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Research Article

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ASSESSMENT OF ORGANIC PHOSPHORUS DISTRIBUTION IN THE SEDIMENT AND WATERS OF LANGAT RIVER, MALAYSIA

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ARTICLE HIGLIGHTS

- Langat River water and sediment quality deteriorate toward downstream.
- Dissolved organic phosphorus increases near agricultural areas.
- Sediment phosphate retention dominated by non-labile organic phosphorus.
- Downstream pollution linked to sediment sorption and nearby agriculture.

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ABSTRACT

The Langat River traverses rapidly developing urban areas in Malaysia and is significantly affected by anthropogenic activities. The introduction of excessive phosphorus into rivers poses a significant ecological issue. Water and sediments were sampled from nine stations at Langat River to evaluate the current and potential impacts of organic phosphorus. The water quality parameters indicated a progressive decline downstream, attributed to allochthonous sources from tributaries and land use practices. Inorganic substances were the principal cause of pollution in the river while degradation of organic pollution biologically is reduced. Dissolved organic phosphorus (DOP) played a significant role at stations that are either relatively unpolluted or in areas with high population density and dominated by agricultural lands, serving as a potential source of bioavailable phosphorus. The total organic phosphorus in the sediment increased downstream, predominantly comprising non-labile fractions (67 – 78%). The labile fractions exhibited strong correlations with dissolved oxygen (DO) (r = -0.797), dissolved organic phosphorus (DOP) (r = 0.931), and conductivity (r = 0.837), suggesting internal loading to the water column. Increased non-labile fractions indicated the sediment's capacity to retain organic phosphorus. The downstream stations exhibited elevated risk owing to high sorption capacity, sediment deposition rate and land use type in the basin.

Keywords: dissolved organic phosphorus, ecological subsidy, organic pollution, sediment phosphate retention, tropical river, water quality

INTRODUCTION

Rivers are a valuable resource for humans providing water, food and transportation (Chan 2012) and this includes the Langat River, Malaysia. In accordance with Ahmed *et al.* (2023), approximately 33% of the population depends on the river for potable water. Ten freshwater treatment plants located in the vicinity of the Langat River Basin supply residential, industrial, and agricultural areas (Abidin *et al.* 2018) but they frequently have to shut down due to contamination (Farid *et al.* 2016).

Pollution that plagues freshwater bodies can be traced to a point-source or to several possible non-point-sources. Point sources, such as water treatment facilities, livestock farms, and industry can be traced back to an origin point unlike non-point sources (Ferrier *et al.* 2001; Mohd *et al.* 2012). Since non-point sources are harder to identify, they are also known as diffuse sources and cause significant pollution in freshwater bodies (Duncan *et al.* 2012). Both type of sources that stem from human activities affect the pollution level in Langat River which includes aquaculture, sewage treatment plant outflow, illicit dumping, irrigation runoff, and river sand mining (Lim *et al.* 2023), with non-point sources accounting for most pollution (Juahir *et al.* 2011).

Suki *et al.* (1988) reported that the Langat River's water quality was classified as moderately contaminated, as confirmed by Hashim *et al.* (2018) thirty years later. Although this suggests that the water quality is stable, it also implies that there have been no improvements in the water quality. Indeed, a review of the index used to grade river water quality in Malaysia is required (Juahir *et al.* 2011). The Langat River Basin is currently Malaysia's most urbanized river due to rising development over time (Selamat *et al.* 2023). In 2013, mangroves and peat swamps decreased significantly from 25.7% to 9.4% of the basin area, while the amount of land used for development increased from 2.4% to 23.5% compared to 1974 (Elfithri 2018). From 2000 to 2013, population growth grew concurrently, rising from 1,184,917 to 4,266,011 (Ahmed *et al.* 2023), thus, the Langat River's ecosystem and water quality have been put under stress due to the growing population and increasing land usage for development and non-point source pollution.

