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Baseline

Improved water quality in response to pollution control measures at Masan Bay, Korea

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ABSTRACT

The total pollution load management system (TPLMS) was first applied in 2007 to the highly developed Masan Bay watershed, Korea. To evaluate the effect of TPLMS on water quality improvement, we analyzed the water qualities in rivers and bay during 2005–2010, targeting chemical oxygen demand (COD), suspended sediment (SS), total nitrogen (TN), and total phosphorus (TP) loads. Land-based pollutant loading all decreased during this period, with a significant reduction in COD and SS loads ($p < 0.01$). The COD reduction in seawater, following the TPLMS implementation, was also significant ($p < 0.01$). Time-lagged responses in COD and Chl-*a* supported an estimated seawater residence time of ~1 month. Land-based nutrient loads were also significantly reduced for TN ($p < 0.01$) and TP ($p < 0.05$), however, significant reductions were not observed in the bay, indicating potential alternative nutrient inputs from non-point sources into the bay system.

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Coastal zones between terrestrial and marine systems that receive large quantities of nutrients from inflowing rivers. Human settlements tend to form along coastal areas, consequently intensifying pollution problems, such as waste disposal and habitat loss, in watershed areas. Some pollutants may lead to eutrophication and health problems that, in turn, adversely affect ecosystems, including nature and humans through a variety of mechanisms (Cloern, 2001). For example, the depletion of dissolved oxygen (DO), which is caused by the aerobic decomposition of organic and inorganic compounds, represents one of the major environmental concerns in many coastal areas throughout the world (Diaz and Rosenberg, 1995). In addition, pathogenic microbes in sewage-born polluted waters pose a serious public health problem, limiting recreational use of those waters (Bartram and Rees, 2000). Fortunately, some of the adverse effects of pollution can be reversed through abatement actions (Cloern, 2001; Garcia-Barcina et al., 2006). However, major environmental spatiotemporal variability and/or time-lags in recovery responses make the evaluation of

the progress towards achieving the goals of pollution reduction difficult in coastal ecosystems (Boesch, 2002).

Masan Bay has been identified as a coastal area that is becoming one of the most polluted estuaries in Korea (Khim and Koh, 2011). During the last 40 years, the natural features of the bay have been dramatically modified by urban, industrial, and port developments, with its tidal wetlands having been reclaimed to accommodate the expansion of a large population and ever-growing industry (Ryu et al., 2011). As a result, the bay system became quickly and heavily polluted by a variety of wastes, including untreated municipal sewage and industrial wastewater, which led to harmful algal blooms, sharp oxygen depletion, loss of aquatic life, and aesthetic problems. In addition, due to significant bacteriological pollution, Gapo Beach (situated near site R3, Fig. 1) was permanently closed in 1975. This was followed by shellfish harvesting being prohibited from 1979 onwards. Since the 1980s, there has been public and regulatory concern about these pollution problems. For example, the central government has been working intensively towards reducing land-based pollution. A wastewater treatment plant (WWTP) was constructed in 1993 with a capacity of 280,000 ton d⁻¹, which could potentially treat about 50% of total freshwater inputs to Masan Bay (MLTM, 2008). In addition, there was also intensive dredging activity in the bay between 1990 and 1994 to remove polluted

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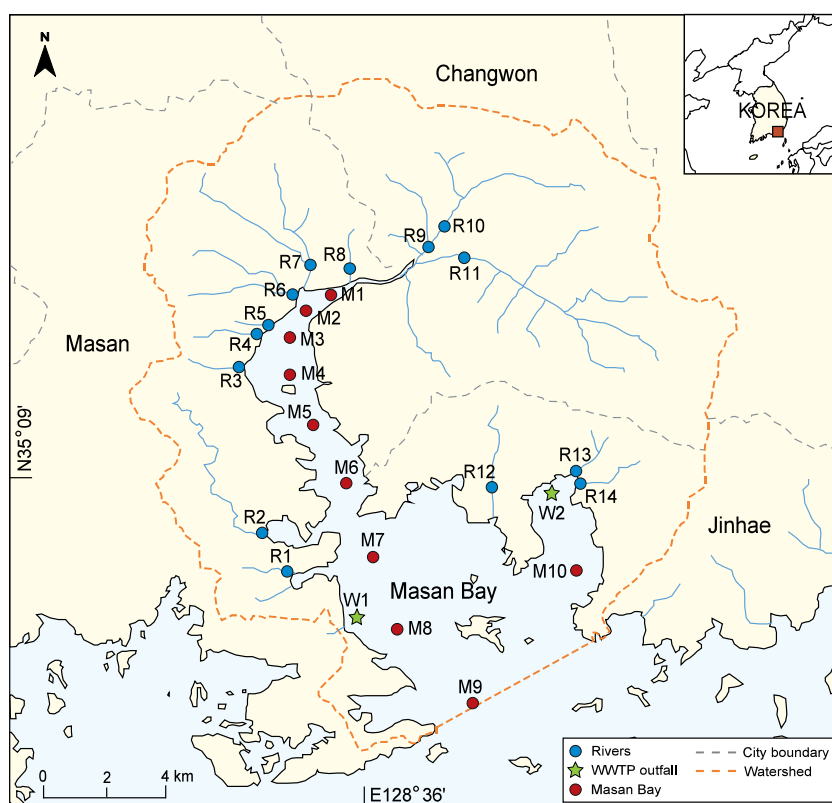


Fig. 1. Map showing the sampling locations in the study area. Sampling details are fully given in Table 1.

sediments, with a total investment of 36 million USD. However, these preliminary efforts failed to improve the quality of the water in Masan Bay.

More recently, a total pollutant load management system (TPLMS) was launched for Masan Bay in 2007, targeting a level of water quality that is suitable for 'swimming and fishing' by 2020 (MLTM, 2008). The TPLMS project comprises a total of 800 km of combined sewer networks that convey sewage to two central WWTPs that have been upgraded with an advanced biological treatment facility and an extended capacity of 500,000 ton d^{-1} (82% of total freshwater input). To date (2007–2010), over 200 million USD has been invested into the TPLMS, with 30% of funding being provided by the private sector. The main purpose of the TPLMS was divided into two stages: (1) the reduction of organic matter (targeting chemical oxygen demand–COD–loads) from point sources in the watershed during 2007–2011 and (2) the reduction of nitrogen and phosphorus related nutrients in the bay through regulating diffuse sources across the watershed during 2012–2016.

Appropriate monitoring is crucial to identify the efficacy of the TPLMS, in addition providing a useful tool to verify the acquisition of the established goals (Boesch, 2002). Therefore, since 2005, comprehensive surveys have been undertaken at periodic intervals to monitor the land-based pollutant load and general water quality of Masan Bay. In this paper, we explore in detail the spatiotemporal responses in water quality of Masan Bay to pollution abatement measures that have been conducted over the last 20 years. To specifically quantify the efficacy of TPLMS implementation, we evaluated the existence and magnitude of temporal trends and variability in water quality over a six-year period (2005–2010) with respect to (1) land-based pollutant loads discharge and (2) univariate parameters of the receiving water quality in Masan Bay.

The target area of Masan Bay (Fig. 1) is located on the southeast coast of Korea (35°3'–35°16'N, 128°32'–44'E) with a watershed of

about 264 km 2 . Fourteen small rivers drain the watershed, supplying 611,000 tons of freshwater on average per day to the bay. The Samho, Nam, and Changweon rivers account for about 55% of the total freshwater inflow. Masan Bay is a semi-enclosed coastal embayment, with a surface area of about 71 km 2 , an average depth of about 10 m, and a total volume of 0.77×10^9 m 3 . The bay has a low water-exchange rate with residence time of bay water having 53.7 days in the inner bay and 23.2 days in the whole bay (Lee et al., 2009). The south part of the bay is connected to the Jinhae Bay system (approximately 637 km 2 in the surface area), which includes Masan Bay and several other small bays. Three cities surround Masan Bay, namely Masan City, Changweon City, and Jinhae City. These three cities are highly industrialized and, in 2010, had a combined population of ca. 1.1 million people. The population density is 3167 persons per km 2 , which is 6.5 times above than the national average. The urban area of the three cities contributes to about 40% of the total watershed area. The relatively small size of the watershed is due to its steep slope towards the coast.

