwara, 2013); also, the annual fishery landings in the Seto Inland Sea have decreased since 1980, and in 2014 were about one-third of what they were in 1980 (Fisheries Agency of Japan, 2016). The facts indicate that the TPLCS needs to be reviewed.

Because of how food webs function, increases and decreases in primary producers such as phytoplankton have direct and indirect effects on fishery landings in coastal areas (Yamamoto, 2003; Philippart *et al.*, 2007; de Mutsert *et al.*, 2016). Since nitrogen and phosphorus are essential nutrients for phytoplankton growth, we hypothesize that the decrease in fishery landings in the Seto Inland Sea may reflect the reduction in nutrient loadings from the surrounding land. Primary production in the Seto Inland Sea was estimated once for 1993 and 1994 (Hashimoto *et al.*, 1997; Tada *et al.*, 1998), but has not been done in recent years because it is not included in either the CWQS or other survey programs of the Japanese Government. In the previous studies investigating the historical changes of primary production, chlorophyll *a* concentrations were used for the estimation (Wetsteyn & Kromkamp, 1994; Philippart *et al*., 2007; Marchese *et al*., 2015), while in the recent one the satellite data were used to determine chlorophyll *a* concentrations (Marchese *et al*., 2015). In the CWQS, chlorophyll *a* concentrations were fortunately measured and available.

It is important however to remember that the sources of nutrients to the Seto Inland Sea are not limited to the surrounding land. In fact, Yanagi & Ishii (2004) reported that the ratios of the land-originated and ocean-originated nitrogen and phosphorus loadings to the Seto Inland Sea were 19:81 and 7:18, respectively, indicating that, while the contributions in each subarea may differ, nutrients originating from the open ocean are the main source for the entire Seto Inland Sea rather than land-originated nutrients. In addition, outer coastal areas may respond less sensitively to the reduction of land-originated nutrient loadings than inner coastal areas (Savchuk *et al*., 2009; Staehr *et al*., 2017). Therefore, questions about whether primary production has decreased in the Seto Inland Sea and how the reductions in the land-originated nutrient loads to the sea impact productivity remain unanswered.

As shown in the schematic view of this study (Figure S1, available with the online version of this paper), we constructed a simple model based on the CWQS dataset and information collected in a seasonal survey of Hiroshima Bay that would allow us to estimate phytoplankton primary production in recent years in the Seto Inland Sea. After validating the model, we investigated whether phytoplankton primary production had decreased across all of the Seto Inland Sea and in its 11 subareas in line with the reductions in loadings of land-originated nutrients required by the TPLCS.

Methods

Outline of the seasonal survey

Most existing mathematical models for estimating primary production by phytoplankton require information on physicochemical and biological parameters, such as light intensity and chlorophyll *a* concentrations (e.g., Hashimoto & Takeoka, 1998; Maar *et al*., 2016). Unfortunately, some of the required parameters, such as the light-reachable depth, are not included in the CWQS, and transparency measured with a Secchi disk is used for apparent turbidity. Therefore, we included water quality in our survey of Hiroshima Bay and examined the relationship between the visible light attenuation coefficient (K_d) and transparency.

Figure 2 shows the monitoring stations in the seven subsections of Hiroshima Bay, determined from the Voronoi tessellation division. Four of the seven stations overlap with those used in the CWQS by the MOE (Table S1, available with the online version of this paper). We did our survey in November 2014 (autumn), February 2015 (winter), May 2015 (spring), and August 2015 (summer).

Measurement of water quality, light attenuation, and primary production

At the seven stations, we measured seawater temperature, photosynthetically active radiation (PAR), and the depth with a probe (AAQ-RINKO, JFE Advantec, Japan) and determined K_d from the

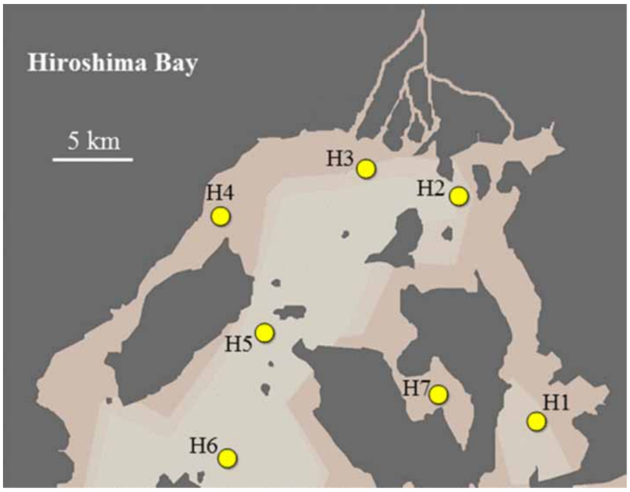

Fig. 2. Seven sampling stations in Hiroshima Bay (H1–7).

