hydrology

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The Seasonality of Phosphorus Concentrations in a Suburban River Catchment by the Assessment of the Dissolved and Particulate Phosphorus Species

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Abstract

Phosphorus (P) contamination in surface water is a major global concern affecting most suburban river catchments worldwide due to the discharge of industrial effluents and urban sewage, a consequence of rapid urbanization and industrialization. The objective of this study was to investigate the concentrations and seasonal patterns of various phosphorus (P) species in water from the Yamato River. Total phosphorus (TP), total particulate phosphorus (TPP), and particulate inorganic phosphorus (PIP) were determined using the molybdenum blue method; particulate organic phosphorus (POP) and total dissolved phosphorus (TDP) were calculated by difference; and particulate phosphorus was fractionated using the sedimentary phosphorus sequential extraction (SEDEX) method. Average TP concentration was 0.31 mg/L (0.14 mg/L–0.77 mg/L), TPP was 0.10 mg/L (0.03 mg/L–0.47 mg/L), PIP was 0.07 mg/L (0.02 mg/L–0.46 mg/L), particulate organic phosphorus (POP) was 0.02 mg/L (0.00 mg/L–0.24 mg/L), and total dissolved phosphorus (TDP) was 0.22 mg/L (0.00 mg/L–0.71 mg/L). There was a decrease in TP levels with increased water levels in summer, possibly due to dilution effects, although other factors may be influencing TP levels. TDP consistently dominated over TPP but was more pronounced in low-flow conditions in winter compared to summer. PIP exceeded POP on most dates, although they both increased with the increased water levels witnessed in the summer. Fe-P was the dominant P species, increasing alongside Ca-P in the summer (irrigation period of paddy fields). The study indicated that the phosphorus pollutants in the river water were inorganic in nature and may have originated from the leaching of inorganic fertilizers from nearby paddy fields, urban surface runoff, wastewater discharge, and construction debris carried as erosion products.

Keywords: eutrophication; phosphorus forms; industrialization; urbanization SEDEX; particulate phosphorus

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Academic Editor: Ioannis Panagopoulos

Received: 2 October 2026
Revised: 25 October 2026
Accepted: 27 October 2026
Published: 30 October 2026

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1. Introduction

Phosphorus (P) is a vital element for humans and other living organisms [1–6]. It is a component of the nucleotides (adenosine triphosphate ATP and adenosine diphosphate ADP) required for cell energy transduction, cell membranes, and metabolism control [1,5,7].

In agriculture, it is critical for sustaining and improving crop yield [8–10]. In contrast, phosphorus is one of the pollutants that have contaminated surface and groundwater in most megacities worldwide alongside nitrates, trace metals, and chlorides, among others. This phenomenon is because megacities often tend to be heavily urbanized and industrialized, and these pollutants result from industrial effluents, agricultural runoffs, and urban sewage pathways that find their way into the water system [11]. Moreover, managing phosphorus is essential in aquatic habitats to prevent or reduce eutrophication [4,6,12–14]. Eutrophication often leads to deterioration of surface water quality, which has become a critical environmental issue for both coastal and inland water bodies [10,14–20]. Transportation of phosphorus in water bodies is rather easy, owing to its adsorption onto particles or sediments (sources and sinks of water-soluble phosphorus) throughout the aquatic system [4,9,21,22]. Watershed factors such as geology, land use, and hydrometeorological conditions substantially regulate the concentrations of phosphorus in freshwater ecosystems [23–25].

The physicochemical fractionation of phosphorus, that is, dissolved inorganic phosphorus (DIP), soluble reactive phosphorus (SRP), dissolved organic phosphorus (DOP), colloidal inorganic phosphorus (CIP), particulate inorganic phosphorus (PIP), and particulate organic phosphorus (POP), according to Yang et al. and others [6,13,21,26–28], determines its fate and its subsequent impact on the environment. Particulate phosphorus (PP) is the fraction of phosphorus retained by a 0.45 μm filter as dissolved phosphorus (DP) passes through [29,30].

The identification of phosphorus sources, its cycling, and its transformations is more difficult than other nutrients [31,32]. This challenge is primarily due to various factors, including (a) the low concentration of readily dissolved phosphorus compared to particulate phosphorus; (b) the complex interactions involved in phosphorus cycling. Additionally, the varying nature of phosphorus transformation between its organic and inorganic forms is another factor contributing to this difficulty; (c) simultaneously occurring biotic and abiotic reactions lead to absorption and desorption, dissolution and precipitation, and cycling of phosphorus under various conditions [33,34]; and (d) different organisms employ various strategies for the uptake and cycling of phosphorus, depending on the concentrations and compositions of organic and inorganic phosphorus compounds. Despite these challenges, anthropogenic activities, such as mining and processing phosphorus minerals to produce phosphorus-containing fertilizers, have been associated with significant phosphorus deposition in the ecosystem [35–37]. These fertilizers and animal manures are overapplied to cultivated lands, which often contain ample amounts of phosphorus [19,38–41]. Consequently, leaching of these excessive amounts of phosphorus, along with overland flow, surface runoff, and soil erosion, as well as the discharge of sewage into water bodies, constitutes the major sources of phosphorus in freshwater environments [3,12,16,35,42,43].