The original morphology of Masan Bay has been strongly modified over the last four decades by the construction of dikes across large areas of tidal wetlands and by extensive dredging activities to maintain navigation routes in the channel. Intensive industrial and residential development resulted in an increase of impervious and paved surfaces in the watershed area, for example, covering about 30% of surface area by 2006.

The survey conducted within the framework of this study can be divided into three categories (Data Sets I–III) based on their specific purposes (Table 1). Details of water sampling, including sampling area, sampling activities, frequency of sampling, target water quality parameters, and other acquired data, are presented with respect to data category.

First, the pollution load of the water from inland areas was monitored by river water sampling (Data Set I). In total, 14 river sites (R1–R14) were surveyed monthly from June 2005 to December

Table 1
Details of field study and data structure, summarized into three parts (Data Set I–III).

| | Data Set I | Data Set II | Data Set III |
|---|--|-----------------------------------|---|
| Data characteristics | Water quality load from inland | Influent & effluent water quality | Water quality in bay system |
| Sampling area | Inland rivers | Waste water treatment plant | Masan Bay |
| Samples | Freshwater | Influent & effluent | Seawater (surface & bottom) |
| # of locations (<i>adjacent city</i>) | | | |
| (Masan) | 8 | 1 (Deokdong WWTP) | 10 (bay area) |
| (Changwon) | 3 | | |
| (Jinhae) | 3 | 1 (Jinhae WWTP) | |
| (total) | 14 | 2 | 10 |
| # of survey (<i>year</i>) | | | |
| (2005) | 7 | 365 | 4 |
| (2006) | 17 | 365 | 8 |
| (2007) | 18 | 365 | 10 |
| (2008) | 18 | 365 | 11 |
| (2009) | 19 | 365 | 12 |
| (2010) | 19 | 365 | 11 |
| (total) | 98 | 2190 | 56 |
| Sampling frequency | Monthly (biweekly during summer) | Daily | Seasonal to monthly |
| Water quality parameters | Temp, Salinity, DO, COD, BOD, SS, TN, TP, NO_3^- -N, NH_4^+ -N, PO_4^{3-} -P, SiO_2 -Si, TOC, POC, DOC | COD, BOD, SS, TN, TP, flow | Temp, Salinity, DO, COD, SS, TN, TP, NO_3^- -N, NH_4^+ -N, SiO_2 -Si, TOC, POC, DOC, Chl- <i>a</i> |
| Key parameters | ←COD, SS, TN, TP→ | | |
| Other data | Rainfall, porewater chemistry, and socio-economic data | | |
| Data presented in | Table 2 and Figs. 2, 5 and 6 | Table 2 and Figs. 3, 5 and 6 | Table 2 and Figs. 4–6 and 6 |

2010. During the summer monsoon season (from June to September), sampling was performed approximately every two weeks. At each site, a surface water sample was collected from the riverbank using a bucket sampler. Various water quality variables were measured, with key parameters comprising COD, suspended sediment (SS), total nitrogen (TN), and total phosphorus (TP).

Second, to trace and monitor effluent water quality of two WWTPs (inland point sources), the water quality data of their influent and effluent were collected from the local WWTP management agency. The two target WWTPs are located in the southern part of Masan Bay, one operating for the cities of Masan and Changwon (W1) and the other for Jinhae City (W2). Daily data for COD, biological oxygen demand (BOD), SS, TN, TP, and flow during the same period as river sampling (i.e., June 2005 to December 2010) were collectively used to calculate land-based pollution loads. Furthermore, the water quality data of the WWTPs were utilized for comparative purposes; viz. land-based pollution loads (rivers or WWTPs) versus bay water quality (Table 2), in terms of cause and effect.

Finally, water quality in the bay system was monitored monthly at 10 sites across the bay (M1–M10) from July 2005 to October 2010. During the summer, water samples were taken twice a month. At each site in the bay, the surface water was sampled on board a boat using a bucket sampler, while bottom water samples (1 m above the seabed) were collected using a Niskin bottle. Target water qualities were almost the same as those of river sampling, measured for the COD, SS, TN, TP, and chlorophyll-*a* (Chl-*a*).

All of the water samples from the inland rivers and Masan Bay were immediately transferred to the laboratory, and water quality variables were analyzed. In brief, COD was measured using the KMnO_4 -based titration method according to the Korean water quality testing guideline (MOE, 2008). For SS, the water samples were filtrated using a GF/F filter, and the filter residues were dried at 105 °C for 2 h, and then weighed. TN and TP were analyzed using an Integral Future Automatic Analyzer (Alliance). Chl-*a* was measured using High Performance Liquid Chromatography.

Other data, such as rainfall (acquired from the Masan Climate Center) and socio-economic data (population, land-use, and TPLMS

related activities), were also collected to aid the understanding and interpretation of Masan Bay water quality during the period of 2005–2010.

Temporal trends of land-based pollution loads and bay water quality for COD, SS, TN, and TP were analyzed using a simple regression model at the scale of the whole bay. This was achieved by averaging the water quality data of 14 river sites for pollution loads and 10 bay sites for bay water quality (all data summarized in Supplementary table: Table S1). In addition, Spearman correlation analysis was conducted for COD, SS, TN, TP, and Chl-*a* to estimate the extent to which temporal changes in pollution loads explain the quality of receiving water parameters in the bay at 95% significance level. Three datasets were used for this analysis to determine the 'lag' effect (if any) of loads (Data I and II) on bay water quality (Data III) with respect to: (1) river/WWTP data and the closest bay sampling date and (2) river/WWTP data followed by a one month lapse in bay sampling. Precipitation and water quality of the influent and effluent at the two WWTPs were represented as monthly averages that were derived from daily datasets.

To test whether the TPLMS has a significant effect on water quality improvement in the bay, two additional comparisons were made. First, an analysis of covariance (ANCOVA) was conducted for COD, SS, TN, and TP after removing the variance of loads during the six sampling years. Second, the summer COD and TN in bottom sediment and bottom seawater of the bay were compared before and after the TPLMS (viz. 2006 versus 2010) for representative bay sites (M2, M5, M8, and M10). All statistical analyses were performed using SPSS 12.0.

During the six-year study period, the total monthly pollution loads discharged into the bay showed large monthly variations with clear seasonal fluctuations (Fig. 2). In general, the concentrations of all key water quality parameters (including COD, SS, TN, and TP) in the rivers decreased from spring to summer, and increased again towards winter. The concentrations of these water quality variables were relatively low during summer (mainly July–August), which reflected the dilution effect of high freshwater summer loading caused by heavier rainfall during this period.

Table 2
Relationship (r^a) of water quality parameters between river water or effluent and bay water (surface and bottom).