attenuation of PAR. We also measured the Secchi depth with a disk with a diameter of 30-cm to determine the depth at which the disk was no longer visible. We collected 4 L samples of sea water at depths where the irradiance was 100, 50, and 10% of that at the surface and analyzed them for nutrients such as dissolved inorganic nitrogen and phosphorus (DIN and DIP), chlorophyll *a* (Chl.*a*), and primary production. Concentrations of DIN and DIP were determined colorimetrically using an autoanalyzer (SWAAT, BLTEC, Japan). Consistent with the CWQS of the MOE, we measured Chl.*a* by the Welschmeyer method (Welschmeyer, 1994), as follows. Three hundred mL of the seawater sample was passed through a glass-fiber filter (GF/F, Whatman, UK), and the residue was extracted with 10 mL of 90% acetone in dark conditions at −20°C for between 16 and 24 h. After sonicating for 20 min, the extract was analyzed fluorophotometrically (10-AU-5, Turner Designs, USA). Prior to measuring the primary production, 200 mL of the seawater sample was filtered through a 220 μm mesh screen to remove large zooplankton. The filtrate was transferred to a 500 mL polycarbonate bottle and the bottles were left to acclimatize to the dark conditions for 30 min. After adding 0.2 mM $NaH^{13}CO_3$ to each bottle, the bottles were immediately incubated in a culture tank at the *in situ* temperature under light intensities of 200-, 100-, and 20-μmol photons m^{-2} sec^{-1} for 4 h. The incubated samples were filtered through pre-combusted GF/F filters (450°C for 4 h), and the residue was treated with 1N HCl to remove inorganic carbon and stored at −20°C until isotope analysis. Finally, the filter was dried at 60°C for 48 h, and the particulate organic carbon (POC) concentration and atom % of ^{13}C in the residue were determined by an elemental analyzer coupled with an isotope ratio mass spectrometer (FlashA EA1112-dDelta V Advantage, Thermo Fisher Scientific, USA). The photosynthetic rate was calculated with the method of Hama *et al*. (1983).

Estimation of primary production

We estimated the phytoplankton photosynthetic rate, *P*r [mg-C m^{-3} h^{-1}], from the Chl.*a* specific productivity, *P*c [mg-C mg-Chl.a^{-1} h^{-1}] (Wetsteyn & Kromkamp, 1994; Bode & Varela, 1998), and Chl.*a* concentration, CHL [mg-Chl.a m^{-3}], with Equation (1):

$$Pr = Pc \times CHL \tag{1}$$

Primary production in the euphotic zone of each station was determined by double integration of the Chl.a specific productivity at the ranges of depth and time (Lawrenz *et al.*, 2013; Marchese *et al.*, 2015) from the surface to where the irradiance is 1% of that at the surface and from sunrise to sunset with Equation (2):

$$PP_{\mathrm{stn}} = \iint [Pc(z, t) \times CHL(z)]dzdt \tag{2}$$

where PP_{stn} is the primary production in the euphotic zone at each station [mg-C m^{-2} d^{-1}]; $Pc(z, t)$ and $CHL(z)$ are the productivity of Chl.a [mg-C mg-Chl.a^{-1} h^{-1}] and Chl.a concentration [mg-Chl.a m^{-3}], respectively, as functions of depth, z [m], and time, t [h].

$CHL(z)$ is expressed with a linear equation (Equation (3)), where z_{12} is the range of depths investigated, and CHL_1 and CHL_2 are the Chl.a concentrations at the corresponding boundary depths:

$$CHL(z) = CHL_1 - \frac{CHL_1 - CHL_2}{z_{12}} z \tag{3}$$

Based on the kinetics of phytoplankton growth, $Pc(z, t)$ can be regarded as a function of growth-limiting factors, light intensity, and nutrients such as DIN and DIP concentrations (e.g., Hashimoto & Takeoka, 1998; Maar *et al.*, 2016); however, as mentioned later, PP_{stn} in Hiroshima Bay was reproduced well by $Pc(z, t)$, a function of light intensity (Equation (4)), as reported by Eilers & Peeters (1988) and Wetsteyn & Kromkamp (1994):

$$Pc(z, t) = \frac{I(z, t)}{a[I(z, t)]^2 + bI(z, t) + c} \tag{4}$$

where $I(z,t)$ is the photon flux density [μmol photons m^{-2} sec^{-1}] dependent on z and t. According to the Lambert Beer's law, $I(z,t)$ is expressed by Equation (5):

$$I(z, t) = I_0(t) \exp(-K_d z) \tag{5}$$

where $I_0(t)$ is the PAR at the surface at time, t [μmol photons m^{-2} sec^{-1}]. $I_0(t)$ was calculated from the maximum photon flux density $I_{\max}$ [μmol photons m^{-2} sec^{-1}] with Equation (6) when β is a constant.

$$I_0(t) = I_{\max} \sin(\beta t) \tag{6}$$

$I_{\max}$ was estimated using the maximum amount of global solar radiation, $SR_{\max}$ [MJ m^{-2} h^{-1}], reported by the Japan Meteorological Agency. $SR_{\max}$ was converted to $I_{\max}$ using Equation (7) (Ishikawa *et al.*, 1988), where the constants, 0.42, 0.79, 23.4, 0.20×10^6, and 2.78×10^{-4} were the ratio of PAR to the amount of global solar radiation (Bassham, 1977), the passage of PAR through the sea surface into its subsurface (Ishikawa *et al*., 1988), the ratio of unit [MJ m^{-2}] to unit [cal cm^{-2}], the factor for converting solar radiation [cal cm^{-2} d^{-1}] to photon flux density [μmol-photons m^{-2} d^{-1}] (Clough & Attiwill, 1980), and the ratio of unit [h^{-1}] to unit [s^{-1}], respectively:

$$I_{max} = SR_{max} \times 0.42 \times 0.79 \times 23.4 \times (0.20 \times 10^6) \times (2.78 \times 10^{-4}) \quad (7)$$

When estimating $Pc(z, t)$ for each season, the corresponding seasonal average of $I_0(t)$ was used, assuming that the value was constant for the investigated period. Briefly, the SR_{max} and the time from sunrise to sunset reported by the Japan Meteorological Agency were averaged for each season of the 30-year period from 1981 to 2010. Using the averages of SR_{max} and the time for Equations (6) and (7), we obtained the seasonal I_{max} and β (Table S2, available with the online version of this paper).