Wang et al. [43], in their studies, found that phosphorus loading to freshwater from Asia accounts for approximately 52.3% of the global total phosphorus (TP), implying that an accurate assessment of the long-term phosphorus budget is required to understand its fluctuations. Although point-source phosphorus discharges into large rivers around the world have decreased as a result of increased wastewater treatment [18,20], eutrophication in enclosed areas has remained a perennial problem despite efforts to implement water quality measures, as nutrient-rich deep waters frequently undermine the effectiveness of these interventions [17]. To address eutrophication and other challenges associated with phosphorus, accurately assessing the concentrations of its different fractions is important for understanding the phosphorus cycling of freshwaters [21,38,44], as most routine water monitoring programs worldwide determine total phosphorus (TP) and dissolved reactive phosphorus (DRP) in water [13,14,33,45]. Knowledge of TP levels, along with different phosphorus speciation, is not only useful for comprehending phosphorus dynamics in river

systems, but it also provides invaluable information for identifying phosphorus sources, assessing anthropogenic pollution [3], and for comprehensive phosphorus management. This is because hydrological seasonal changes can influence these dynamics, leading to variations in dilution, surface runoff, sediment resuspension, and transport of dissolved and particulate phosphorus.

The Yamato River drains a densely populated part of the Osaka metropolitan area. It carries with it large volumes of sewage, overused fertilizers, and industrial wastewater into the Osaka Bay, hence its ranking among the most polluted rivers in Japan [3,46–48], yet the forms and seasonal sources of its particulate phosphorus remain poorly characterized. This study aimed to determine the concentrations of various forms of phosphorus in the Yamato River, assess the short- and long-term variations in the concentrations of different fractions of phosphorus present in water in relation to observed water-level variations, and identify the likely sources of the various fractions of phosphorus in the Yamato River.

2. Materials and Methods

2.1. Study Area

The Yamato River is one of the major rivers that originates from the northern part of Nara Prefecture and flows through the Osaka Plain before discharging into Osaka Bay, located in the southern part of the Osaka megacity. It runs along the boundary between Osaka and Sakai City and spans a length of 68 km [49–51]. The catchment area (Figure 1a) encompasses 1077 km^2 and experiences annual precipitation of approximately 1360 mm, with rainfall particularly pronounced during the East Asian rainy season mainly in June and July [43]. This river basin serves a population of over 2 million people as recorded in 2009 [46]. The catchment area enjoys a Seto Inland Sea climate, characterized by generally warm temperatures throughout the year, with an average temperature of 15 °C [43].

The soil types in the catchment area primarily consist of Regosols and Fluvic Gleysols (32% and 29%, respectively), followed by Cambisols (15%), Acrisols and Luvisols (15%), and Ochric Gleysols at 7%, while Fluvisols, Lithosols, and Andosols each account for less than 1% [43,51]. There are three types of land use within the catchment area, which include agricultural activities (mainly rice paddies), forested areas (bamboo, deciduous, and evergreen forests), and residential areas (constructed land) (Figure 1b). With increased population, the constructed land has also significantly increased from about 16.9% in the 1970s to about 29.3% in the 2010s, while the total built-up land covers up to 34.5%. Conversely, the agricultural land has seen a decline from about 32.4% to about 19.4% in the same period, while rice paddy fields cover up to 15.2% [51,52]. However, the forested land has remained unchanged and accounts for approximately 40% of the catchment area [53]. Five wastewater treatment plants (WWTPs) in the Yamato River catchment area were established between 1970 and 1985, and another one was built in 1963 in the estuary area that processes both municipal and human waste before being released into the Yamato River system [43]. Despite the implementation of numerous protective measures to restore the water environment in the Yamato River by the local agencies, water pollution and eutrophication have remained a persistent challenge [51].