Phosphorus (P) plays a key role in natural environments (Maitra *et al.* 2015) but excessive phosphorus due to anthropogenic activity can lead to eutrophication which in turn can lead to large-scale deaths of aquatic organisms (Zhu *et al.* 2015). Therefore, monitoring phosphorus levels in aquatic environments like rivers and lakes is necessary to maintain a balanced ecosystem. In aquatic environments, the particulate and dissolved phosphorus forms exist in equilibrium with the major portion found in the solid phase. Under certain conditions such as an increase in pH, the absence of oxygen, microbial activity, and changes in salinity, the reduction of external phosphorus inputs can cause sediment-bound phosphorus to be released into the water to maintain the equilibrium (Spivakov *et al.* 1999; Gardolinski *et al.* 2004). However, the sediment sorption capacity is also important in the maintenance of the equilibrium in aquatic environments. Sediments can accumulate up to 1,000 times the concentration found in the waters (Pardo *et al.* 1998) with pore water found in sediments containing 103 times more phosphorus than the water overlying the sediment (Li *et al.* 2013). Its ability to release sorbed phosphorus can ensure that phosphorus levels remain high despite efforts to reduce the introduced phosphorus pollution load (Søndergaard *et al.* 1999; Yu *et al.* 2017).

No studies have evaluated the phosphorus status in the waters and sediments of Langat River to date and generally, the river exceeds the permissible phosphorus levels (Al-Badaii *et al.* 2013). In aquatic environments, phosphorus compounds are generally divided into organic and inorganic phosphorus. The former can be found relatively abundant and is often classified as biologically unavailable unlike orthophosphate (McKelvie 2005). Organic phosphates are usually found in the form of nucleic acids, phospholipids, phosphonates and inositol phosphates which are important in the building and maintenance of living organisms (Baldwin 2013). However, there is insufficient knowledge on the relative contribution of organic phosphorus and the influence that the sediments have in organic phosphorus cycling. Therefore, this study determined the extent of organic phosphorus pollution in one of the more important rivers of Selangor, Langat River by characterizing the water physicochemical parameters, as well as the association between the organic phosphorus fractions and sediment sorption capacity to the dissolved organic phosphorus (DOP) in the water.

MATERIALS AND METHODS

Sampling Stations and Process

The Langat River Basin catchment area covers 1,815 km2 most of which is located in the state of Selangor and the main course of the river is 141 km long. (Ahmed & Mokhtar 2020). The river originates from the Titiwangsa Range in the District of Hulu Langat and drains at the Straits of Malacca (Lim *et al.* 2023). According to Abidin *et al.* (2018), the major land use in the radius of 1 km from Langat River is commercial plantation which includes palm oil and rubber estates. In addition, Langat River is supplied by at least three main tributaries including Lui, Semenyih and Beranang Rivers (Juahir *et al.* 2011). Other anthropogenic activities such as sand dredging also occur especially at the tributaries like Semenyih River which is also classified as moderately polluted (Kasmuri *et al.* 2021).

The nine sampling sites were selected based on their positions in order to cover as much of the length of the river as possible as accessibility can be a limiting factor (Abd Rahman 2020; Table 1; Fig. 1).

The physicochemical parameters, pH, DO, and electric conductivity were measured using a Hach portable multi-meter (Model HQ 2200). Water samples for the biological oxygen demand (BOD) and chemical oxygen demand (COD) were collected in clean Schott bottles and stored on ice before laboratory analyses. The water samples were collected just below the water surface and closer to the riverbanks whenever possible using a horizontal water sampler. Sediment samples were collected between 5 to 10 cm deep using an Ekman Grab dropped from an overpass or bridge where the river is deep or inaccessible from the bank, but a bucket was used for collection in locations where the water level was less than one meter and when the sediment consisted of mostly sand. Collection was performed from November 2022 to June 2023 which fall within the Northeast monsoon (November to March) and part of the Southwest monsoon (May to September). However, the distinctive monsoons and dry seasons are not readily apparent in the areas of Langat River Basin that are closer to the coast due to unpredictable and increased rainfall (Ahmed 2016). Therefore, no seasonal effects were scrutinized in this study.