The primary production in each season, PP_{area} [mg-C m^{-2} d^{-1}], was calculated for each subarea with Equation (8):

$$PP_{area} = \sum (PP_{stn} \times A_{subsec})/A_{area} \quad (8)$$

where A_{subsec} is the area of each subsection as determined by the Voronoi tessellation [m^2], and A_{area} is that of each Bay and Nada [m^2].

Validation of the model to estimate primary production

As mentioned later, when Pc was calculated with Equation (1) using the measured Pr of the seawater samples collected in Hiroshima Bay, the nutrient availability in spring and summer indicated by the low Pc was low compared with that in winter and autumn (Figure 3, Tables S3 and S4). Therefore, we decided to use the model to estimate PP_{area} for autumn and winter. When validating the model, we compared the estimates of PP_{area} for each subarea for winter and autumn in 1993 with the estimates of Hashimoto *et al.* (1997) and Tada *et al*. (1998). To estimate the PP_{area}, PP_{stn} was calculated using the Chl.*a* concentration, surveyed depth, and transparency (Secchi depth) measured by the MOE at the observation station(s) in each subsection of the investigated subareas (Figure 1) in autumn and winter of 1993. The Secchi depth was converted to K_d using Equation (9), where SD, m, and n were the Secchi depth [m] and constants determined from the data we collected in Hiroshima Bay (Figure S2). (Tables S3 and S4 and Figure S2 are available with the online version of this paper.)

$$K_d = \frac{m}{SD} + n \quad (9)$$

By inserting the Chl.*a* concentration, surveyed depth, K_d, I_{max}, and β (Table S2) into Equations (2)–(6), PP_{stn} can be calculated for each station. In the case that there was more than one station in a subsection, we used the average PP_{stn} value for the subsection. The PP_{area} was then estimated for each subarea by entering the PP_{stn} or the averaged PP_{stn} into Equation (8).

Effect of the TPLCS on primary production in the Seto Inland Sea

We estimated primary production in the 11 subareas of the Seto Inland Sea for the autumn and winter seasons from 1981 to 2010. First, the seasonal averages of the Chl.*a* concentrations and the Secchi depth

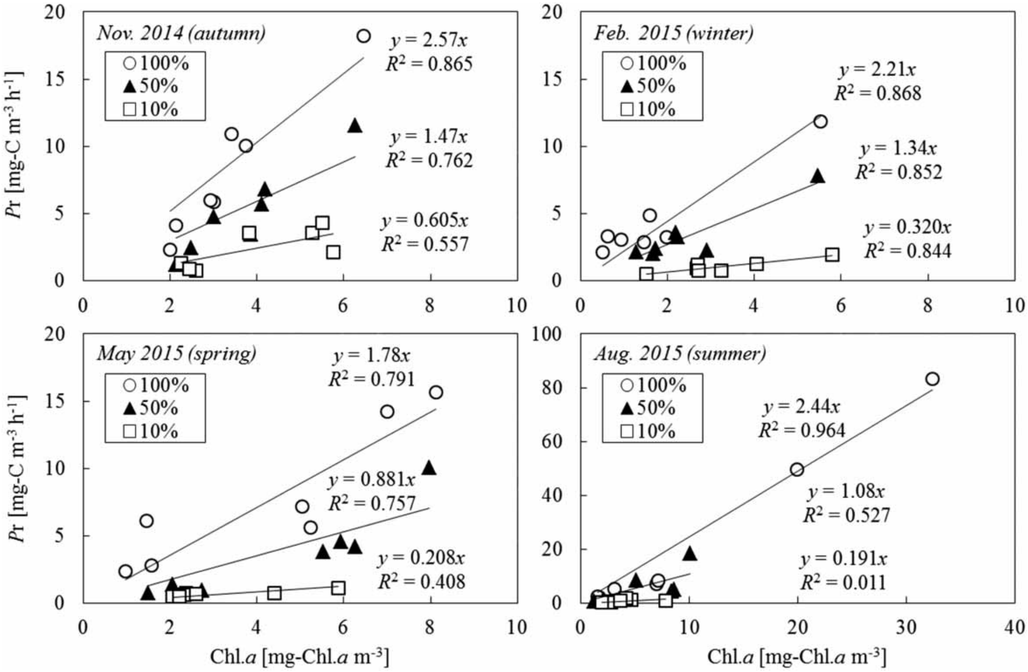

Fig. 3. Linear relationships between the Chl.*a* concentration and primary production rate (*P*r) of the seawater sampled at depths with the indicated percentage of surface radiation at the stations H1–H7 in each season. The percentages correspond to the light intensities, namely 200-, 100-, and 20-μmol photons m^{-2} sec^{-1}, at which primary production was measured.

were calculated using the CWQS data provided by the MOE. After K_d was estimated from the Secchi depth (Equation (9)), the resultant K_d, Chl.*a* concentration, and the $I_0(t)$ calculated by Equation (6) using the I_{max} and β in the corresponding season (Table S2 in the Supplementary materials) were inserted into Equations (2)–(5) to determine the PP_{stn} of each station. Finally, the seasonal PP_{area} in each subarea was estimated with Equation (8). The PP_{area} for each year was calculated by averaging the seasonal PP_{area}.