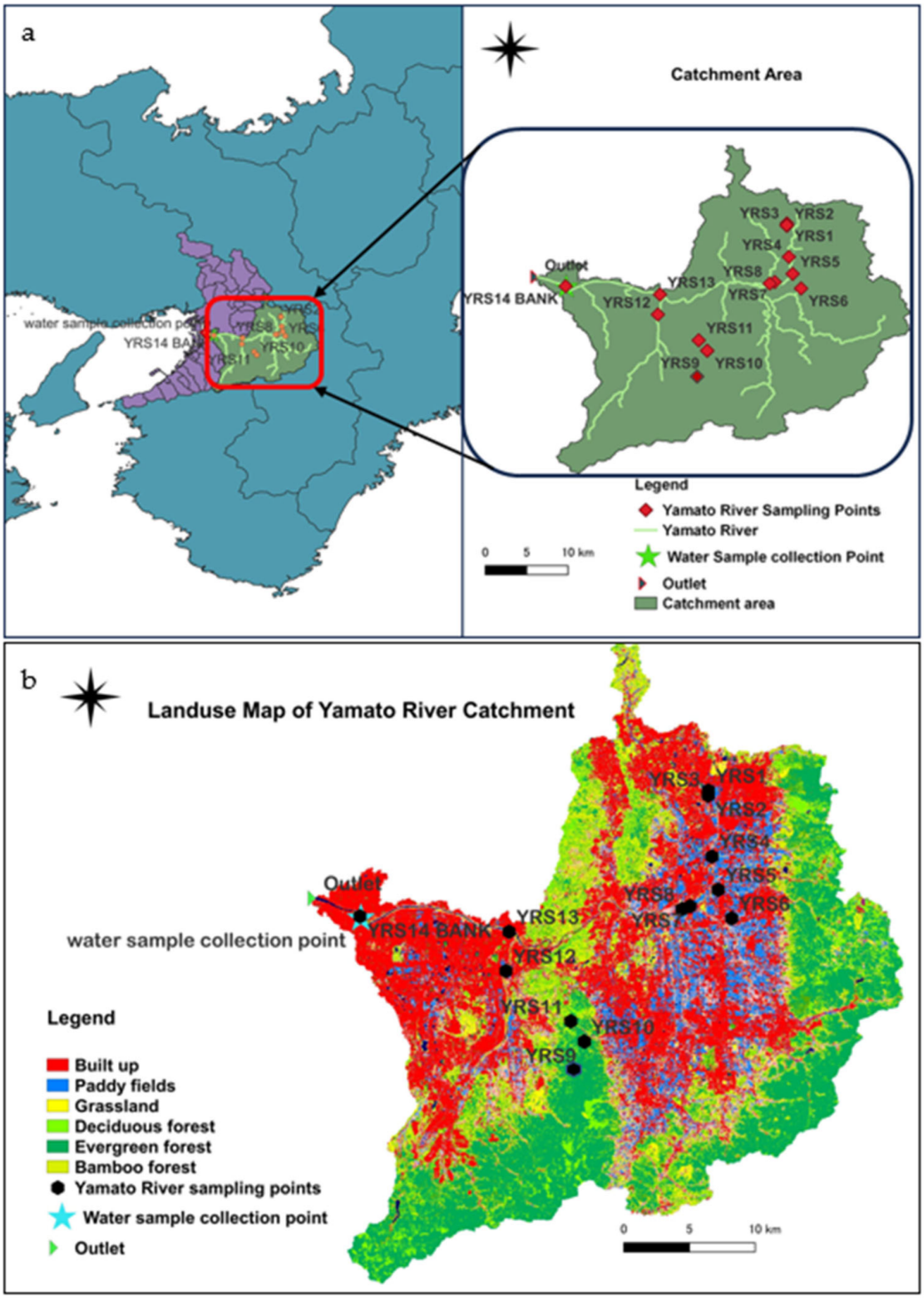


Figure 1. (**a**) Map of the study area showing the locations of different sample collection points and (**b**) land use in the Yamato River catchment area, including the sample collection points.

2.2. Sample Collection

River water samples were collected weekly from the left bank of the Yamato River in the downstream site (latitude 34°35′25.50″ N, longitude 135°29′52.43″ E), located adjacent to the estuary, from October 2022 to December 2024. A total of 119 water samples were collected between 10:00 and 12:00 from approximately 5 cm below the surface of the water on Wednesday of every month, except during extreme rainfall or whenever conditions did not allow. Unfiltered water samples for total phosphorus (TP) analysis were collected directly

into 10 mL plastic vials. Water samples for the different phosphorus fraction analyses were filtered using 0.45 µm membrane filters to capture the particulate phosphorus. The membrane filters and unfiltered water samples were stored in a freezer until analysis.

Additional samples collected for the analysis of various phosphorus fractions included three surface soil samples from paddy fields (YRS 3, 5, and 6), three soil samples from the forested regions (YRS 9, 10, and 11), two riverbank soils from the Yamato Riverbank (YRS 8 and 14) and seven sediment samples from the Yamato River riverbed (YRS 1, 2, 4, 7, 12, 13, and 15). Sample YRS 7 corresponded to a sediment sample obtained from the riverbed adjacent to the sewage treatment facility upstream of the Yamato River (see Figure 1a). In total, there were 16 sampling sites in the study area. The soil samples from the riverbed and riverbank of the Yamato River, as well as from the surrounding forested areas and paddy fields, were collected into clean plastic bags and placed in a cooler box, then maintained under refrigeration until analysis.

2.3. Precipitation and Water Level

The Sakai and Oriono weather stations continuously monitored precipitation levels in the surrounding area while the water levels of the Yamato River were recorded at the gauging station near the Oriono bridge. The collected data were used to evaluate how the water levels changed with varying amounts of precipitation. However, the water-flow (discharge) data for the Yamato River were unavailable as the Oriono station did not possess the dataset for the study period. Figure 2 illustrates these variations in water level in relation to varying precipitation.

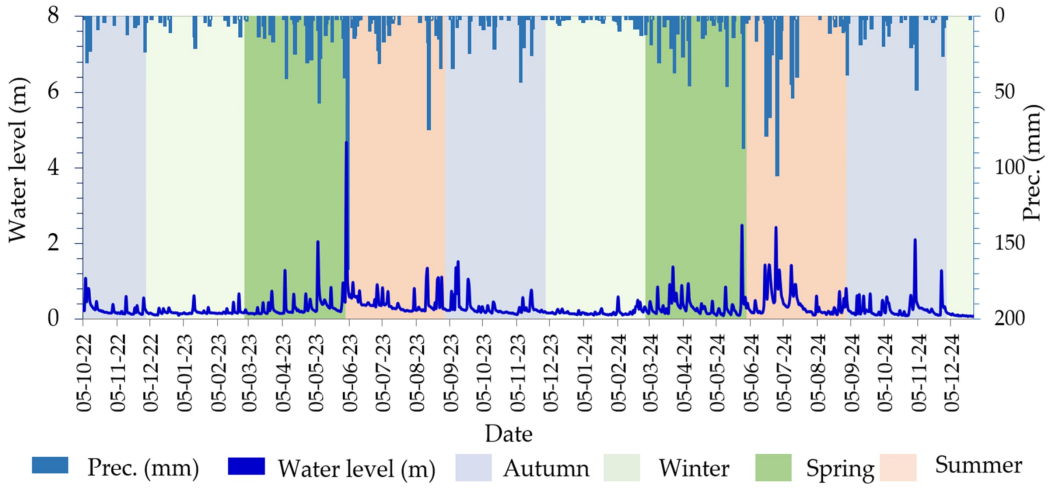


Figure 2. Daily precipitation and water level of the Yamato River throughout the study period, covering all seasons (Source: Sakai and Oriono weather stations).