Table 1 Sampling stations and notes of the land use type in its vicinity

Site	Coordinates	Notes
PRA	3°12'35" N 101°52'24" E	Close to Pangsun Recreational Area, villages
UP4	3°09'17" N 101°50'36" E	Small town center, close to villages
M1	3°07'36" N 101°49'34" E	Dusun Tua area, close to villages
M2	3°05'40" N 101°47'56" E	Sg. Long Quarry Road, close to business centers and stone quarry.
M3	3°00'51" N 101°46'12" E	Sg. Balak, close to residential areas, water treatment plant and cement factory.
M4	2°55'06" N 101°45'36" E	Bangi, close to village
M5	2°51'15" N 101°40'52" E	Dengkil, close to residential areas, plantations and market
D1	2°48'46" N 101°38'33" E	Bukit Changgang, close to oil palm plantations
D2	2°48'16" N 101°37'21" E	Labohan Dagang, close to villages and oil palm plantations

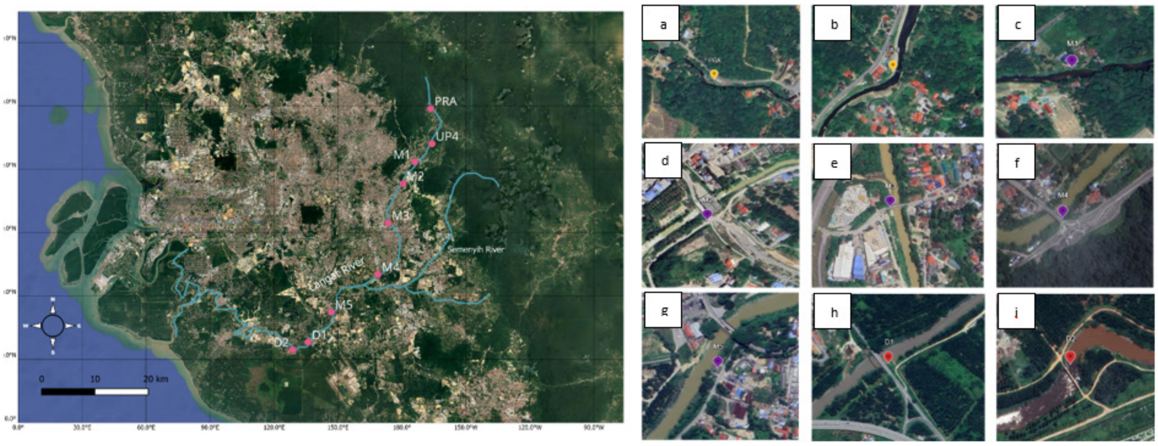

Figure 1 Map of Langat River in Peninsular Malaysia (left). The nine sampling sites and its land use types surrounding the river are depicted on the right

Notes: a = PRA; b = UP4; c = M1; d = M2; e = M3; f = M4; g = M5; h = D1; i = D2.

Oxygen Demand Determination

Biological oxygen demand (BOD) was determined using the LDO 101 probe (detection range was from 0.05 mg/L to 20.0 mg/L) connected to Hach HQ 2200 portable multi-meter unit where the dissolved oxygen was read on the day of collection prior to incubation and on the fifth day after incubation in the dark at 20 ± 5 °C. The difference between readings was taken as the BOD (Rubel *et al.* 2019). The LDO 101 probe was calibrated in 100% water-saturated air before each use according to the manufacturer's protocol. The chemical oxygen demand (COD) was assessed using Hach COD Digestion Vials (low range, 3 – 150 mg/L) following the Hach Protocol Method 8000 on a Hach DR900 colorimeter.

Dissolved Organic Phosphorus Determination

The dissolved organic phosphorus was determined based on methods by Murphy and Riley (1962) and Rowland and Haygarth (1997). Since dissolved organic phosphorus here is defined as soluble unreactive phosphorus, the orthophosphate was first determined from undigested water samples and then the water samples were digested using potassium peroxodisulphate and sulphuric acid to quantify the total dissolved phosphorus. The orthophosphate (soluble reactive phosphorus) concentration was considered part of the inorganic pool. The dissolved organic phosphorus is obtained through subtraction of the orthophosphate concentration from the total dissolved phosphorus concentration. Concentration of both total dissolved and dissolved inorganic phosphorus were determined through colorimetry using Hach DR900 colorimeter. The profile used for the determination of orthophosphate and total dissolved phosphate was Hach Method 8048 and the detection limit is 0.02 mg/L phosphate ion (PO_4^(3-)). The values obtained here were then converted to represent phosphorus, P instead of phosphate ion.

