Numerical Assessment of Total Nitrogen (Tn) Load Discharged from Rivers into Harima-Nada, the Seto Inland Sea, Using A Numerical Coupled Hydrological-Water Quality Model

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Abstract. Harima-Nada region, located in the east of Seto Inland Sea, is a semi-enclosed sea-area next to Osaka Bay that has been suffering a sustained oligotrophication process during the past 25 years. A hydrogeological model, coupled with a water quality model, was applied to assess the numerical discharge of freshwater and total nitrogen (TN) load from rivers into the sea. The model was validated for the main rivers from the northern and southern regions (the Kako River and the Yoshino River, respectively). The obtained results showed that the main contribution of freshwater and TN load to the sea comes from Hyogo Prefecture Rivers. The type of activities developed on each watershed showed to be substantially more relevant on TN load contribution than the size itself of the considered watershed. For the eight years considered in this study (2009 - 2016), the annual average of TN discharged into Harima-Nada was almost constant, while the volume of discharged freshwater showed more variations. Additionally, the sustained decrement in nitrogen concentration in Harima-Nada seemed not to be directly bonded with riverine TN load, suggesting that nitrogen availability is closely related to sea nutrients mechanisms more than to TN inland contribution.

1. Introduction
Water pollution is one of the most critical consequences of economic and industrial development for countries all around the world. The effects are so broad as the different types of existent pollutants and could affect as much water ecosystems as human health. During the late '60s, Japan experienced an intensive industrial development that not only positioned it on the top of world economies but also caused big environmental problems all around the Japanese archipelago.

Because of the geographical advantages of the Seto Inland Sea coastal areas, the region experienced a sustained development during the past century and nowadays locates around 30% of Japanese manufacturing industries and 28% of Japan's population [1]. Eutrophication problems caused for the intensive discharge of nutrients on its waters promoted the establishment of a special law in 1973, which became in 1979 the Law Concerning Special Measures for Conservation of the Environmental of the Seto
Inland Sea, intending to regulate organic matter and nutrients effluent's discharge. The development of rigorous environmental policies to revert the eutrophication, plus the construction of multiple dams in most of the rivers of the region, have caused since then a sustained decrement in nutrients discharge from rivers in the area [1, 2].

Harima-Nada is one of the numerous basins of the Seto Inland Sea, and as a consequence of its proximity to Osaka Bay, the enclosed geography of the region, and the numerous rivers basins that surround the area, it is strongly influenced by riverine nutrients and freshwater discharged (figure 1) [2]. The eutrophication problems were partially reverted after the reinforcement of environmental policies, but new problems arose. A sustained oligotrophication process has been occurring during the past 25 years in many areas of the Seto Inland Sea, but the problem is particularly severe in Harima-Nada and the southern side of Osaka Bay, where the values of Dissolved Inorganic Nitrogen (DIN) have suffered a decrement of around 0.2 µM per year since the '90s [3, 4]. This continuous depletion of nutrients has caused a pronounced reduction in fishery, nori production, and has affected many pelagic species of the region, putting them in an endangered situation [1, 3].

Much research and studies have been done in this local area to clarify nutrients dynamics, some focusing on numerical estimations, others based on observed data series of the region, and some of them combining both types of techniques [1, 4, 5, 6, 7, 8]. However, the nutrient load from rivers is not always estimated in detail. The reason is that most of the existing data on nutrients were measured at non-flooding conditions, and the effects of heavy rain and flooding could not be fully evaluated. The present work aims to introduce a quantification of the freshwater and TN land-derived discharged into the sea, using a numerical coupled hydro-chemical model (CHCM) from the main rivers of the region between 2009 and 2016. The CHCM was able to reproduce the changes in flow rate and TN load at the time of rain and flood events, which was confirmed with the validation of the model in the most important watersheds of northern and southern areas of Harima-Nada (Kako River and Yoshino River). Additionally, nonpoint and point source nitrogen contributions were considered to quantify the TN fluxes of the entire study area.