2.4. Sample Analysis

Unfiltered water samples were analyzed for total phosphorus (TP) using the peroxydisulfate digestion method at 120 °C for 30 min in an autoclave [9,33,54]. Total particulate phosphorus (TPP) was assessed using the high-temperature hydrochloric acid method [7,55], which involved extracting the samples with 1 M HCl and shaking for 24 h after adding a few drops of 1 M $Mg(NO_3)_2$, followed by combustion at 550 °C in a muffle furnace. Particulate inorganic phosphorus (PIP) was quantified using the hydrochloric acid extraction method, where 1 M HCl was added to the samples and shaken for 24 h. After extraction, the phosphorus concentration was determined using the molybdenum blue (colorimetric) method [6,9,56,57]. Particulate organic phosphorus (POP) was calculated as

the difference between TPP and PIP (POP = TPP–PIP) [9], while total dissolved phosphorus (TDP) was derived from the difference between TP and TPP (TDP = TP −-TPP) [9,54,58].

The sequential extraction method (SEDEX) (Table 1) which isolates several phosphorus species in sediments according to their reactivity and solubility was used to analyze various fractions of particulate phosphorus, including exchangeable phosphorus (Ex-P) or loosely bound phosphorus that is easily released into the water column, iron-bound phosphorus (Fe-P) produced by co-precipitation with Fe/Mn oxides and hydroxides, calcium-bound phosphorus (Ca-P) including $CaCO_3$ from biological apatite such as teeth and sea shells, detrital phosphorus (Det-P) an inert coarse-grained fraction derived from erosion of rocks, and organic-bound phosphorus (Org-P) that is biologically formed from sinking organic matter [3,59,60]. Subsequently, the phosphorus content from each extraction step was quantified using the molybdenum blue method at a wavelength of 880 nm with a UV-Vis spectrophotometer [3,60].

Table 1. Shows the summary of the SEDEX method used for the extraction of different phosphorus fractions from the dried sediment and soil samples [3,44,59].

SEDEX Extraction Method		
Extractant	**Conditions**	**Expected P fraction**
1 M $MgCl_2$	2 h	Exchangeable or Soluble P (loosely sorbed) (Usable by algae or phytoplankton
BD solution	4 h	Iron-bound P (Fe-P) Released from anoxic or hypoxic environment
1 M NaCl rinse	2 h	
1 M NAcetate Buffer	6 h	Authigenic Apatite or Ca-P (Transformed from Fe-P and Org-P in early diagenetic reactions)
1 M $MgCl_2$ rinse	2 h	
1 M HCl	16 h	Detrital Apatite Inert coarse-grained fraction from erosion of rocks
HCl (after combustion at 550 °C)	16 h	Refractory Organic P Biologically formed and also sensitive to physicochemical changes

Following the procedure in Table 1 above as outlined by Gamage et al., Rahman et al., and Zhang et al. [3,44,59], filter papers containing the dried sediment samples (separate filters from TPP and PIP) were transferred into 50 mL centrifuge tubes. For the extraction of the Ex-P, a 1 M $MgCl_2$ solution was added to each sample and shaken for 2 h. The extraction of Fe-P was conducted using a BD (bicarbonate dithione) solution, shaking for 4 h, followed by a rinse with 1 M NaCl and an additional 2 h of shaking. The combined supernatants were collected in clean plastic bottles. For Ca-P extraction, a 1 M NaAc buffer (1 M CH_3COONa + conc. Acetic acid) was utilized, with shaking for 6 h, followed by a 1 M $MgCl_2$ rinse for 2 h. Subsequently, a 1 M HCl solution was added to the sediment sample, followed by 16 h of shaking to extract Det-P. Finally, the sediment samples were transferred into clean crucibles and combusted for 16 h at 550 °C in a furnace (Yamato FO300 muffle furnace from Yamato Scientific Co., Ltd., Tokyo, Japan). The samples were then transferred into centrifuge tubes, and 1 M HCl was added to extract refractory Org-P. At the end of the extraction steps, the samples were centrifuged and then filtered through 0.2 µm glass filters before analysis. To determine the phosphorus concentration in all the fractions, the molybdate blue method and a UV/Vis spectrophotometer (Thermo Scientific MULTISKAN GO, from Thermo Fischer Scientific, Waltham, MA, USA) at a wavelength of 880 nm were employed.

For the additional samples, which include surface soil samples from paddy fields and forested regions, as well as riverbank and riverbed sediment samples, a total of 0.4 g of dried homogenized sample was weighed and placed into a 50 mL centrifuge tube. The procedure outlined in Table 1 was then followed.

3. Results and Discussion

3.1. Seasonal Variation in the River Water Level with Precipitation

There was a notable fluctuation in precipitation levels observed throughout the study period. Precipitation was relatively low during the autumn-to-winter period from October 2023 to February 2024, followed by a gradual increase in the spring-to-summer period between June and August 2024. A similar pattern was observed in water levels, with significant changes corresponding to variations in precipitation. The maximum water levels coincided with the highest daily precipitation of the study period (167 mm on 2 June 2023), whereas the minimum occurred during the rain-free period (Table 1). These data indicate that precipitation directly impacts water levels in the Yamato River, as increases in precipitation resulted in corresponding increases in water levels and vice versa.

3.2. Seasonal P Fractions Concentrations and Their Variations with Water Level

Table 2 presents the average concentrations of TP, TDP, TPP, PIP, and POP, including their minimum and maximum values for the individual years 2022, 2023, and 2024, as well as for the combined study period from 2022 to 2024.

Table 2. Maximum, minimum, and average values for daily precipitation, water level, and different phosphorus species in Yamato River water.