| Target | | Bay (surface) | | | | | Bay (bottom) | | | | | |
|----------------------------------|-----------------|---------------|--------|--------|-------|---------------|--------------|-------|--------|---------|---------------|--|
| | | COD | SS | TN | TP | Chl- <i>a</i> | COD | SS | TN | TP | Chl- <i>a</i> | |
| River versus Bay (closest date) | <i>River</i> | | | | | | | | | | | |
| | COD | -0.04 | 0.07 | -0.25* | -0.19 | -0.18 | 0.35** | 0.20 | -0.20 | -0.32** | 0.47** | |
| | SS | 0.13 | 0.24* | -0.09 | -0.02 | 0.03 | 0.25* | 0.28* | -0.10 | -0.17 | 0.38** | |
| | TN | 0.19 | 0.14 | -0.07 | 0.03 | -0.13 | 0.39** | 0.23 | -0.11 | -0.10 | 0.34** | |
| | TP | 0.13 | 0.08 | -0.25* | -0.12 | -0.17 | 0.42** | 0.17 | -0.23* | -0.22 | 0.31 | |
| | <i>Effluent</i> | | | | | | | | | | | |
| | COD | 0.30* | 0.18 | 0.04 | -0.14 | 0.02 | 0.43** | 0.18 | 0.09 | -0.23 | 0.26* | |
| | SS | 0.33** | 0.02 | 0.05 | -0.01 | 0.13 | 0.40** | -0.02 | 0.14 | 0.03 | 0.34** | |
| River versus Bay after one month | <i>River</i> | | | | | | | | | | | |
| | COD | 0.29* | 0.16 | 0.20 | -0.12 | 0.06 | 0.40** | 0.23 | -0.12 | -0.44** | 0.49** | |
| | SS | 0.21 | 0.22 | 0.00 | -0.11 | -0.03 | 0.47** | 0.29* | -0.16 | -0.26* | 0.39** | |
| | TN | 0.35* | 0.23 | 0.13 | 0.03 | 0.36** | 0.41** | 0.18 | -0.05 | -0.19 | 0.49** | |
| | TP | 0.24* | 0.11 | 0.04 | 0.01 | 0.17 | 0.29* | 0.13 | -0.20 | -0.26* | 0.39** | |
| | <i>Effluent</i> | | | | | | | | | | | |
| | COD | 0.36** | 0.13 | -0.11 | -0.01 | 0.06 | 0.55** | 0.13 | 0.01 | -0.01 | 0.47** | |
| | SS | 0.14 | 0.08 | 0.31* | 0.07 | 0.17 | 0.17 | -0.03 | -0.05 | -0.09 | 0.24* | |
| TN | 0.35** | 0.15 | 0.07* | -0.16 | 0.19 | 0.45** | 0.15 | 0.04 | -0.16 | 0.34** | | |
| TP | 0.29* | 0.34* | -0.07* | 0.18 | 0.05 | 0.47** | 0.27 | 0.11 | 0.18 | 0.50** | | |

^a Statistically significant at * $p < 0.05$ or ** $p < 0.01$.

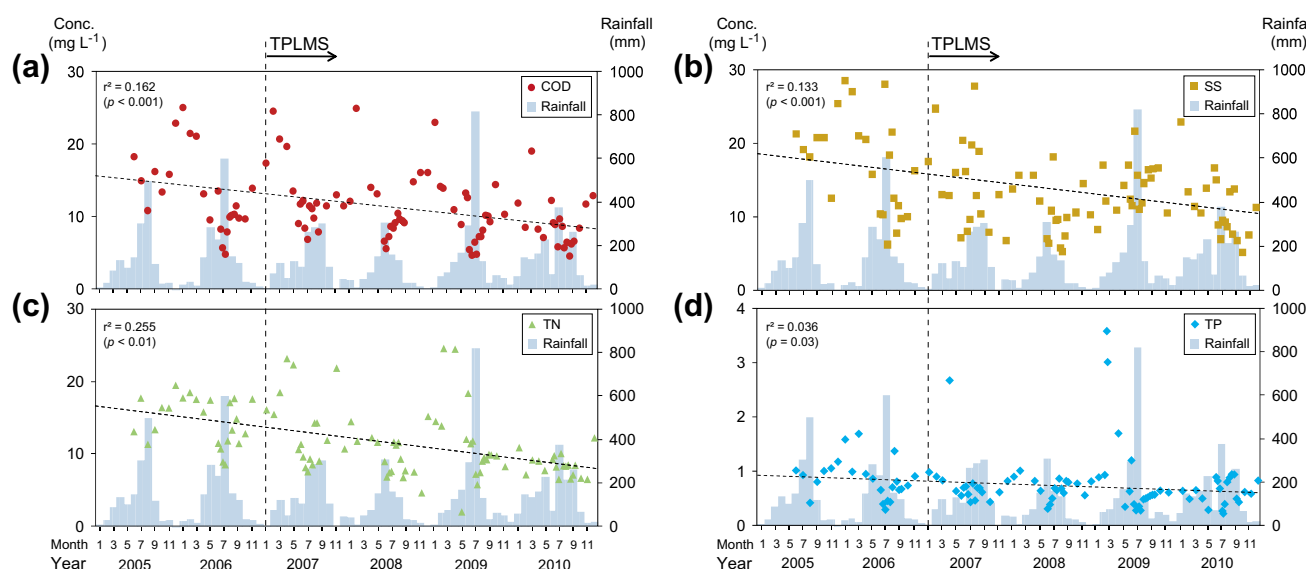


Fig. 2. Temporal changes in concentrations (mg L^{-1}) of (a) COD, (b) SS, (c) TN, and (d) TP in freshwater measured from 14 sites (given as mean) in the inland rivers over the six-year period (2005–2010), rainfall data (mm) during the corresponding period was given as background information.

Among the measured water quality parameters (Table S1), COD most strongly mirrored variation in the rainfall data (Fig. 2a). This indicates the physically associated response of COD against increased freshwater input in the river system. Compared to Korean water quality standards (WQS), the monthly average COD levels (mean = 12.8 mg L^{-1}) exceeded the WQS_{COD} level-IV (9 mg L^{-1} = moderately polluted), except during summer (mean = 8.6 mg L^{-1}), indicating severe water pollution in the Masan Bay watershed area.

The seasonal pattern of riverine SS was less distinctive (Fig. 2b). However, SS concentrations rapidly declined with increasing rainfall during the summer rainy season, which was consistent with the COD seasonal pattern. When compared to the corresponding WQS_{SS} ($< 25 \text{ mg L}^{-1}$ = not polluted), monthly SS water quality remained good across all survey years (mean = 13.0 mg L^{-1}), except for a couple of months in 2005–2007.

TN and TP also showed a clear seasonal pattern (Fig. 2c and d), but with less characteristic variation, particularly for TP, compared to COD and SS. However, exceptionally elevated concentrations of

TN (max = 21.3 mg L^{-1}) and TP (max = 3.07 mg L^{-1}) were often found during spring, indicating that spring rainfall runoff contained higher nutrients levels possibly accumulated during the winter. Alternatively, these high concentrations may be attributed to the spring thaw effect (Novotny et al., 2008).

Without exception, riverine water variables showed a decreasing trend from 2005 to 2010. Specifically, COD ($p < 0.001$, $r^2 = 0.162$) and SS ($p < 0.01$, $r^2 = 0.133$) showed much clearer decreasing trends compared to TN ($p < 0.01$, $r^2 = 0.255$) and TP ($p < 0.05$, $r^2 = 0.036$) (Fig. 2).

Over the six-year period, although the WWTPs recorded an increase in the COD and SS influent loads, the corresponding COD and SS effluent loads remained consistent (Fig. 3). This observation indicates that a certain portion of the total land-based COD and SS loading to the bay may be attributable to point sources. In parallel, the efficacy of the wastewater treatment parameters gradually increased over the years. Similar to the temporal pattern of the river waters, both the influent and effluent of COD and SS in the