The MOE has estimated the loadings of T-N and T-P from the land into the Seto Inland Sea every five years for the TPLCS, i.e., in 1984, 1989, 1994, 1999, 2004, 2009. Nutrient concentrations including T-N, T-P, DIN, and DIP were determined annually. Therefore, we estimated the five-year-averaged PP_{area} in the 11 subareas for each five-year period from 1981 to 2010 and looked for a causative relationship with the land-originated T-N and T-P loadings. We also calculated the nutrient stocks in each subarea of the Seto Inland Sea using the T-N and T-P concentrations and the seawater volume contained in each subarea. We were not able to calculate the PP_{area} in the Suo Nada, Hiroshima Bay, and Iyo Nada subareas from 1996 to 2000 because there were no data for Chl.*a*. The total of the PP_{area} calculated for each of the 11 subareas was taken as the primary production for the Seto Inland Sea.

Statistical analysis

To investigate whether the PP_{area} had decreased, we tested the null hypothesis, wherein slope for the relationship between the estimated values and time is zero. In addition, decorrelation was tested for the relationships between the PP_{area} and loadings of land-originated nutrients.

Results and discussion

*Changes in the Chl.*a *concentrations in Hiroshima Bay over recent years*

We compared the Chl.*a* concentrations from data collected from November 2014 (autumn) to August 2015 (summer) with the seasonal average concentrations from 2005 to 2012 calculated from the CWQS data (Table S5, available with the online version of this paper). At the station H5, our observed Chl.*a* concentrations were consistently lower than the seasonal average from the CWQS data and, at H6, the values in winter and summer were lower than the minimum of the CWQS data. No such trend was observed at H1 and H2. While our data only provide a glimpse of the full situation, the results point to a possible reduction in primary production in some subsections of Hiroshima Bay; however, we could not find a consistent causative relationship between the Chl.*a* concentrations and DIN or DIP concentrations through all four seasons from our limited data (Tables S3, S4, and S5).

Construction of the model to estimate primary production

As shown by Figure 3 and the results from Equation (1), the relationships between the Chl.*a* concentrations and the measured values of *P*r of the seawater samples collected at stations H1–H7 for each season were linear. As expected, the *P*r was highest in summer; however, the slope for the summer *P*c was not the highest but was lower than the slope for the autumn. In addition, even though the spring temperature was higher than the winter temperature, the *P*c was higher in winter than in spring (data not shown). The DIN and DIP concentrations were higher in autumn and winter than in summer and spring, respectively (Tables S3 and S4). The lower *P*c in the summer and spring might therefore reflect the fact that the nutrient availability was lower in these seasons than in autumn and winter. Since the available N and P concentrations were not included to estimate *P*c in Equation (4), we decided to calculate *P*c(*z, t*) and use it for further investigation.

PP_{stn} is estimated using *P*c(*z, t*) (Equation (2)), and the parameter for *P*c(*z, t*) is the light intensity, $I(z,t)$ as defined in Equation (4) (Eilers & Peeters, 1988; Wetsteyn & Kromkamp, 1994). If the light intensity is too high, Equation (4) is used to express the occurrence of photoinhibition. The maximum light intensity of 200-µmol photons m^{-2} sec^{-1} used in our study to measure *P*r was much lower than that of the estimated I_{max} in all seasons; we were unable to measure *P*r values at greater than 200-µmol photons m^{-2} sec^{-1} because of limitation in our experimental apparatus. Since we could not experimentally determine the photoinhibitory light intensity, we assumed that a value of 307.4-µmol photons m^{-2} sec^{-1} was appropriate (18.3 klux, Kadowaki *et al*., 1993), and used this value and the observed data in Equation (4) to determine the constants *a, b*, and *c* for each season (Figure S3). The calculated constants are summarized in Table S6. (Figure S3 and Table S6 are available with the online version of this paper.)

In the CWQS, the Secchi depth, rather than K_d, is measured as an index of apparent turbidity. In our survey of Hiroshima Bay, we therefore measured both the Secchi depth and K_d to obtain an equation so that we could convert the values measured by the MOE to K_d and calculate $I(z,t)$ using Equation (5). As shown in Figure S2, there was a linear relationship between the inverse of the Secchi depth and K_d when the Secchi depth was between 0.1 and 0.4, and *m* and *n* in Equation (9) were determined as 0.64 and 0.14, respectively.

Validation of the model to estimate primary production

Hashimoto *et al*. (1997) and Tada *et al*. (1998) measured primary production in the Seto Inland Sea by the ^{13}C method. To verify our simple model, we calculated PP_{area} in the subareas of the Seto Inland Sea using the CWQS data, and compared our estimates of PP_{area} with those measured by Hashimoto *et al*. (1997) and Tada *et al*. (1998). While the PP_{area} was estimated from only light intensity and Chl.*a* concentrations in Equations (2)–(9), the estimates showed good agreement with the values reported previously for each subarea (Figure S4, available with the online version of this paper). Models based on only light intensity and Chl.*a* have been successfully used in previous studies (Lawrenz *et al*., 2013; Marchese *et al*., 2015), and therefore we decided to use Equations (2)–(9) to estimate historical changes in primary production in autumn and winter in the Seto Inland Sea and its subareas.