**Figure 1.** Harima-Nada location, river domains studied, and location of monitoring stations.
2. Methodology

2.1. Study site and modeling framework
Harima-Nada locates on the east side of the Seto Inland Sea, between the Bisan Strait in the west, the Osaka Bay on the eastern side, and the Kii Channel in the south (connected by the Naruto Strait), as figure 1 shows. The considered domains were each watershed, for the eleven rivers that directly discharge in Harima-Nada from Hyogo and Kagawa Prefectures, and the Yoshino River System basin (figure 1). The horizontal resolution used for calculations was 1x1 km, and the number of grids was dependent on the particular river watershed size.

Geographical datasets were obtained from the website of the Ministry of Land, Infrastructure, and Transport of Japan (MLIT), with a resolution of 100x100 m for river watershed and 1x1 km for elevation and land usage. Meteorological information used corresponds to the Automated Meteorological Data Acquisition System (AMeDAS) database from the Japan Meteorological Agency. The observed data used to perform water discharge validation was from the Water Information System of MLIT. For Kako and Yoshino Rivers, hourly discharged water data was available. For TN concentration, the monthly load was also obtained from the Water Information System. Additional information on the TN load to perform validation was complemented with previous research and monitoring campaigns conducted by Hyogo Prefectural Institute of Environmental Sciences (HIES) [9].

A total of 501 point sources (PS) was considered in the Prefecture to estimate the TN load of each river. The number of point-sources considered depended on population density and the economic activities of each site. HIES also provided the location and daily/monthly TN load of point sources in Hyogo Prefecture.

2.2. Numerical coupled hydro-chemical model (CHCM)
A schematic layout of the CHCM used in this research is shown in figure 2a. The model consists of two big coupled blocks, in which the first one is a hydrological model, followed by a water quality model, each of them composed of different submodules.

2.2.1. Hydrological model. This model is based on the hydrological model proposed for Kojiri et al. (2008) [10]. For each study site, the surface was divided into a 1x1 km horizontal mesh to generate a calculation grid. Meteorological input data (rain, solar radiation, etc.) was assigned per cell to the correspondent values in the closest meteorological station. The slope-terrain model was coupled to a river channel model to obtain river water level and river water discharge. In the vertical, four layers were considered to represent the hydraulic behavior of the soil in the considered basin.

Four different types of land use (farm, forest, urban area, and rice field or paddy) and their respective ratio in each cell were used to estimate the hydraulic characteristics of each cell. Farms, forests, and urban areas were modeled using a Kinematic Wave Model, while for paddy, a tank model was used due to the specific flow conditions that this type of field has. Evapotranspiration and vegetation canopy was also added to the model to make the estimations more realistic. A scheme of the hydrological model is presented in figure 2b.

2.2.2. Water quality model. The water quality model has its basis on the cesium transport model developed by Ikenoue et al. (2020) [11], and it was configured to give an estimation of TN load on each cell of the mesh. The used inputs were the hydrological model results, land use information and wet deposition parameters for nonpoint source contribution, and point-source observed data (industries, wastewater treatment plants, sewage, etc.). Nitrogen transport was calculated considering advection and diffusive mechanisms and the lateral inflows caused by the terrain slope (runoff), according to the land usage ratio on each cell. Fertilizer loads were considered temporarily, according to seasoning fertilization, and were estimated as a constituent of runoff nitrogen load.
2.3. Model validation

The model validation was performed separately for the north and the south of Harima-Nada because the human activities, population density, and meteorological conditions of the inland areas are slightly different. Tuning was performed on the most significant contributors of the north and south areas (the Kako River and the Yoshino River), respectively (figure 1). Validation was conducted between 2009 and 2016 for water discharge in six points, three on the main river, and three in tributary rivers. For TN load, six points on the main river and one point on a tributary river were used (figure 1). In Shikoku Island, it was decided to use the Yoshino River System to perform calibration and validation of the model. The Yoshino River does not discharge directly into Harima-Nada, but it has the biggest and more important watershed of Shikoku Island, with an approximate basin area of 3750 km² (figure 1). Based on the previous considerations, it is the most representative river of the region, and water discharge validation was conducted in three points (figure 1), to extend its use to the Kagawa Prefecture rivers. The values of TN load were estimated for all Shikoku Island rivers without considering PS contribution due to the inaccessibility to any PS load data in the region.