Organic Phosphate Fractionation

The sediment fractionation scheme was conducted according to Wan *et al.* (2020). The collected sediments were first freeze-dried and then crushed using a pestle and mortar in preparation for fractionation. Fractionation was done in triplicate for each of the sites selected. One gram of finely crushed sediment was dissolved in 50 mL of extraction solutions and shaken at 200 rpm sequentially to extract phosphate from the sediments as follows: 0.5 M NaHCO3 (pH = 8.5, 16 hours), 1.0 M HCL (3 hours), 0.5 M NaOH (16 hours) and 1.0 M H2SO4 (24 hours). At every extracting stage, samples were centrifuged at 4,000 rpm for 20 minutes and the supernatant was filtered through Whatman 41 filter paper before phosphorus concentration determination. Inorganic phosphorus was determined using the molybdenum blue assay (Murphy & Riley 1962).

The total digestion method (Rowland & Haygarth 1997) was performed with a modification to the digestion time, three hours of digestion as opposed to one-hour digestion since the extracts may contain different types of organic phosphorus that require longer digestion time for complete oxidation (Ma *et al.* 2017). The available organic phosphate was categorized into labile, moderately labile, and non-labile fractions according to Wan *et al.* (2020) with the organic phosphorus extracted from NaHCO3 considered labile, and moderately labile was determined by the sum extracted from HCL and fulvic acid fraction obtained from the acidification of NaOH extracts. This acidification produced both fulvic and humic acid fractions. The non-labile fraction is the sum of humic acid and extracts of H2SO4 (Residual P). All extracts were neutralized before the phosphorus determination. The organic phosphorus for each fraction was calculated from the difference between the total phosphorus and its inorganic phosphorus concentration. The phosphate concentrations were determined through colorimetry using Hach DR900 unit following Hach Method 8048. Molybdenum blue was prepared as presented by Murphy and Riley (1962) and are reported as P mg/kg.

Sediment Sorption Capacity (Q_{max}) Determination

The sorption capacity (Q_{max}) of sediment samples from each sampling station was determined using the P sorption index (PSI) method (Bache & Williams 1971). One gram of sediment was weighed and added to 20 mL of 75 mg/L P sorption solution. To avert any effects due to microbial activity, two drops of chloroform were added before the mixture was centrifuged at 2,000 rpm for 30 minutes and the supernatant filtered through Whatman 41 filter papers. The final concentration was determined through colorimetry using Multiskan Sky Microplate spectrophotometer (Thermo Scientific) and compared to a standard curve. The average precision calculated using relative standard deviation is 2.12%. The Q_{max} was calculated as follows:

$$\text{PSI (L/kg)} = \frac{X}{\log C}$$

where:
X = P sorbed (mg P/kg)
C = P concentration at equilibrium (mg/L)

Data Analyses

All statistical analyses including one-way ANOVA, Tukey test and Pearson's correlation were performed using IBM SPSS Statistics software version 29.0.0.0(241).

RESULTS AND DISCUSSION

River Status Based on Physicochemical Parameters

Overall, the water quality of Langat River deteriorated from upstream to downstream, as reported by Hashim *et al.* (2018). Other authors who studied the water quality of Langat River have also shown that the river is moderately polluted based on the Water Quality Index (Syaiffudin & Toriman 2020; Basheer *et al.* 2017; Gasim *et al.* 2015). This study used several physicochemical parameters to assess the water quality, which can provide current indications of the river's health status and the impact of the surrounding environment.

In Langat River, the BOD levels increased downstream with an exceptional rise at M3. This station also recorded the highest COD value. Table 2 lists the results of Tukey's test of physicochemical parameters used in this study between the sampling stations and supports the significant difference of COD level at M3 than the other stations with exception of station D2.