Year		Parameter						
		Prec. (mm)	**WL (m)**	**TP (mg/L)**	**TDP (mg/L)**	**TPP (mg/L)**	**PIP (mg/L)**	**POP (mg/L)**
2022	Max:	47.00	2.14	0.34	0.25	0.28	0.08	0.24
	Min:	0.00	0.10	0.21	0.00	0.08	0.04	0.03
	Ave:	3.02	0.34	0.29	0.17	0.12	0.06	0.06
2023	Max:	167.00	4.67	0.64	0.58	0.44	0.30	0.14
	Min:	0.00	0.12	0.14	0.00	0.03	0.03	0.00
	Ave:	3.48	0.32	0.30	0.19	0.10	0.08	0.02
2024	Max:	105.50	2.48	0.77	0.71	0.47	0.46	0.15
	Min:	0.00	0.06	0.17	0.00	0.03	0.02	0.00
	Ave:	3.92	0.30	0.34	0.25	0.08	0.07	0.02
2022–2024	Max:	167.00	4.67	0.77	0.71	0.47	0.46	0.24
	Min:	0.00	0.06	0.14	0.00	0.03	0.02	0.00
	Ave:	3.61	0.31	0.31	0.22	0.10	0.07	0.02

Pearson correlation between water level and the P fractions is shown in Figure 3. TP demonstrated a weak negative correlation with water level. TPP displayed a moderate positive correlation with water level and a strong correlation with both PIP and POP. Conversely, TDP showed a moderate negative correlation with water level.

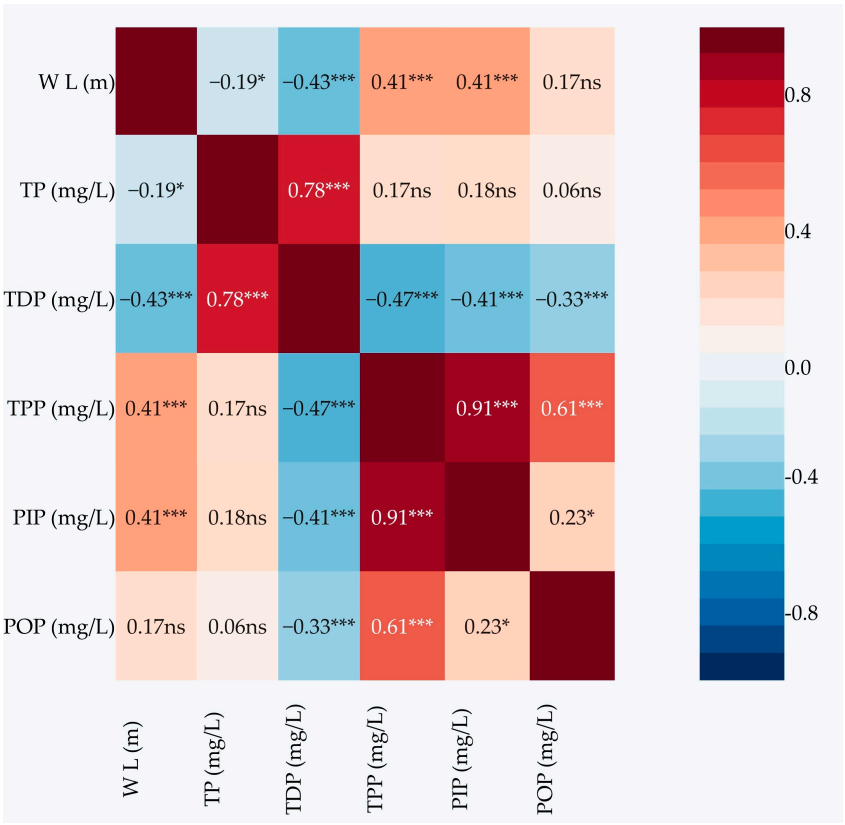


Figure 3. Pearson correlation of the various P fractions with water levels and other fractions. Pearson r with significance (*** $p < 0.001$, * $p < 0.05$ and ns = not significant).

The variations in the various P fractions with water level throughout the study period are illustrated in Figure 4.

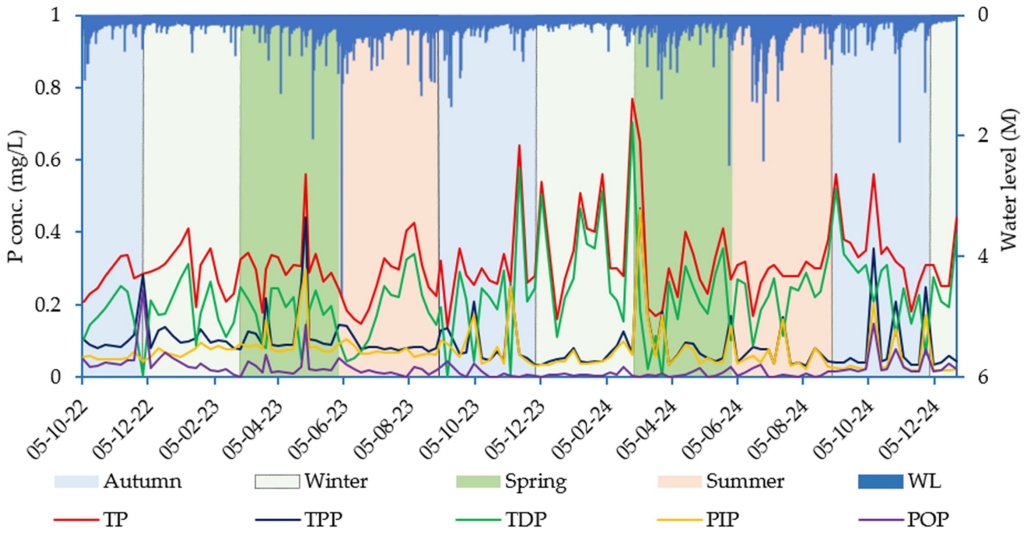


Figure 4. Variations in TP, TPP, TDP, PIP, and POP with water level. The different color bands represent the different seasons throughout the study period.