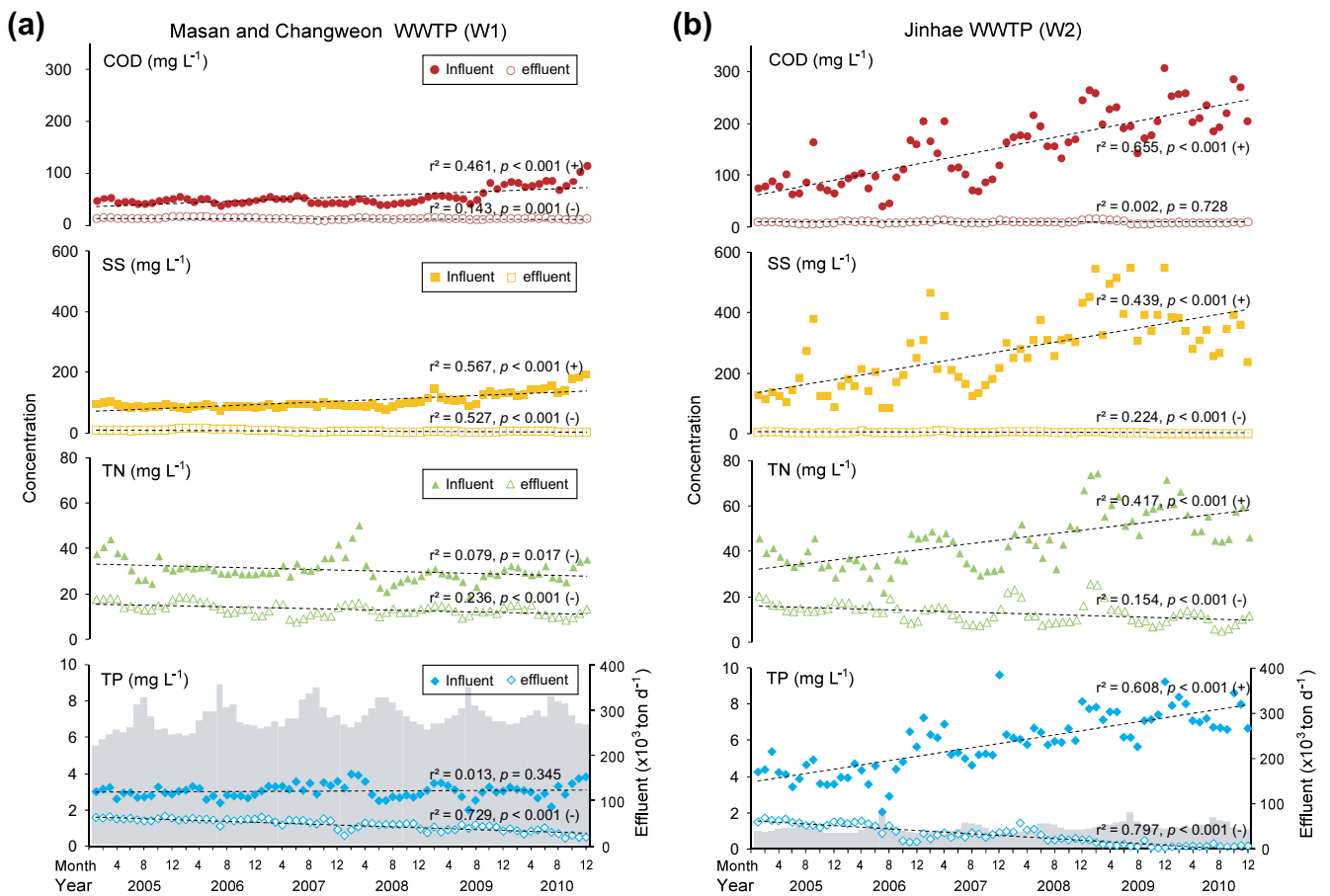


Fig. 3. Temporal changes in concentrations (mg L^{-1}) of COD, SS, TN, and TP in influent and effluent measured from (a) Masan and Changweon WWTP (W1) and (b) Jinhae WWTP (W2) over the six-year period (2005–2010), total effluent load ($\times 10^3 \text{ ton d}^{-1}$) during the corresponding period was given as background information.

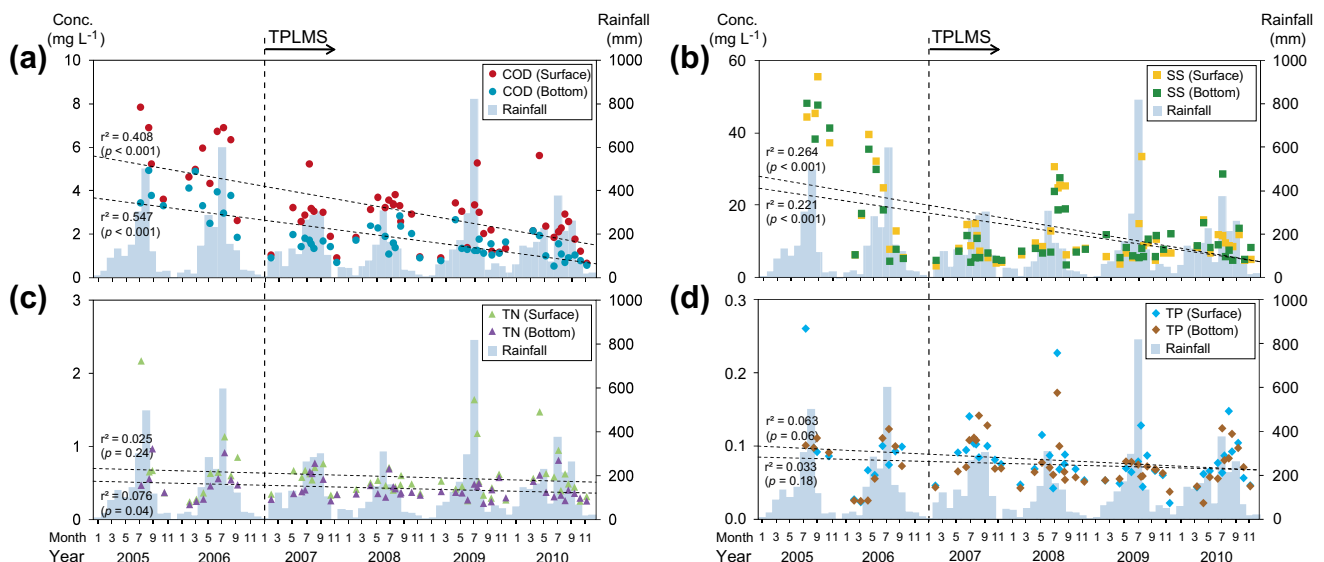


Fig. 4. Temporal changes in concentrations (mg L^{-1}) of (a) COD, (b) SS, (c) TN, and (d) TP in seawater (surface and bottom) measured from 10 sites (given as mean) in the Masan Bay over the six-year period (2005–2010), rainfall data (mm) during the corresponding period was also given as background information.

WWTPs also showed seasonal fluctuations in response to rainfall. Such seasonal patterns strengthened the COD and SS values in the WWTP of Jinhae (W2), which may be explained by the sewer system being combined with the rainwater pipe in Jinhae.

Again, the TN and TP concentrations in the influent and effluent of WWTPs showed seasonal fluctuation patterns in both W1 and W2 (Fig. 3), but tended to decrease over the survey period. These declines in TN and TP were obvious for W2, which was possibly

Table 3
comparison of water quality flux between sediment, porewater, and water from selected locations in Masan Bay between 2006 and 2010.

| Location | Year | Year 2006 | | | | | | Year 2010 | | | | | |
|----------|------|-------------------------|-----|--------------------------------------|-----|----------------------|-----|------------------------|-----|--------------------------------------|------|----------------------|-----|
| | | Sediment | | Porewater flux ^a | | Bottom seawater | | Sediment | | Porewater flux | | Bottom seawater | |
| | | COD | TN | DIN | DIP | COD | TN | COD | TN | DIN | DIP | COD | TN |
| | | (mg g ⁻¹ dw) | | (mgm ⁻² d ⁻¹) | | (mgL ⁻¹) | | (mgg ⁻¹ dw) | | (mgm ⁻² d ⁻¹) | | (mgL ⁻¹) | |
| M2 | June | 32.4 | 2.3 | 19.3 | 4.7 | 3.9 | 0.6 | 23.0 | 1.6 | 93.7 | 7.5 | 0.6 | 0.4 |
| | July | 27.0 | 1.7 | 22.7 | 9.2 | 2.1 | 0.8 | 23.2 | 2.0 | 73.9 | 23.0 | 1.8 | 0.3 |
| | Aug | 31.0 | 2.4 | 31.3 | 1.1 | 2.4 | 0.7 | 24.3 | 2.1 | 75.0 | 13.2 | 1.5 | 0.5 |
| | Sep | 29.9 | 2.1 | 9.1 | 1.5 | 1.8 | 0.5 | 25.8 | 2.3 | 94.6 | 26.0 | 0.9 | 0.5 |
| M5 | June | 23.3 | 2.1 | 13.6 | 1.8 | 3.0 | 0.5 | 28.7 | 2.4 | 204.9 | 15.8 | 0.8 | 0.3 |
| | July | 20.4 | 1.0 | 12.7 | 3.0 | 1.7 | 0.7 | 22.1 | 2.5 | 46.1 | 2.2 | 0.7 | 0.3 |
| | Aug | 37.1 | 3.9 | 60.7 | 2.5 | 2.6 | 0.6 | 28.8 | 1.7 | 95.3 | 9.1 | 0.7 | 0.4 |
| | Sep | 37.8 | 3.9 | 92.7 | 8.7 | 0.9 | 0.5 | 40.8 | 3.7 | 69.8 | 9.3 | 1.2 | 0.4 |
| M8 | June | 20.0 | 2.1 | 17.1 | 4.7 | 4.3 | 0.4 | 33.4 | 2.3 | 79.4 | 3.2 | 0.3 | 0.3 |
| | July | 18.8 | 3.1 | 32.2 | 1.7 | 2.2 | 0.6 | 25.1 | 2.5 | 20.1 | 1.8 | 0.7 | 0.1 |
| | Aug | 31.2 | 3.0 | 44.6 | 2.2 | 2.3 | 0.7 | 26.6 | 2.3 | 73.7 | 14.2 | 0.9 | 0.5 |
| | Sep | 33.8 | 3.2 | 31.2 | 2.5 | 1.2 | 0.5 | 25.4 | 2.9 | 183.7 | 21.6 | 1.1 | 0.3 |
| M10 | June | 21.9 | 1.3 | 13.1 | 3.0 | 3.1 | 0.2 | 42.2 | 2.9 | 175.4 | 16.7 | 0.5 | 0.3 |
| | July | 25.9 | 1.1 | 5.2 | 0.0 | 3.3 | 0.9 | 18.5 | 2.3 | 146.9 | 17.8 | 0.9 | 0.1 |
| | Aug | 27.8 | 2.3 | 24.5 | 1.6 | 4.8 | 0.2 | 21.0 | 2.0 | 146.1 | 15.9 | 1.2 | 0.2 |
| | Sep | 23.0 | 1.6 | 0.0 | 0.3 | 2.6 | 0.4 | 23.1 | 2.1 | 49.5 | 12.2 | 0.9 | 0.3 |