Nonetheless, the increase in BOD and COD levels detected at M3 was not accompanied with lower DO values. In reaches with faster river flows as observed at M3, aeration of the water was increased and thus, more oxygen was dissolved into water. Therefore, during the data collection which was done at the field, the DO still recorded relatively high values (Bulbul *et al.* 2022). Both BOD and COD are strongly correlated ($r = 0.795$, $P = 0.01$) and their significance is demonstrated through the BOD5 : COD ratio, also known as the Biodegradability Index (Rim-Rukeh 2013).

Our study adopted the Biodegradability Index based on the suggestion by Lee and Nikraz (2015) which assesses the toxicity of wastewater to examine the biodegradation ability in the Langat River. The Biodegradability Index indicates the amount of biodegradable organic matter in the total organic matter, where a ratio greater than 0.5 means that it is easily biodegraded, 0.4 – 0.5 means that it has average biodegradability, and 0.2 – 0.4 is very slowly biodegraded (Saravanathamizhan & Perarasu 2021).

BOD is defined as the concentration of dissolved oxygen used by aquatic organisms to oxidize organic matter, whereas COD is the dissolved oxygen consumed to oxidize organic matter and typically, the COD should exceed BOD (Najafzadeh & Ghaemi 2019). All ratios were below 0.2, except for station M5 (BOD5 : COD = 0.21), thus, it was surmised that: a) generally in the Langat River, the biodegradability of organic matter is low and pollution is most likely from inorganic sources; and b) the Langat River cannot naturally degrade organic matter efficiently through biological means.

Organic matter can originate from microorganisms and plants, as well as leach from the banks (Loh *et al.* 2016) and it is also a source of organic phosphorus (Lü *et al.* 2018). In urbanized rivers like the Langat River, the impact of excess phosphorus is of concern and this is usually linked to the anthropogenic activity in the basin. The ability of the river to transform, hold, and sequester organic phosphorus depends on the chemical forms of organic phosphorus introduced and the microorganisms that regulate phosphorus (Watson *et al.* 2018). A reduced capacity to cycle phosphorus, especially in areas where the river functions as drainage for human activities, can lead to increased phosphorus concentrations in the water and sediments possibly triggering eutrophication.

In 2021, up to 59.65% of Langat Basin land was used for agriculture (Selamat *et al.* 2023) and land use patterns, especially irresponsible use directly impact the aquatic environment and are the source of many water quality-related problems (Bu *et al.* 2014). There is an increasing trend observed in the electric conductivity of the Langat River (Fig. 2e) with electric conductivity values well below the Class I limit (1,000 µS/cm) of the National Water Quality Standard Malaysia (NWQS), where the lowest value (27.30 µS/cm) was recorded at

PRA and the highest value (137.34 µS/cm) was recorded at D2. The increase in Langat River electric conductivity can be caused by several factors including pollution from tributaries and land use patterns (Duncan *et al.* 2012).

The land use types around the sampling stations and the anthropogenic activities observed in the vicinity of the sampling station were presented in Table 1. According to Ahmed *et al.* (2022), the midstream to downstream section of Langat River carry high concentrations of heavy metals such as Al, Pb, As and Cd originating from shipping traffic, construction sites, and landfill where rechargeable batteries are dumped. In addition, large-scale plantations have been observed at the downstream stations, D1 and D2 (Fig. 1h and i), where the risk of agricultural runoff containing high concentrations of phosphate and nitrate salts explains the relatively high electric conductivity values downstream. Conductivity measures the electrical conductivity of water and can be used to estimate the total dissolved ions present (Samal *et al.* 2015) and is used as an indicator of mineral-related pollution (Yap *et al.* 2013). The current data in the Langat River supports this assertion, as electric conductivity is strongly positively correlated to BOD and COD and strongly negatively correlated to pH and DO. The negative correlation between electric conductivity and pH suggests that the concentration of hydrogen ions in the water is rising downstream. Among the possible factors that can cause the decrease of pH as electric conductivity increases include leaching of acid and metals into the water although natural decomposition of organic matter by microorganisms can also contribute (Misman *et al.* 2023).

Briefly, the increased pollution from mineral salts causes the deterioration of the other parameters. Electric conductivity does not identify the chemical species present in the waters but can help to understand the pollution trend of the river (Shrestha & Basnet 2018).