Model performance was evaluated for both rivers comparing observed data with simulation results. Daily and monthly comparisons were statistically performed following the guidelines proposed by Moriasi et al. (2007) [12] on discharged water for Kako and Yoshino Rivers. Additionally, TN load was validated using the same guidelines, between April 2010 and March 2012, in the Kako River.

3. Results and discussion

3.1. Model performance

The statistical evaluation for the CHCM was based on the indices recommended in Moriasi et al. (2007) for monthly temporal distribution. The results for the coefficient of determination (R²), Nash-Sutcliffe efficiency (NSE), percent bias (PBIAS), and the root mean square error-observation standard deviation (RSR) are presented in table 1 for discharged volume and TN load.

For discharged water, the degree of collinearity between observed and simulated results is very high, as it is shown in figures 3a and 3b. The model overestimates observed values by around 25%, as the negative values of PBIAS in both rivers show, but all the indices indicate a good performance (table 1.).

For TN load, a high correlation between observed and calculated monthly average was also observed (figure 4c), but the model underestimates the computed values by around 14% (positive value of PBIAS).
The obtained results (table 1.) show a very good model performance for discharged water and TN load according to the model evaluation guidelines proposed for Moriasis et al. (2007).

3.2. North Harima-Nada, hyogo prefecture
The results are shown in table 2 for all the rivers considered in this study. The river flow (Q) of the Kakogawa was $6.55 \times 10^6$ m$^3$/day, representing about 50% of the total rivers calculated. It was observed that the biggest the watershed area, the highest values for Q. Additionally, Q values vary from year to year, as it could be seen in figure 4a.

TN load results were obtained by considering the land usage in the basin with PS contribution and without PS contribution (non-PS contribution). The Kako River watershed is the most important contributor, and it maintains the same trend as for Q, being responsible for around 50% of the Prefecture's TN load into the sea. For TN load, the correlation with the basin's size does not seem as direct as for discharged (table 2). For example, the Mihara River results show that the annual and average TN load discharged is the highest per unit area of all the Prefecture (20% of Kako river TN load). Still, Mihara's watershed size is 14 times smaller than the Kako River basin.

The PS contribution is very significant in Kako and Ichi River basins, where it represents a respective increment of more than 30% and around 20%, while for the rest of the rivers is less than 10%, and for the Mihara River, represents less than the 1.0%. Not significant annual variations on the TN load can be observed from the Prefecture, being the average nitrogen discharge around 3500 tons/year. (figure 4b).

3.3. South Harima-Nada, Kagawa prefecture and Yoshino river system
The obtained results for Q in Kagawa Prefecture and Yoshino River follow the same trend for Hyogo Prefecture, showing a direct size-discharge correlation. Kagawa Prefecture watersheds are comparatively smaller than those in Hyogo Prefecture, having similar sizes than the basins of Awaji Island. The total freshwater discharged from Kagawa Prefecture Rivers accounts for less than 10% of Hyogo Prefecture Rivers. On the other hand, the Yoshino River watershed has more than double the Kako River area, and it discharges around the double of freshwater to the sea than all Hyogo Prefecture rivers together. Annual variations in Q are more pronounced for Yoshino River than for any of the other studied rivers for the considered years, as shown in figure 4c.

The results for Kagawa Prefecture on the southern area show that the nitrogen river contribution to the sea is around 20% of Hyogo's TN load contribution and does not show any specific trend with the watershed size as for Hyogo Prefecture. On the contrary, the Yoshino River results are around the same values as the total TN load discharged from Hyogo Prefecture (table 2). The total nitrogen load from Shikoku Island shows the same behavior as for Hyogo Prefecture, being practically constant for the whole period (figure 4d).

| Table 1. Statistical evaluation of monthly model performance. |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| Parameter       | River name      | Obs (m$^3$/s)  | Sim (m$^3$/s)  | R$^2$           | RSR             | NSE             | PBIAS           |
| River Discharge | Kako River      | 48.6           | 60.4           | 0.96            | 0.37            | 0.86            | -24.3           |
| (2009-2016)     | Yoshino River   | 151.6 (m$^3$/s) | 187.5 (m$^3$/s) | 0.93            | 0.27            | 0.93            | -23.7           |
| TN load         | Kako River      | 84.8 (kg/s)  | 72.8 (kg/s)   | 0.97            | 0.32            | 0.89            | 14.2            |
| (April 2010-March 2012) | | | | | | | |
Figure 3. Validation results for discharged freshwater in Kako River (a) and Yoshino River (b), and TN validation results in one of the stations of Kako River (c).