Historical changes in primary production and loadings of land-originated nutrients

Figure 4 shows historical changes in the PP_{area} estimated for the Seto Inland Sea and its subareas, where the former was determined by averaging the latter, and the size of each subarea was considered as in Equation (8). So that we can check whether the PP_{area} has decreased, the parameters obtained from linear regression analysis of the data in Figure 4 are summarized in Table 1, showing the statistical test result for a null hypothesis wherein slope is zero. Because there were no Chl.*a* data for Hiroshima Bay, Iyo, and Suo Nadas from 1996 to 2000, we were not able to calculate the PP_{area} for the entire Seto

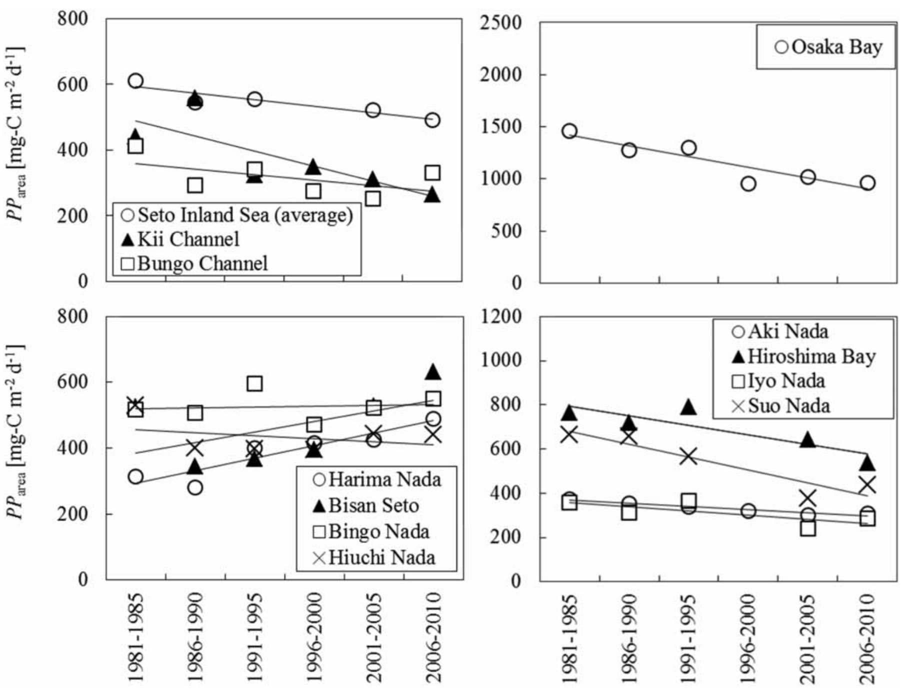

Fig. 4. Temporal variations in the five-year-averaged PP_{area} in the Seto Inland Sea. The parameters from linear regression are summarized in Table 1.

Table 1. Parameters obtained by linear regression for changes in the PP_{area} (Figure 4).

Whole/Subarea	Slope	R^2
Seto Inland Sea	−16	0.59
Osaka Bay	−104**	0.83
Aki Nada	−14***	0.94
Hiroshima Bay	−43*	0.74
Iyo Nada	−18	0.53
Suo Nada	−59*	0.87
Harima Nada	38**	0.85
Bisan Seto	32	0.28
Bingo Nada	2.5	0.012
Hiuchi Nada	−9.1	0.11
Kii Channel	−23	0.47
Bungo Channel	−0.73	9.9×10^{-4}

*$p < 0.05$.
**$p < 0.01$.
***$p < 0.001$ for a null hypothesis wherein slope is zero.

Inland Sea for these years; however, regression analysis shows that the PP_{area} has decreased across all of the Seto Inland Sea since 1981.

PP_{area} decreased in 4 of the 11 subareas, namely, Aki and Suo Nadas, and Hiroshima and Osaka Bays. Although not confirmed by statistical significance, the Iyo Nada ($p = 0.16$) and Kii Channel ($p = 0.13$) showed a tendency to decrease, indicated by the slopes, −18 and −23, and the coefficients of determination, 0.53 and 0.47, respectively. While the slope was negative in the Hiuchi Nada and Bungo Channel, it was difficult to determine whether the PP_{area} had reduced because of the high p values ($p > 0.52$). The PP_{area} has increased over the past 30 years in the Harima Nada despite the implementation of the TPLCS. While the slope was positive in the Bisan Seto and Bingo Nada, the increase of PP_{area} was not statistically significant ($p > 0.29$). The results confirm that PP_{area} in the Seto Inland Sea has decreased in line with the TPLCS implementation, but the reductions are not consistent in every subarea.

The changes in the five-year-averaged PP_{area} and the land-originated loadings of T-N and T-P are compared in Figures 5 and 6, and the parameters obtained from linear regression analysis of the data are summarized in Table 2 with the results of the statistical testing for decorrelation. While all the T-N and T-P constituents in seawater are not available to phytoplankton, there are inherent proportional relationships because nitrogen and phosphorus are essential for phytoplankton growth. The PP_{area} and the land-originated loadings of T-N and T-P were strongly and positively correlated across all of the Seto Inland Sea, thereby indicating the effect of the reductions in nutrient loadings because of the TPLCS on the total primary production in this semi-enclosed sea. The two subareas of the Kii and Bungo Channels, contiguous to the Pacific Ocean, were expected to be insensitive or less sensitive to changes in the loadings of land-derived nutrients. While the slope for the relationship between the PP_{area} and the land-originated loadings of T-N and T-P was positive in the Kii and Bungo Channels, the correlation was not statistically significant ($p > 0.18$) (Figures 5 and 6, Table 2).