^a DIN = Dissolved Inorganic Nitrogen; DIP = Dissolved Inorganic Phosphorus.

due to the W2 plant having the higher reduction efficiency for lower effluent loading ($4.8 \times 10^4 \text{ ton d}^{-1}$) compared to W1 ($3.0 \times 10^5 \text{ ton d}^{-1}$). The significant increase of TN and TP in W2 may also be due to its influent sewerage containing food sludge, which is composed of a relatively large amount of nutrients, whereas food sludge was excluded from W1. Of note, the large-scale sewer system repair plan was initiated in 2004, and was further strengthened after 2006, which led to the successful reduction in point sources loadings.

The bay waters showed a clear seasonal fluctuation in water quality, which was similar to the temporal trends of key river water (and WWTP effluent) variables (Fig. 4). In contrast to the land-based loads, peaks of COD, SS, TN, and TP concentrations in the bay occurred during the summer season when rainfall

increased. This could be due to the rapid input of large amounts of land-based pollution loading into the bay during the rainy season. In addition, the time-lag (or accumulated) effect of river water entering the bay may result in river water variables being strongly correlated with bay surface water after one month (Table 2). This observation may be explained by the nutrient residence time in seawater. The residence time was measured from the mass balance model of ²²⁶Ra, which was conducted as part of this study, to be 53.7 (inner bay) and 23.2 days (entire bay) (Lee et al., 2009).

Both the bay COD and SS significantly decreased from 2005 to 2010 (Fig. 4a and b), with a noticeably dramatic decrease in 2007 (Table S1), immediately following the implementation of the TPLMS. Regression analysis showed a significant decreasing trend for both the surface and bottom layers ($p < 0.001$; Fig. 4); however,

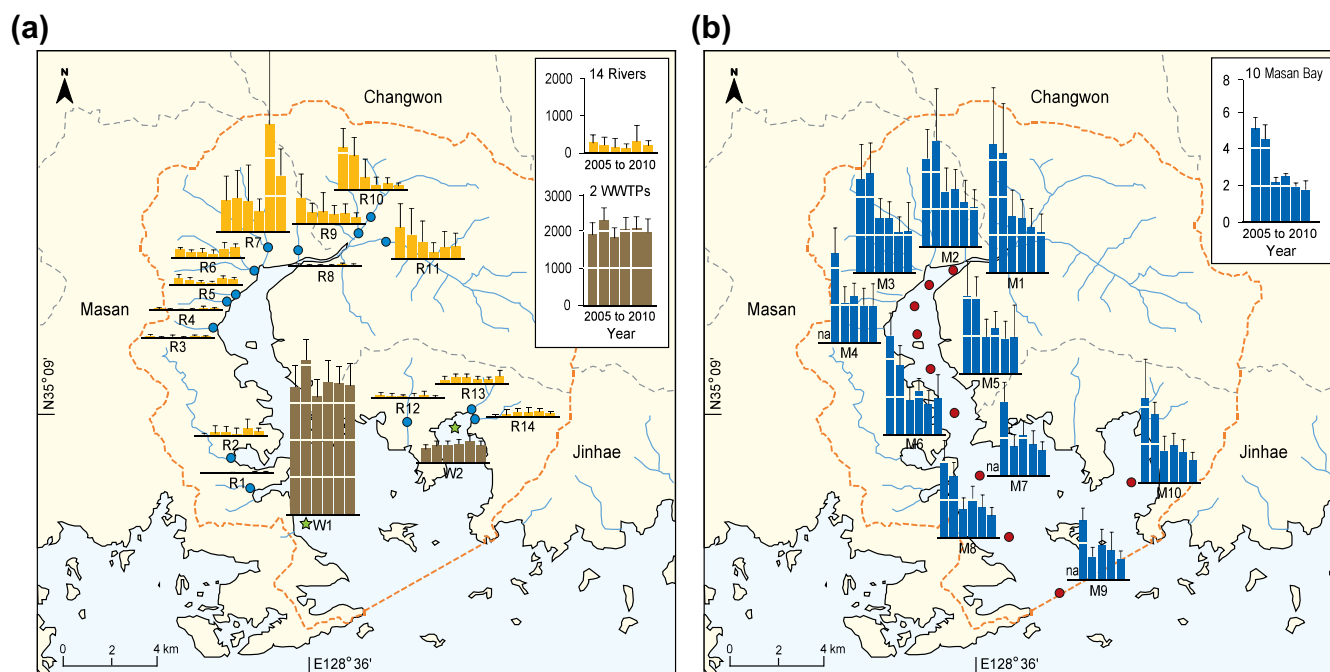


Fig. 5. Spatiotemporal distribution of COD budget in Masan Bay, showing (a) annual mean COD loads (kg d^{-1}) from the inland rivers and WWTPs and (b) annual mean COD concentrations (mg L^{-1}) in the Masan Bay over the six-year period (2005–2010).

the bottom waters showed less variation, indicating a relative lack of flushing in the semi-closed bay system. The TN and TP loads also peaked in the bay during summer; however, no significant temporal trend was observed during the study period, except for TN in the bottom layer ($p = 0.04$, $r^2 = 0.076$).

Although the reduction of TN and TP loading from rivers and WWTPs was evident (Fig. 4c and d), the bay system did not reflect a similar decline, suggesting possible nutrient addition from non-point sources, such as the input of resuspended loading from the bottom sediment or elution from the porewater nutrients (Pitkänen et al., 2001; Almroth et al., 2009). A simple comparison for the nitrogen element between sediment (TN), porewater (dissolved inorganic nitrogen; DIN), and bottom seawater (TN) from selected representative sites located in Masan Bay (M2, M5, M8, and M10) in 2006 versus 2010 indicated that such additions may occur (Table 3). In particular, the DIN porewater flux was as much as 2500 times (mean = 4 times) greater in 2010 compared to 2006. This observation suggests that another pathway of additional nutrient loading exists in the bay. Therefore, future monitoring efforts should incorporate ways to find and monitor these additional pathways, with a greater resolution in time and space.

Among the water quality variables monitored in the Masan Bay system, COD was the most distinctive key parameter, producing consistent seasonal fluctuations over the sampling period, in addition to being a relatively consistent and relevant responsive element to land-based pollution loading. To obtain a fine scale understanding of the relationship and characteristics between land-based pollution loading (rivers and WWTPs) and the response of the receiving seawater (bay), spatiotemporal comparisons of the inland COD load and bay concentrations were further compared (Fig. 5).