Table 2. River simulation results.

| Region         | River name          | River area (Km²) | $\bar{Q}$ a (x10⁶ m³/day) | No. of PS considered | TN load (Ton/year) a | % of PS contribution on TN load |
|----------------|---------------------|------------------|-----------------------------|----------------------|----------------------|---------------------------------|
| Hyogo Prefecture | Kako                | 1730.0           | 6.55                        | 273                  | 1748.5               | 2411.0                          | 32.2%                           |
|                 | Ibo                 | 810.0            | 3.30                        | 63                   | 437.9                | 481.6                           | 10.0%                           |
|                 | Chikusa             | 730.0            | 2.53                        | 63                   | 386.3                | 420.0                           | 8.8%                            |
|                 | Ichi                | 496.0            | 2.08                        | 67                   | 305.4                | 362.0                           | 18.6%                           |
|                 | Yumesaki            | 205.0            | 0.71                        | 17                   | 120.8                | 125.6                           | 4.0%                            |
|                 | Mihara              | 123.7            | 0.65                        | 18                   | 517.2                | 521.6                           | < 1.0%                          |
|                 | Gunge               | 26.3             | 0.06                        | 23                   |                      | 23.5                            |                                 |
| **Total**       |                     | **4121.0**       | **15.88**                   | **501**              | **3539.6**           | **4321.8**                      | **22.0%**                       |
| Kagawa Prefecture | Kasuga/Shin system | 131.9            | 0.60                        |                      |                      | 414.4                           |                                 |
|                 | Minato              | 51.6             | 0.23                        |                      |                      | 44.5                            |                                 |
|                 | Kamobe              | 68.0             | 0.22                        |                      |                      | 132.9                           |                                 |
|                 | Tsuda               | 43.7             | 0.15                        |                      |                      | 71.1                            |                                 |
| **Total**       |                     | **295.2**        | **1.2**                     |                      |                      | **662.9**                       |                                 |
| Yoshino system  | Yoshino             | 3750.0           | 18.7                        |                      | 3450.0               |                                 |
Figure 4 Representation of annual averages of discharged freshwater and TN load per river of Hyogo prefecture (a and b) and Shikoku Island (c and d).

4. Conclusions
CHCM model showed a good performance reproducing observed values of discharged water and TN load for the most representative watersheds of the Harima-Nada region. Results for discharged water confirmed that the Kako River is the most important direct-contributor on freshwater to the area. Yoshino River discharges every year into the sea, more freshwater than all Harima-Nada rivers, so it is necessary a most profound understanding of the percentage of freshwater that is exchanged through the Naruto Strait. For the studied area, results showed that there is a direct correlation between watersheds size and discharged volume. Also, meteorological conditions showed to have more incidence on Shikoku Island rivers than in Hyogo Prefecture, as it could be seen in figures 4a and 4c.

TN load results for Hyogo and Shikoku Island showed that annual riverine loads into the sea are practically constant for the considered years. It is also noticeable a high degree of independence between the TN load that is annually discharged and the meteorological events that directly affect the discharge volumes. Point source contribution showed to be strongly linked to the activities developed in the watershed. For the Kako River basin, which locates significant urban areas, the PS contribution represents around 30% of the total load, while for farming areas like the Mihara basin, the land usage is the principal source of nitrogen load. Based on this, and because of the size and type of economic activities develop on
Kagawa Prefecture basins, we assume that the results obtained are acceptable, even when validation could not be conducted. Hyogo Prefecture rivers account for approximately 80% of the nitrogen directly discharged into Harima-Nada, which is according to the results of previous research in the region[2, 4]. The TN load from the Yoshino River is comparable to the entire TN load directly discharged into Harimanda, reinforcing the importance of quantifying the nutrients exchange in the Naruto Strait. The sustained oligotrophication process in Harima-Nada cannot be explained under the consideration of an annual decrement in riverine nitrogen loads during the past decade. On the other hand, sea nutrients mechanisms and dynamics seem to be more important to explain nutrient variations than the riverine discharge for the area.

5. References

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