Since 1981, the PP_{area} has generally decreased in 4 of the 11 subareas, the Osaka and Hiroshima Bays and Aki and Suo Nadas in line with decreases in the T-N and/or T-P loadings from the

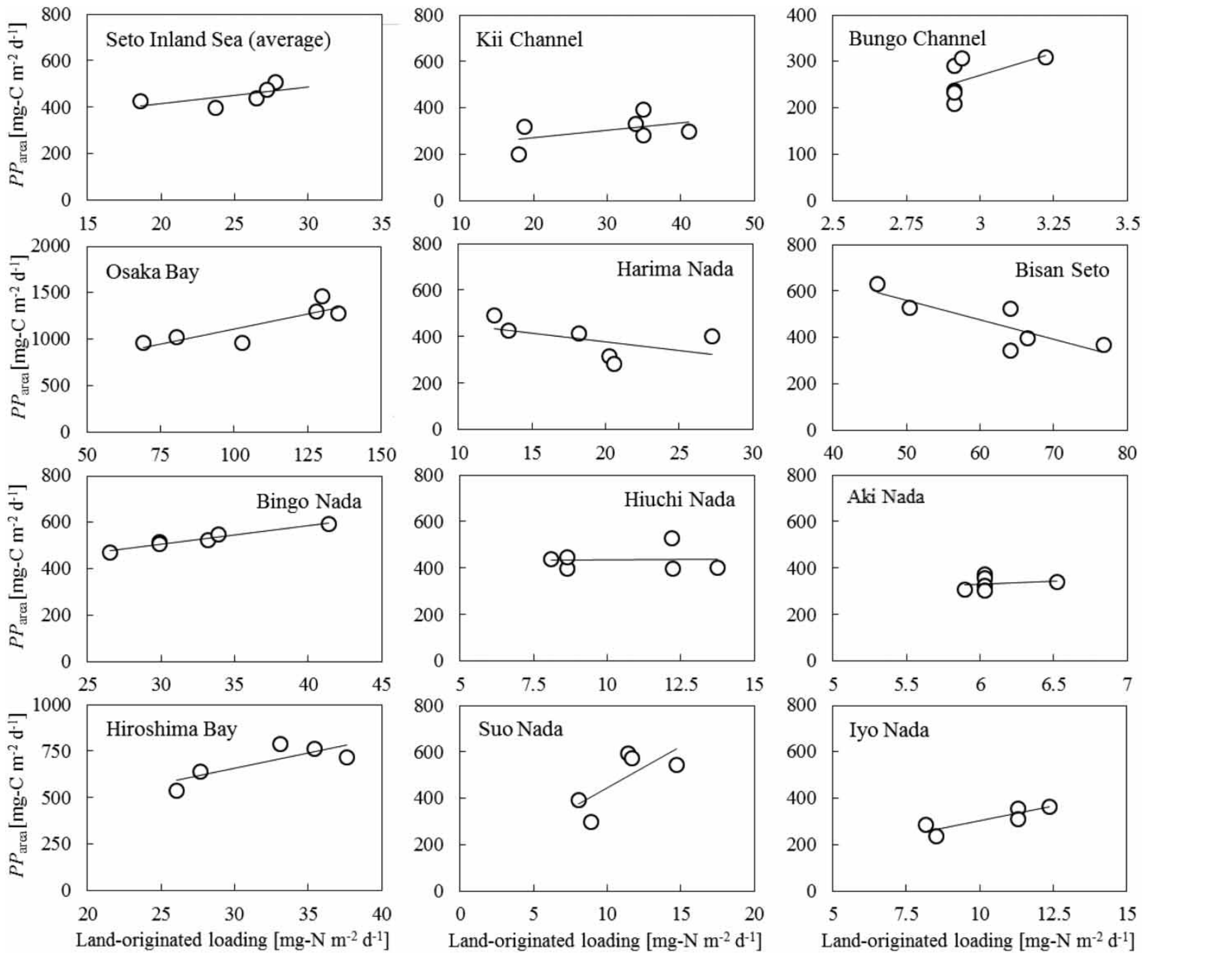

Fig. 5. Relationships between the five-year-averaged PP_{area} and the land-originated N loadings in the Seto Inland Sea. Each of the land-originated T-N loadings measured in 1994, 1999, 1994, 1999, 2004, and 2009 was used for the corresponding five-year-averaged PP_{area}, 1981–1985, 1986–1990, 1991–1995, 1996–2000, 2001–2005, and 2006–2010.

surrounding land. Surprisingly, the PP_{area} has been increasing rather than decreasing in the Harima Nada since 1981 (Figure 4, Table 1), resulting in negative correlations with the land-originated loadings of both T-N and T-P (Figures 5 and 6, Table 2). For the Bisan Seto, the increase of PP_{area} was not statistically significant but the slope was positive over the 30-year period (Figure 4, Table 1). Although a statistical significance was not confirmed for the PP_{area} and the land-originated T-N and T-P loadings in the Bisan Seto ($p > 0.09$) (Figures 5 and 6, Table 2), the slope was negative with $R^2 = 0.55$ and $R^2 = 0.47$, respectively. In the Iyo, Bingo and Hiuchi Nadas, a correlation was not observed ($p > 0.1$) (Figures 5 and 6, Table 2). The results confirm that these four subareas are insensitive or less sensitive to reductions in land-originated nutrients generated by the TPLCS.