The relatively low TP concentrations observed during high-water levels may partly reflect a dilution effect associated with increased water levels [9,18,34,58]. This is consistent with the weak Pearson correlation of −0.19 ($p < 0.05$) between the measured TP concentrations and the river water levels, although the weak correlation suggests the presence of

other factors influencing the TP concentrations. In contrast, the increase in TP observed under lower water-level conditions may be linked to the degradation of organic matter, weathering of phosphorus materials, internal loading, and agricultural emissions that may have found their way into the river system [19,20,32,61].

Savenko S. et al. and Ellison M. et al. [25,62] in their studies suggest that the growing population around the catchment area could contribute to increased total phosphorus (TP) concentrations over an extended period. High population density often leads to an increase in pet waste and the discharge of substantial volumes of sewage and other household waste, including detergents, into the river. Furthermore, increased fertilizer use may also significantly contribute to rising TP concentrations due to phosphorus discharge into the waterway [63].

The presence of water containment structures for irrigation in the upstream paddy fields may have influenced the TP levels observed in the study. Friedl, G. and Wüest, A. Hillbricht-Ilkowska, A. and Shimizu, Y. et al. [64–66] noted in their studies that these reservoirs create stagnant sections, which in turn affect the residence time, a factor that controls processes such as nutrient assimilation by phytoplankton.

Weirs tend to reduce the water flow velocity and increase water residence time, which influences the phosphorus dynamics. The reduced water flow and increased residence time favor retention and accumulation of phosphorus within the water system [41]. Under low-flow conditions or relatively stagnant conditions, phosphorus may be retained through sedimentation and adsorption to suspended particles and sediments [19]. As a result, TP concentrations in the downstream samples may be affected by the presence and operations of these structures, as well as by the discharge at the time of sampling [18].

TDP was calculated as the difference between the measured TP and TPP concentrations (TDP = TP − TPP) [9,58]. Figure 4 shows the seasonal variation in TDP and TPP in relation to varying water levels. Throughout the study period, TDP consistently dominated over the TPP, with a slightly more pronounced increase during low-flow conditions compared to high-flow conditions. During the low-flow period, phosphorus that had accumulated in the deposited sediments may have been released into the overlying water as dissolved phosphorus, following a reduction in soluble phosphorus levels [67].

During high-flow conditions, abiotic interactions may have facilitated the adsorption of soluble phosphorus onto suspended sediments, leading to a transition from the dissolved to the particulate phase. This transition, alongside dilution effects, resulted in decreased TDP concentrations [9,68,69]. Additionally, Ellison and Brett, Jordan, T. et al. and Luo, P. et al. [62,70,71] observed that precipitation significantly increased the load and concentration of suspended sediments while influencing riverbank soil inflow, overland runoff flow from adjacent agricultural land, and the urban canals [42,58]. Resuspension of phosphorus contained in riverbed sediments also occurred due to rising water levels during high flow [9,14,61], causing an increase in TPP. In contrast, the settling of particulate matter to the riverbed sediments during low-flow conditions resulted in lower TPP concentrations [25].

Containment structures (weirs) located upstream may also influence TPP and TDP levels. These structures reduce the river flow, leading to increased water depth as water accumulates. The reduced water flow may then promote sedimentation and trapping of particulate phosphorus from tributaries and other sources [72]. During high-flow conditions, rising water levels can cause the resuspension of trapped phosphorus, which is then released along with accumulated sediments, resulting in increased TPP downstream [41].

The POP concentrations were calculated as the difference between the determined TPP and PIP concentrations (POP = TPP – PIP) [9]. Figure 4 illustrates the variation in PIP and POP across different water levels. The results indicated that PIP exceeded POP on most sampling dates (mean 0.07 vs. 0.02 mg/L; Table 1). In relation to water levels, both PIP and

POP concentrations showed an increase (Pearson correlation of 0.43 and 0.41, respectively) (Figure 3).

Elevated PIP values observed under high flow conditions may be linked to urban surface runoff resulting from heavy precipitation, wastewater discharge, and the leaching of inorganic fertilizers from nearby paddy fields. According to Owens, P. et al. and Gebus-Czupyt and Wach [1,61], a significant amount of phosphorus in river systems is bound to inorganic material particles, with PIP being more prevalent in rivers receiving industrial effluents. Regelink, I. et al. [29] in their study also noted that waters in catchment areas adjacent to agricultural setups are primarily affected by inorganic phosphorus fertilizers, as well as the timing of their applications in nearby farmlands.

POP accounted for a lower fraction of TPP compared to PIP. This finding suggests that the variations in TPP relative to water levels are mainly due to changes in PIP, consistent with the study conducted by Nguyen, T. et al. [9]. Potential sources of POP in river water during high precipitation include urban stormwater, sewage discharge, and runoff of decayed organic matter and animal manure from agricultural lands [7,73]. The concentrations of POP during the low precipitation period could be attributed to the deposition of naturally decaying aquatic plants [74].

3.3. Comparison of the Phosphorus Speciation with Other Studies

The comparison of the various phosphorus in the water samples from other studies with our study is presented in Table 3.

Table 3. The comparison of the TP, TDP, TPP, PIP and POP concentrations (mg/L) in river water from different Rivers with the Yamato River.