Tropical rivers tend to have well-established vegetation on their banks that reduces erosion (Zhang *et al.* 1999) but since a substantial portion of the Langat River Basin is used for agriculture, the introduction of excess nutrients into the river can happen either through surface runoff during precipitation or leaching from soil subsurface. Often, these nutrients are assimilated by aquatic organisms but affect the normal functioning of the ecosystem when in excess (Aristi *et al.* 2015). Limiting nutrients such as phosphates and nitrates are crucial to biological productivity. An increase in biological productivity, such as photosynthesis, is linked to an increase in biomass production, which, when no longer viable through decomposition reduces the amount of oxygen available for living organisms (Spivakov *et al.* 1999). This is evident as compared to the other physicochemical data, oxygen levels in the Langat River deteriorated downstream (e.g., Class IV at station D2).

Table 2 Tukey's HSD test of the water physicochemical parameters including dissolved organic phosphorus in water and total organic phosphorus extracted from the sediments with respect to river sections

Station	pH	DO (mg/L)	BOD (mg/L)	COD (mg/L)	Electric conductivity (µS/cm)	DOP (mg/L)
PRA	7.25 ± 0.11	9.66 ± 2.30a	0.65 ± 0.47	N.D.	27.30 ± 2.82a	0.05 ± 0.03a
UP4	7.38 ± 0.35	8.47 ± 0.53ab	0.25 ± 0.07	9.50 ± 3.50a	27.79 ± 1.36a	0.02 ± 0.01a
M1	7.31 ± 0.15	7.97 ± 0.20ab	0.21 ± 0.20	6.75 ± 1.25a	31.24 ± 3.51ab	0.08 ± 0.08a
M2	7.29 ± 0.04	6.22 ± 1.12abc	0.85 ± 0.80	3.00 ± 0.00a	66.14 ± 31.25abc	0.05 ± 0.02a
M3	7.14 ± 0.46	6.74 ± 0.10abc	3.32 ± 1.47	35.75 ± 0.75c	86.10 ± 23.41abcd	0.05 ± 0.01a
M4	7.21 ± 0.10	4.19 ± 1.36bc	1.75 ± 0.85	13.00 ± 4.00ab	109.30 ± 3.65cd	0.19 ± 0.03b

Station	pH	DO (mg/L)	BOD (mg/L)	COD (mg/L)	Electric conductivity (µS/cm)	DOP (mg/L)
M5	7.20 ± 0.03	5.52 ± 0.55abc	2.48 ± 0.67	11.75 ± 0.75a	91.63 ± 6.05bcd	0.25 ± 0.02b
D1	6.78 ± 0.20	4.09 ± 0.43bc	3.11 ± 1.23	16.50 ± 8.50ab	90.32 ± 14.04bcd	0.22 ± 0.01b
D2	6.90 ± 0.12	2.72 ± 0.16c	2.71 ± 0.67	33.25 ± 5.25bc	137.35 ± 9.57d	0.36 ± 0.01c

Notes: The means with standard deviation values in the same column having different superscript letters indicate a significant difference between them ($P < 0.05$); N.D. = values not available.