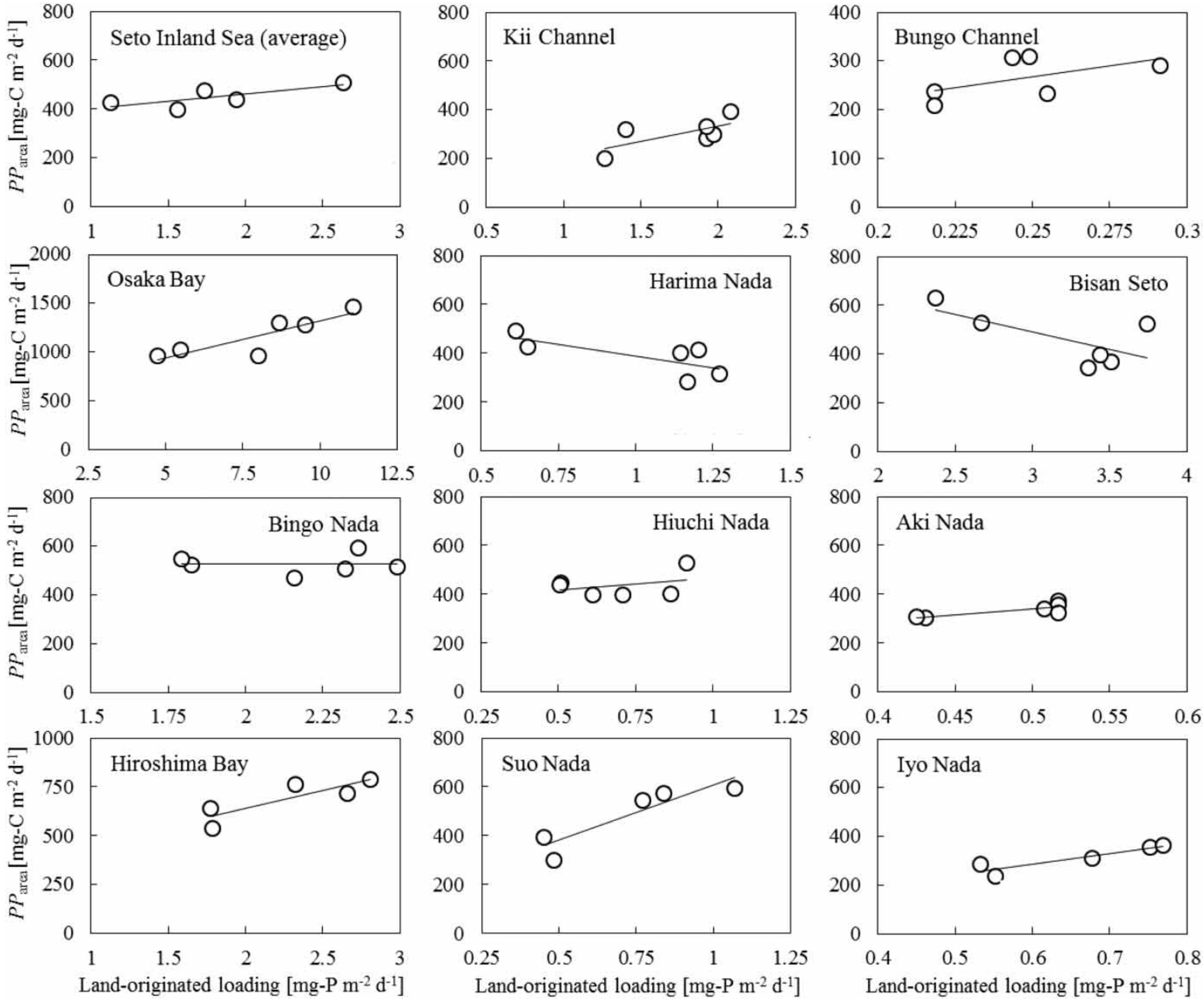

Fig. 6. Relationships between the five-year-averaged PP_{area} and the land-originated P loading in the Seto Inland Sea. Each of the land-originated T-P loadings measured in 1994, 1999, 1994, 1999, 2004, and 2009 was used for the corresponding five-year-averaged PP_{area}, i.e., for 1981–1985, 1986–1990, 1991–1995, 1996–2000, 2001–2005, and 2006–2010.

Even though all of the T-N and T-P constituents in seawater are not available for phytoplankton, the reduced sensitivity or insensitivity of these four subareas to reductions in the land-originated loadings of T-N and T-P may reflect other major nutrient sources. The annual changes in the land-originated loadings of T-N and T-P and the stocks of T-N and T-P in each subarea of the Seto Inland Sea presented in Figures S5 and S6 show that there were reductions in the land-originated loadings of nutrients in all subareas since 1981 except Bingo Nada and Bungo Channel, where T-N did not decrease. Despite this, the stocks of T-N and T-P in Harima Nada and Bingo Nada have not decreased, nor has the PP_{area} decreased, over this 30-year period. In addition, there were no obvious reductions in the stock of T-N in Hiroshima Bay and Aki Nada. The results therefore indicate that nutrients from sources other than the surrounding land control the phytoplankton growth in these subareas.