The spatial distribution of COD load clearly indicated that pollution loads were concentrated in the innermost rivers (Samho-R7, Naedong-R9, Changweon-R10, and Nam-R11) and WWTPs sites, accounting for 86–91% of total land-based COD loads during the study period (Fig. 5a). In general, COD loads from rivers showed relatively consistent and/or decreasing trends (innermost river sites) over the six-year period. However, the COD load from Samho River (R7) was exceptional, with a dramatic increase (upto 5-fold) in 2009 compared to previous years. The two WWTPs caused major COD loading in the bay, with the cities of Masan and Changweon (W1) being considered as hot point sources for COD discharge

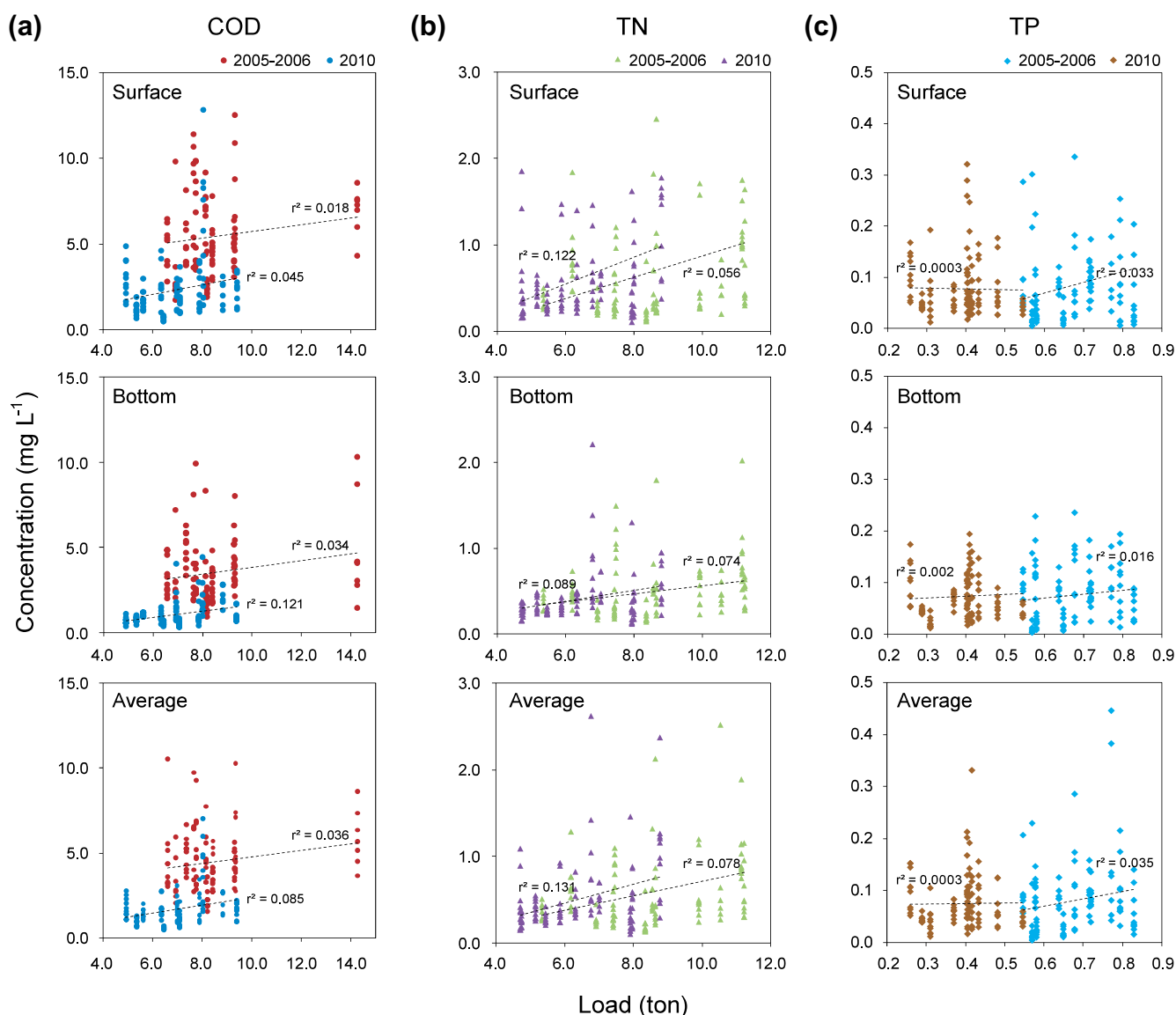


Fig. 6. Plots of (a) COD, (b) TN, and (c) TP concentrations (mg L^{-1}) in seawater against the corresponding land-based load (ton) measured from the inland rivers and WWTPs. Data presented for before (2005–2006) and after (2010) the TPLMS, for comparison.

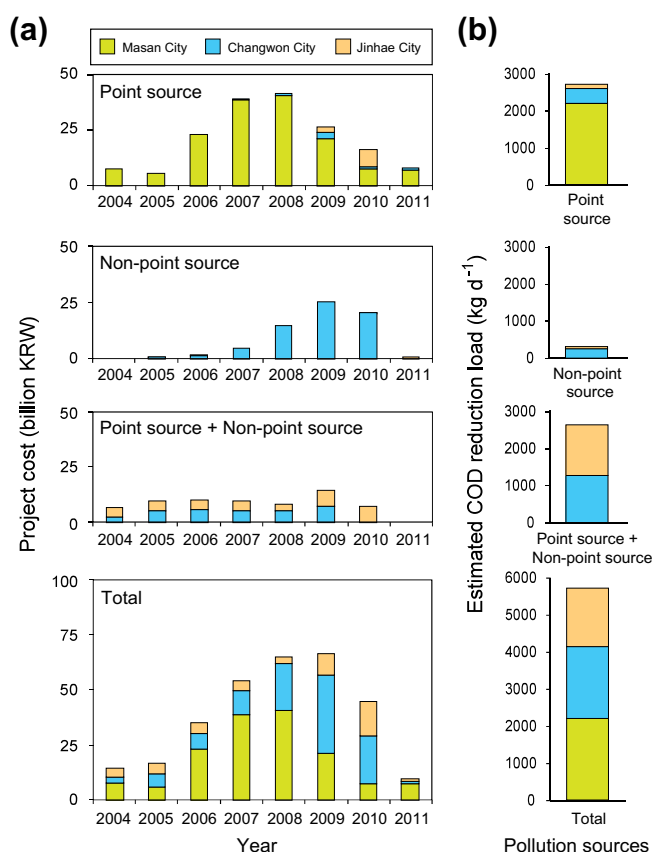


Fig. 7. Annual project cost (billion KRW = million USD) for the pollution abatement measures (namely TPLMS) in the Masan Bay case since 2004, where (a) budget allocations given for source types and/or target cities and (b) estimated COD reduction loads by pollution sources given as well.

throughout the sampling period. While yearly total COD loads did not show inter-annual variation in 2005–2010, COD concentrations in the bay showed a significant decline ($p < 0.01$) at nearly all sampling sites (Fig. 5b), with a noticeably rapid drop in 2007 immediately following the TPLMS implementation.

Furthermore, we used ANCOVA to evaluate the trends in COD response in the bay. This analysis facilitated the comparison of the response variable (i.e., bay COD) in the two groups (2005–2006 versus 2010 data) by considering the variability of other variable, termed covariate (i.e., land-based COD loads) (Fig. 6). More simply an ANCOVA can be utilized to control for the effect of a covariate (land-based COD loads) before making inferences on the TPLMS effects (Flores-López et al., 2010). We found that bay COD (average of surface and bottom water) declined significantly in 2010 (Fig. 6a), when removing the effect of land-based COD loads. This trend indicates that the reduction in COD ($<2 \text{ mgL}^{-1}$) might be attributed to the TPLMS, viz. COD targeted abatement action. Meanwhile TN and TP levels in the bay did not show any significant reduction (Fig. 6b and c) after the implementation of the TPLMS.