Study Area	TDP (mg/L)	TPP (mg/L)	PIP (mg/L)	POP (mg/L)	TP (mg/L)	References
Fox River	0.115–0.209	0.096–0.117	0.046–0.055	0.050–0.062	0.210–0.326	[6]
Changjiang River	0.012–0.046	0.019–0.201	0.019–0.201	-	0.031–0.246	[75]
Milwaukee River	0.025–0.123	0.023–0.054	0.012–0.022	0.011–0.032	0.048–0.177	[76]
Dafengjiang River	0.010–0.077	0.003–0.001	0.003–0.010	-	0.013–0.087	[77]
Yamato River	0.00–0.71	0.03–0.47	0.02–0.4	0.00–0.2	0.14–0.77	This study

The TP concentrations from our study had the highest maximum value (0.77 mg/L), although its minimum value (0.14 mg/L) was lower than that of the Fox River. TDP from our study had a substantially higher maximum (0.71 mg/L) and also exhibited the widest range (0.00–0.71 mg/L) compared to the other Rivers, indicating pronounced variability. The maximum TPP concentrations in the Yamato River (0.47 mg/L) also exceeded the upper limits of all the comparison sites. Similarly, the maximum PIP value from the Yamato River (0.4 mg/L) exceeded those of the other Rivers, although the minimum value (0.02 mg/L) was comparable to the Chanjiang River (0.019 mg/L) and significantly higher than the Dafengjiang River (0.003 mg/L). The Yamato River recorded the lowest POP concentration (0.00 mg/L), although its maximum value (0.2 mg/L) exceeded the values reported for the Milwaukee River (0.032 mg/L) and the Fox River (0.062 mg/L). In comparison to the other study sites, the Yamato River was characterized by higher peak concentrations as well as wider variability in the phosphorus fractions. These results suggest a stronger phosphorus enrichment in the Yamato River compared to the other study sites. This may be due to differences in catchment activities and population, as well as hydrological differences.

3.4. Sequential Extraction of P Species and Their Variation with Water Level

Sequential extraction of particulate phosphorus in the water samples yielded the following phosphorus (P) species: Ex-P, Fe-P, Ca-P, det-P, and org-P. The minimum, maximum, and average values for these species are presented in Table 4. The average TPP concentration was 0.10 mg/L, with a minimum value of 0.03 mg/L and a maximum value of 0.47 mg/L (Table 2).

Table 4. Maximum, minimum, and average values for the different phosphorus species in Yamato River water from the SEDEX extraction.

Year		Phosphorus Fraction				
		Ex-P (mg/L)	**Fe-P (mg/L)**	**Ca-P (mg/L)**	**Det-P (mg/L)**	**Org-P (mg/L)**
2022	Max:	0.04	0.05	0.07	0.05	0.18
	Min:	0.01	0.01	0.01	0.02	0.00
	Ave:	0.02	0.03	0.03	0.04	0.03
2023	Max:	0.05	0.11	0.06	0.03	0.08
	Min:	0.00	0.01	0.00	0.00	0.00
	Ave:	0.02	0.05	0.02	0.01	0.02
2024	Max:	0.12	0.23	0.04	0.02	0.04
	Min:	0.01	0.02	0.01	0.00	0.01
	Ave:	0.04	0.09	0.02	0.01	0.02
2022–2024	Max:	0.12	0.23	0.07	0.05	0.18
	Min:	0.00	0.01	0.00	0.00	0.00
	Ave:	0.02	0.05	0.02	0.01	0.02

During autumn and winter (Figure 5), Org-P species were elevated on several dates. The Ca-P and Fe-P species were also detected at the onset of autumn, although their concentrations decreased throughout winter before increasing towards the end of the season, coinciding with rising water levels. The observed Org-P concentrations during this period may have resulted from contributions from domestic waste, septic tank discharges, and sewage treatment works [34,78] that entered the river.

Notable amounts of Det-P species were recorded multiple times during this period, potentially from eroded soils from the nearby paddy fields as well as sediment resuspension and decayed organic matter from the forested areas [60,79,80]. The Ex-P species was relatively low compared to the other species, exhibiting fluctuations with changes in water level.

In the spring-to-summer season, which is characterized by elevated water levels, Fe-P emerged as the dominant component of TPP. This may have been as a result of erosion of Fe-oxide-rich soil, resuspension of bed sediment, and flooded paddy soils that tend to release Fe-bound P under reducing conditions. The next most abundant species during this period was Ca-P. This pattern coincided with the local rice planting season, suggesting that fertilizers applied in paddy fields may have leached or washed into the Yamato River.

The observed levels of Ex-P, Org-P, and Det-P during this season may have resulted from the remobilization of riverbed sediments and riverbank erosion [61], as well as urban storm runoffs, runoff from forested and agricultural areas, sewage treatment works, and domestic septic discharges [34] during this time.