Table 2. Parameters obtained by linear regression for the relationship between the PP_{area} and land-originated nutrient loadings (Figures 5 and 6).

Whole/Subarea	Land-originated T-N loading		Land-originated T-P loading	
	Slope	R^2	Slope	R^2
Seto Inland Sea	10*	0.76	77**	0.95
Osaka Bay	9.1*	0.82	110**	0.87
Aki Nada	93	0.19	840*	0.67
Hiroshima Bay	13*	0.80	120	0.66
Iyo Nada	21	0.46	410	0.63
Suo Nada	32	0.41	470*	0.87*
Harima Nada	−15	0.27	−440*	0.63
Bisan Seto	−7.7	0.55	−150	0.47
Bingo Nada	4.3	0.51	13	0.016
Hiuchi Nada	−3.6	0.060	16	6.4×10^{-3}
Kii Channel	5.7	0.26	200	0.39
Bungo Channel	96	0.044	1500	0.52

*$p < 0.05$.
**$p < 0.01$ for decorrelation test.

Adjacent subareas may be important nutrient sources (Staehr *et al.*, 2017). For example, Harima Nada is next to Osaka Bay and the Kii Channel, and nutrient transport from the Pacific Ocean is also well documented (Yanagi & Ishii, 2004). Sediment may also be a source of nutrients (Savchuk *et al.*, 2009; Jikumaru *et al.*, 2015; Nguyen & Maeda, 2016). In addition, seawater velocity may affect primary production by not only changing the retention of nutrients (Savchuk *et al.*, 2009) but also light conditions (Wetsteyn & Kromkamp, 1994).

The results show that the stocks of T-N and T-P and primary production have decreased in the four subareas of the Seto Inland Sea since 1981 because of the TPLCS. However, PP_{area} values have not decreased in all subareas. Despite the implementation of the TPLCS, the reductions in PP_{area} were not statistically significant in the Bungo and Kii Channels, Iyo, Bingo, and Hiuchi Nadas, and Bisan Seto, and the PP_{area} actually increased in Harima Nada over the 30-year period. Since primary production in all subareas of the Seto Inland Sea was not controlled by only reducing the land-originated loadings of nutrients, the sources of nutrients that control primary productivity need to be identified for each subarea for better control of primary production.

Policies for better management

In the Seto Inland Sea, the eighth TPLCS has been implemented since 2016. Since the Seto Industrial Zone lies on the coast of the Seto Inland Sea, a special council for the TPLCS in the MOE required the Japan Chemical Industry Association and Japan Iron and Steel Federation to report the results of total pollutant load control according to the seventh TPLCS implementation as well as their opinions on the next one in 2015. The former pointed out the necessity of reviewing the effects of TPLCS including the lack of nutrients associated with the fishery production (Japan Chemical Industry Association, 2015), while the latter mentioned that the TPLCS should be accompanied with actions such as restoration of seaweed beds and tidal flats towards a realization of bountiful seas (Japan Iron and Steel Federation, 2015). Later in 2015, the Law Concerning Special Measures for Conservation of the Environment of the Seto Inland Sea was amended, where its values and functions are fully exerted to achieve the goal of a beautiful and bountiful sea. Under the amended law, the conservation countermeasures are to be taken considering the circumstances of subareas in the Seto Inland Sea. Therefore, the eighth TPLCS has been implemented to accomplish water quality controls suitable for each subarea in the Seto Inland Sea in combination with conservation and regeneration of seaweed beds and tidal flats and restoration of the sediment environment (Negi, 2016). The future environmental management measures need to be implemented in response to the sources of nutrients for better control of primary production towards the realization of beautiful and bountiful seas.

Conclusion

In this study, a simple model, based on CWQS water quality data (MOE, Japan), was constructed to estimate the PP_{area} by phytoplankton over the 30-year period from 1981 to 2010, and to determine whether the PP_{area} in the Seto Inland Sea had been successfully controlled because of the implementation of the TPLCS. The model was validated by comparing the PP_{area} estimated for subareas of the Seto Inland Sea in the autumn and winter in 1993 with those from previous studies. The estimated PP_{area} values for the study period showed that the PP_{area} had generally decreased across the Seto Inland Sea, and had decreased in 4 of its 11 subareas since 1981 in line with decreases in the T-N and/or T-P loadings from the surrounding land. However, there were either increases or insignificant change in the PP_{area} of the remaining seven subareas despite the implementation of the TPLCS. In two of these seven subareas, the Harima Nada and Bingo Nada subareas, the stocks of T-N and T-P did not decrease over the 30-year study period, indicating that the surrounding land was not the main source of nutrients.

The inconsistent responses of the 11 subareas to the TPLCS suggest that the characteristics of each subarea should be considered when environmental management measures are established and implemented in the Seto Inland Sea. Since primary production in all subareas of the Seto Inland Sea cannot be controlled by only reducing the land-originated loadings of nutrients, the sources of nutrients that control primary productivity in the subareas of the Seto Inland Sea should be identified in future studies for better management of nutrients to establish a healthy and productive aquatic ecosystem.

Acknowledgement

Special thanks are given to the MOE and MLIT, Japan, for supplying data from their investigation of the Seto Inland Sea. This study was supported by a grant from the Environment Research and Technology Development Fund (S-13) of the MOE, Japan (S-13).

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Received 22 June 2017; accepted in revised form 29 January 2018. Available online 19 March 2018