During the study period, COD levels declined below the specified target level (2.5 mgL^{-1}), which appeared to indicate the success of the TPLMS, particularly considering the rapid decline in COD loading in response to the point source control of the cities of Masan, Changweon, and Jinhae. Following the establishment of the TPLMS plan in December 2004, the total project cost increased annually until 2009 (Fig. 7a). Based on our results, it is difficult to isolate any single component as directly contributing to the reduction of land-based COD load in the bay. However, it is reasonable to conclude that the reduction of land-based COD load in 2006 (Fig. 5a), followed by the decline in COD concentrations in Masan

Bay since 2007 (Fig. 5b), may primarily result from the point source control of the nearby three cities. Of note, the project cost for non-point source control received greater investment in 2008–2010 (Fig. 7b), after the regulation to reduce pollution loading of the point sources was working relatively well, similar to the previous case in the Hudson River (Brosnan and O’Shea, 1996). This transition suggests that the adaptive management concept (Folke et al., 2005) was carefully applied during the first phase of Masan Bay TPLMS implementation (2007–2011).

The TPLMS plan estimated a reduction in daily COD load of up to about 5700 kg, while the actual reduction in daily load calculated from the concentrations measured in rivers and WWTPs was about 2000 kg. This difference between the estimated and measured COD load, by a factor of ~ 3 , was due to a technical aspect. Specifically, the estimated COD reduction utilized the 95-percentile of COD load values in all years, while the actual reduction was calculated from the yearly average of COD load values. In addition, unknown factors exist, such as the influence of groundwater (Lee et al., 2009; Kim et al., 2010). In addition, certain changes in socio-economic conditions over time could also cause variation in the reduction of COD levels in target areas. While this calculation (viz., using 95-percentile load values to estimate a COD reduction target) has certain limitations in data handling, it is very useful to reach the TPLMS’s goal as long as policy targets are set based on precautionary approaches, incorporating evidence-based site-specific information.

The present study aimed to evaluate the contribution of the TPLMS plan and implementation, the goal of which was the ‘reduction’ of the land-based pollution loads to Masan Bay by primarily targeting COD loading. Overall, the survey datasets assimilated over the six-year period provided a relatively clear explanation about the mass balance between land-based pollution loads (rivers and WWTPs) and the receiving bay water quality. The policy goal of the TPLMS, specifically COD reduction in Masan Bay, seemed to be achieved, and the key contributors towards successfully managing COD loadings comprised (1) sewer system improvement, (2) technical upgrading of the two WWTPs, and (3) an intensive river clean-up. While, land-based COD and SS loading was successfully managed through these management practices, the reduction of TN and TP remains a challenge due to their complex behavioral mechanisms in the aquatic environment. The second phase of the TPLMS plan was to direct attention towards the control of non-point sources (e.g., groundwater monitoring), which may potentially contribute to the reduction of TN and TP in subsequent years. In addition, the introduction of advanced sewage treatment (e.g., the A₂O system with membrane reactor) and Low Impact Development practices to control diffuse sources should be pursued in the second phase. Overall, such abatement action plans should consider long-term practices towards improving the sustainability of the TPLMS.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.marpolbul.2011.11.011.

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Table S1. Data summary for water quality variables (annual mean and range) in rivers, wastewater treatment plants (effluent), and Masan Bay, Korea, in 2005–2010