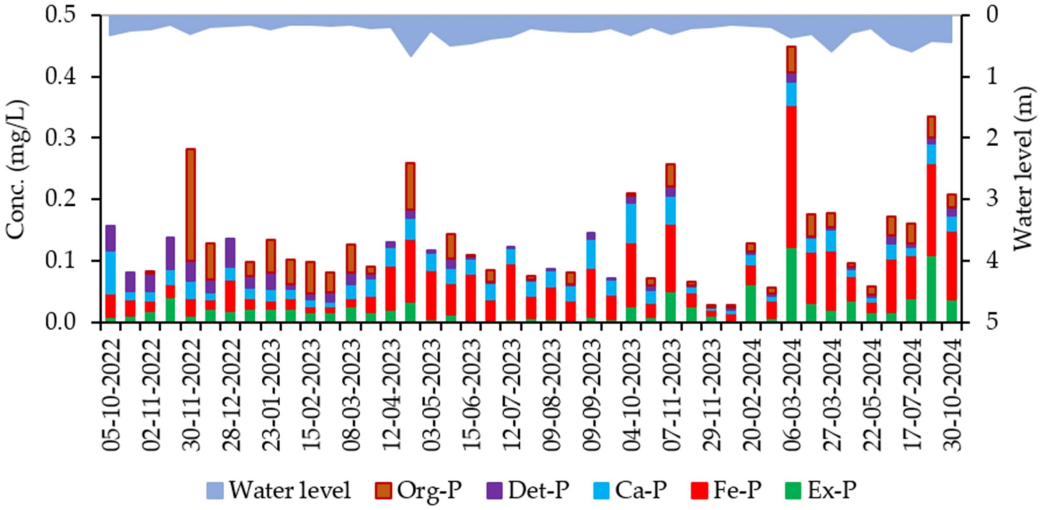


Figure 5. Concentrations (mg/L) of the particulate phosphorus species in Yamato River water determined by SEDEX.

The results of the analysis of the additional samples for various phosphorus species are presented in Table 5. Fe-P was the most dominant fraction in all the soils collected from all the sampling locations, although it was highest in the paddy field samples alongside Ca-P. One paddy field sample (YRS 5) and a forested area sample (YRS 10) had Det-*p* values of over 200 mg/kg, while the rest of the samples recorded values of 150 mg/kg and below. The Ex-P fraction recorded the highest values in the paddy field samples YRS 3, followed by YRS 5. Unfortunately, we were unable to analyze our additional samples for Org-P as our samples were destroyed during the combustion process.

Table 5. Maximum, minimum, and average values of the different phosphorus species for the additional samples collected from riverbed, paddy fields, riverbank, forested area and near the wastewater treatment plant.

		Phosphorus Fraction			
Sample Area		**Ex-P (mg/kg)**	**Fe-P (mg/kg)**	**Ca-P (mg/kg)**	**Det-P (mg/kg)**
Riverbed	Max:	6.04	310.94	82.45	132.30
	Min:	3.10	132.41	17.54	23.20
	Ave:	4.98	251.39	38.23	65.44
Paddy field	Max:	141.10	725.29	273.55	249.74
	Min:	8.82	650.60	160.23	39.89
	Ave:	64.45	685.47	232.34	144.28
Wastewater treatment plant (WWTP)	Conc:	4.34	483.20	30.94	65.23
Riverbank	Max:	12.06	704.69	106.66	145.28
	Min:	7.58	433.75	53.60	72.03
	Ave:	9.82	569.22	80.13	108.65
Forested area	Max:	6.35	364.72	47.94	275.08
	Min:	3.1	130.87	11.36	14.24
	Ave:	4.85	216.55	28.88	102.32

Finally, this study mainly focused on measuring and analyzing various forms of phosphorus concentrations (mg/L) and the identification of their sources. The phosphorus loads (mass per unit time) were not calculated; therefore, our results describe changes in the phosphorus concentration and speciation but do not quantify the total amount of phosphorus from the catchment. Therefore, for comprehensive nutrient management, we recommend future studies to integrate both concentration and load management to assess total phosphorus export from the catchment.

4. Conclusions

The TP concentrations were generally higher during autumn and winter, characterized by lower water levels compared to spring and summer, when water levels were higher. TDP consistently dominated over TPP but was more pronounced in winter compared to summer. This observation suggests that the dissolved and particulate forms of phosphorus respond differently to seasonal hydrological changes. PIP exceeded POP on most sampling dates, although both increased with rising water levels and decreased with declining water levels in summer and winter, respectively. Fe-P was the dominant P species in the river water, which increased alongside Ca-P in the summer, coinciding with the irrigation of paddy fields. Fe-P was highest in the surface soils from the paddy fields, although it also dominated other species in the soil and sediment samples. The results suggest that the phosphorus pollutants in the river water were mostly inorganic in nature, perhaps due to the seepage of chemical fertilizers from nearby paddy fields, wastewater discharge, street dust, and construction debris carried as erosion products.

The seasonal interpretation in this study is based on the weekly sampling and the locations used. Therefore, storm events, extreme rainfall times, and short-term phosphorus changes may not have been fully factored in. We suggest the inclusion of additional upstream and downstream sampling locations, event-based and daily sampling, as well as continuous discharge monitoring to calculate the loads for the phosphorus fractions and the incorporation of physicochemical, isotope or tracer analysis and hydrological modeling to test the relative contributions of the different phosphorus sources.

Author Contributions: Conceptualization, S.-i.O., M.S. and T.I.; methodology, T.I. and M.S.R.; formal analysis, W.Z., D.N.M. and S.B.K.; formal analysis, W.Z. and D.N.M.; resources, S.-i.O., M.S. and T.I.; writing—original draft preparation, D.N.M.; writing—review and editing, D.N.M.; supervision, S.-i.O., M.S. and T.I.; project administration, S.-i.O., M.S. and T.I.; funding acquisition, S.-i.O., M.S. and T.I. All authors have read and agreed to the published version of the manuscript.

Funding: This work was funded by the Asia-Pacific Network for Global Change Research (No. CRRP2019-09MYOnodera) and the Research Grant of Preservation, Restoration and Creation in a Coastal Environment in Osaka Bay area by Osaka Bay Regional Offshore Environmental Improvement Center (PI: Mitsuyo Saito, No. 040006, 050005, 060003).

Data Availability Statement: The data presented in this study are available upon request from the corresponding author.

Acknowledgments: We sincerely appreciate Tetsuo Kamei and his colleagues for their assistance with the weekly river water sampling for this study and the JICA-AGRINET for funding David MOENGA's PhD study.

Conflicts of Interest: The authors declare no conflicts of interest.

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