| Sites / Samples | Year | Number of Survey | Data | Temp. | Salinity | DO | COD | BOD | SS | TN | TP | NO ₃ ⁻ -N | NH ₄ ⁺ -N | PO ₄ ³⁻ -P | SiO ₂ -Si | TOC | POC | DOC | Chl-a | Flow | | |
|-----------------------------|------|------------------|-------------------|---------------------|---------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|---------------------------------|---------------------------------|----------------------------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|---------------------------|---------------------------|-------------------------|
| | (y) | (n) | | (°C) | (psu) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg L ⁻¹) | (mg m ⁻³) | (ton d ⁻¹) | | |
| Rivers (n=14, R1-R14) | 2005 | 7 | Min.-Max. Mean | 5.2 - 23.4 15.8 | 0.2 - 0.6 0.4 | 3.4 - 6.3 4.8 | 9.2 - 21.3 12.6 | 6.2 - 20.5 11.3 | 8.7 - 21.9 15.2 | 7.1 - 16.5 11.3 | 0.22 - 0.96 0.65 | nd ^a - 2.09 1.30 | nd - 9.07 4.83 | na ^b | na | 6.86 - 16.81 13.32 | na | 2.58 - 6.85 4.93 | na | | | |
| | 2006 | 17 | Min.-Max. Mean | 6.5 - 27.1 17.9 | 0.1 - 1.0 0.3 | 4.3 - 8.1 5.9 | 4.2 - 23.3 10.9 | 3.3 - 20.3 9.2 | 5.3 - 28.2 14.1 | 5.6 - 15.6 10.7 | 0.22 - 1.41 0.67 | 0.98 - 3.74 2.22 | 0.99 - 9.86 4.23 | na | na | 2.80 - 17.16 8.07 | na | 1.72 - 6.60 4.56 | na | | | |
| | 2007 | 18 | Min.-Max. Mean | 5.9 - 25.3 17.2 | 0.1 - 1.1 0.4 | 4.2 - 8.8 6.3 | 5.3 - 21.1 11.0 | 4.9 - 20.0 8.2 | 4.7 - 28.0 12.4 | 5.7 - 21.3 11.3 | 0.28 - 2.46 0.62 | 0.03 - 3.01 1.95 | 0.33 - 9.14 2.88 | 0.02 - 0.55 0.25 | 4.71 - 12.21 6.87 | 5.26 - 12.65 8.89 | 1.45 - 9.80 5.13 | 2.56 - 5.88 3.76 | na | | | |
| | 2008 | 18 | Min.-Max. Mean | 4.2 - 26.2 18.3 | 0.1 - 1.1 0.4 | 3.8 - 8.0 5.9 | 5.3 - 21.4 9.7 | 4.4 - 17.4 7.6 | 4.1 - 15.4 8.9 | 3.7 - 12.8 8.1 | 0.24 - 0.82 0.54 | 0.69 - 2.63 1.62 | 1.39 - 10.05 2.96 | 0.09 - 0.51 0.33 | 2.03 - 8.91 5.88 | 2.99 - 9.25 4.69 | 0.68 - 2.88 1.41 | 0.84 - 6.37 3.09 | na | | | |
| | 2009 | 19 | Min.-Max. Mean | 5.7 - 26.1 19.4 | 0.1 - 1.6 0.5 | 4.5 - 7.4 5.5 | 4.5 - 19.8 9 | 4.0 - 11.6 7.6 | 6.7 - 28.4 13.4 | 1.3 - 20.8 10.1 | 0.20 - 3.07 0.64 | 0.83 - 4.30 2.02 | 0.66 - 7.59 3.65 | 0.04 - 0.83 0.31 | 1.73 - 7.77 5.38 | 1.63 - 6.76 3.62 | 0.43 - 1.56 0.86 | 1.20 - 5.32 2.86 | na | | | |
| | 2010 | 19 | Min.-Max. Mean | 6.7 - 27.0 19.0 | 0.1 - 1.9 0.5 | 3.2 - 6.9 5.0 | 4.3 - 16.2 7.9 | 3.0 - 12.4 6.5 | 3.9 - 19.7 9.8 | 5.3 - 10.2 7.3 | 0.20 - 0.88 0.52 | 1.08 - 2.70 1.74 | 1.12 - 6.28 3.17 | 0.08 - 0.36 0.21 | 1.88 - 6.81 3.92 | 3.31 - 9.13 6.41 | 2.66 - 6.94 4.46 | 0.65 - 3.25 1.95 | na | | | |
| WWTP 1 effluents (W1) | 2005 | 365 | Min.-Max. Mean | na na | na na | na na | 8.9 - 16.9 13.1 | 8.7 - 17.3 13.7 | 4.0 - 15.0 8.1 | 9.4 - 20.7 15.6 | 0.73 - 1.78 1.51 | na na | na na | na na | na na | na na | na na | na na | na na | 198528 - 447107 268240 | | |
| | 2006 | 365 | Min.-Max. Mean | na na | na na | na na | 10.1 - 16.8 14.7 | 6.7 - 17.2 12.0 | 4.2 - 15.6 11.7 | 7.4 - 20.1 14.8 | 0.35 - 1.80 1.43 | na na | na na | na na | na na | na na | na na | na na | na na | 213460 - 476517 287539 | | |
| | 2007 | 365 | Min.-Max. Mean | na na | na na | na na | 6.4 - 15.3 10.9 | 1.3 - 8.9 4.2 | 1.2 - 9.0 4.4 | 3.1 - 18.0 11.3 | 0.26 - 1.78 1.35 | na na | na na | na na | na na | na na | na na | na na | na na | 213593 - 466514 297894 | | |
| | 2008 | 365 | Min.-Max. Mean | na na | na na | na na | 8.1 - 15.8 12.2 | 0.1 - 7.9 4.0 | 0.8 - 5.0 3.2 | 4.0 - 17.4 13.2 | 0.16 - 1.40 1.11 | na na | na na | na na | na na | na na | na na | na na | na na | na na | 231159 - 452192 296725 | |
| | 2009 | 365 | Min.-Max. Mean | na na | na na | na na | 7.2 - 16.0 12.3 | 0.4 - 8.0 3.9 | 0.7 - 5.0 3.1 | 6.2 - 16.9 12.7 | 0.22 - 1.39 0.98 | na na | na na | na na | na na | na na | na na | na na | na na | na na | 222490 - 468659 294444 | |
| | 2010 | 365 | Min.-Max. Mean | na na | na na | na na | 7.6 - 15.9 12.0 | 0.6 - 7.9 4.4 | 1.1 - 4.9 3.4 | 5.2 - 17.4 11.8 | 0.14 - 1.40 0.72 | na na | na na | na na | na na | na na | na na | na na | na na | na na | 70990 - 553670 295831 | |
| WWTP 2 effluents (W2) | 2005 | 365 | Min.-Max. Mean | na na | na na | na na | 3.0 - 14.8 8.7 | 2.9 - 15.5 8.1 | 1.2 - 16.0 5.3 | 7.6 - 32.4 15.6 | 0.63 - 2.48 1.48 | na na | na na | na na | na na | na na | na na | na na | na na | na na | 24615 - 115696 44806 | |
| | 2006 | 365 | Min.-Max. Mean | na na | na na | na na | 4.4 - 14.5 10.4 | 0.7 - 14.9 8.5 | 0.8 - 14.0 6.0 | 3.3 - 26.6 13.9 | 0.15 - 2.34 1.09 | na na | na na | na na | na na | na na | na na | na na | na na | na na | 30645 - 171102 47024 | |
| | 2007 | 365 | Min.-Max. Mean | na na | na na | na na | 6.1 - 17.5 11.3 | 1.0 - 18.0 7.4 | 2.0 - 18.8 7.3 | 2.9 - 19.5 11.5 | 0.10 - 3.39 0.79 | na na | na na | na na | na na | na na | na na | na na | na na | na na | 22078 - 135837 44435 | |
| | 2008 | 365 | Min.-Max. Mean | na na | na na | na na | 7.8 - 18.2 11.3 | 1.2 - 9.8 5.9 | 2.0 - 9.8 6.9 | 4.3 - 27.0 12.9 | 0.22 - 2.15 0.80 | na na | na na | na na | na na | na na | na na | na na | na na | na na | na na | 30334 - 107822 45740 |
| | 2009 | 365 | Min.-Max. Mean | na na | na na | na na | 5.5 - 18.8 11.1 | 1.9 - 7.8 5.1 | 0.8 - 7.6 4.9 | 3.6 - 29.9 13.5 | 0.01 - 0.87 0.24 | na na | na na | na na | na na | na na | na na | na na | na na | na na | na na | 37988 - 158590 54536 |
| | 2010 | 365 | Min.-Max. Mean | na na | na na | na na | 6.9 - 13.6 9.3 | 0.8 - 7.3 4.1 | 0.8 - 4.0 1.9 | 2.0 - 16.3 9.9 | 0.09 - 0.88 0.17 | na na | na na | na na | na na | na na | na na | na na | na na | na na | na na | 30546 - 135392 52017 |
| Masan Bay (n=10, M1-M10) | 2005 | 4 | Min.-Max. Mean | 14.3 - 24.7 21.2 | 28.4 - 32.5 30.3 | 4.4 - 7.4 6.3 | 3.4 - 5.9 4.9 | na | 39.4 - 51.6 44.8 | 0.37 - 1.32 0.78 | 0.09 - 0.18 0.12 | 0.05 - 0.14 0.09 | 0.20 - 0.55 0.32 | na | 0.5 - 0.9 0.6 | 1.82 - 5.88 3.89 | 0.29 - 1.19 0.73 | 1.53 - 4.69 3.16 | 9.0 - 12.9 11.7 | | | |
| | 2006 | 8 | Min.-Max. Mean | 6.36 - 25.5 17.0 | 20.7 - 33.0 29.5 | 5.9 - 9.6 7.4 | 2.2 - 5.3 4.3 | na | 5.5 - 37.5 17.1 | 0.22 - 1.03 0.53 | 0.02 - 0.10 0.07 | nd - 0.32 0.07 | 0.03 - 0.23 0.12 | na | 0.1 - 1.3 0.6 | 1.90 - 4.07 3.09 | 0.45 - 1.29 0.92 | 1.44 - 2.78 2.18 | 3.7 - 24.8 15.9 | | | |
| | 2007 | 10 | Min.-Max. Mean | 7.4 - 24.3 19.4 | 28.4 - 34.2 32.1 | 4.2 - 9.5 6.4 | 0.8 - 3.5 2.1 | na | 4.1 - 13.2 7.5 | 0.30 - 0.74 0.51 | 0.04 - 0.12 0.09 | 0.01 - 0.14 0.05 | 0.04 - 0.25 0.10 | na | 0.2 - 1.4 0.6 | 0.81 - 4.06 2.00 | 0.61 - 2.31 1.35 | 0.20 - 1.75 0.64 | 4.4 - 28.2 12.1 | | | |
| | 2008 | 11 | Min.-Max. Mean | 5.0 - 24.7 18.8 | 31.1 - 33.4 32.4 | 3.9 - 12.2 7.3 | 0.9 - 3.1 2.4 | na | 5.0 - 27.3 13.8 | 0.35 - 0.70 0.45 | 0.04 - 0.20 0.08 | nd - 0.09 0.02 | 0.10 - 0.15 0.08 | na | 0.1 - 1.0 0.5 | 0.95 - 1.80 1.52 | 0.09 - 0.37 0.21 | 0.86 - 1.43 1.32 | 3.2 - 30.6 11.9 | | | |
| | 2009 | 12 | Min.-Max. Mean | 7.4 - 23.8 17.5 | 22.9 - 34.4 31.3 | 4.6 - 12.1 7.4 | 0.9 - 3.3 1.9 | na | 4.7 - 20.8 9.1 | 0.26 - 1.06 0.48 | 0.03 - 0.09 0.06 | nd - 0.30 0.06 | 0.01 - 0.26 0.12 | na | 0.2 - 2.3 0.7 | 1.83 - 5.72 2.76 | 0.65 - 4.04 1.41 | 1.18 - 1.68 1.35 | 0.2 - 72.0 13.8 | | | |
| | 2010 | 11 | Min.-Max. Mean | 9.5 - 24.7 18.8 | 26.1 - 32.8 30.3 | 3.8 - 10.5 6.8 | 0.6 - 3.8 1.7 | na | 5.0 - 20.2 10.0 | 0.27 - 1.04 0.51 | 0.04 - 0.12 0.07 | 0.02 - 0.39 0.08 | 0.04 - 0.21 0.12 | na | nd - 1.7 0.8 | 1.68 - 4.00 2.74 | 0.49 - 2.13 1.34 | 1.18 - 1.86 1.40 | 0.4 - 4.0 1.7 | | | |

^a nd: not detected.^b na: not analyzed.