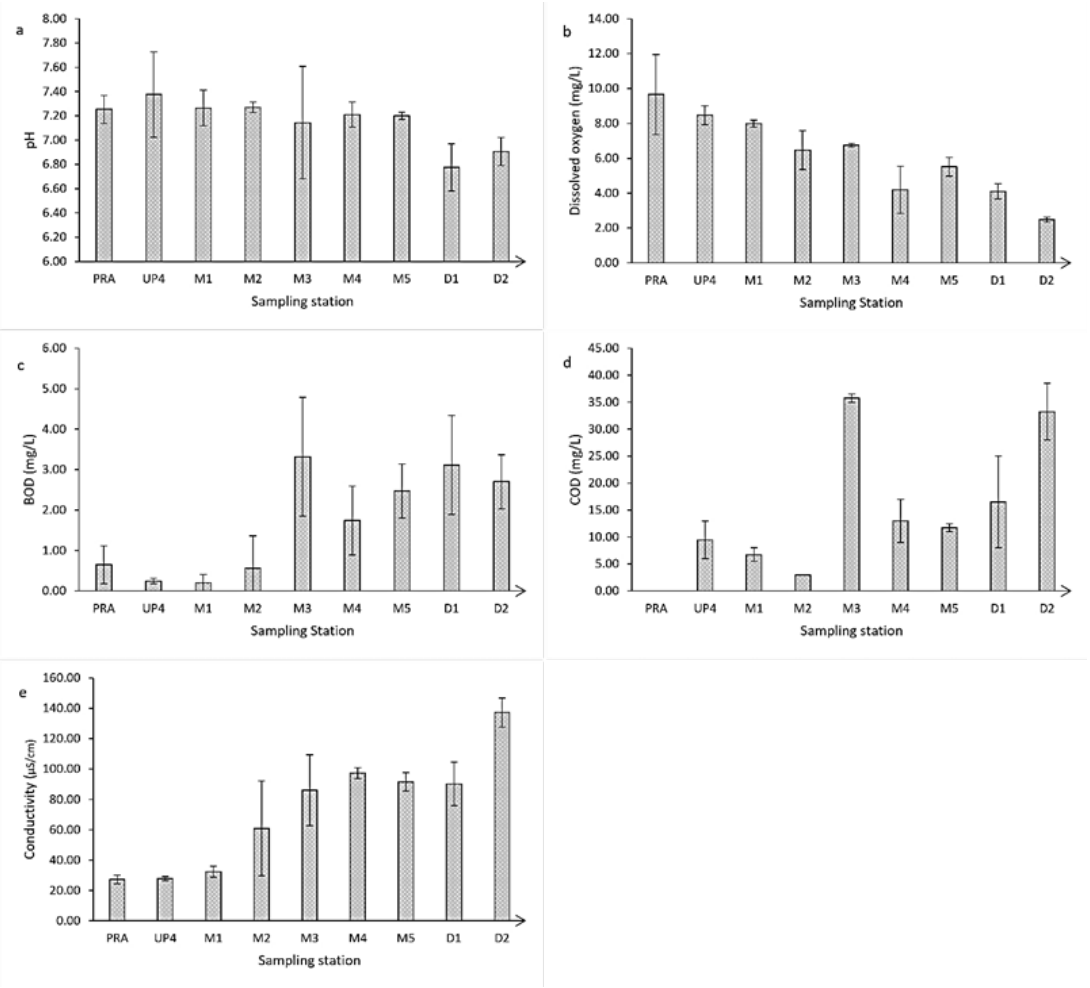

Figure 2 Physicochemical parameters of Langat River measured at the nine sampling stations

Notes: a = pH; b = Dissolved oxygen; c = Biological oxygen demand (BOD); d = Chemical oxygen demand (COD); and e = Electric conductivity. Error bars indicate the standard deviation of the mean values.

Dissolved Organic Phosphorus (DOP) in Langat River

Due to its greater biological activity, dissolved inorganic phosphorus, also known as orthophosphate, is frequently used to quantify the amount of phosphorus in water bodies. This allows for the determination of whether the water body contains enough phosphorus to enable the exponential growth of algae. According to the NWQS, the maximum phosphorus concentration is 0.2 mg/L, which is classified as Class IIA/B. This means that conventional water treatment for human use is needed and is the limit for sensitive aquatic organisms in fisheries. Figure 3 (b) shows the DIP concentration in the Langat River and two stations, M3 and D2 exceeded the limit set by NWQS.

The current standard in phosphorus monitoring is not all-encompassing as it ignores the contribution of DOP. In recent years, DOP has been understood as a potential source of bioavailable phosphorus important for algal growth and eutrophication (Ni *et al.* 2021). Mineralization of DOP through acid and alkaline phosphomonoesterases, nucleotidases and phytases produced by plants and microorganisms is a primary process in phosphorus cycling of DOP into bioavailable orthophosphate (Li *et al.* 2021). At each station selected for this study, the relative contribution of DOP to the total dissolved phosphorus varies. The lowest percentage contribution was found at M3 (11.5%) and the highest at PRA (64.1%) (Fig. 4). Despite the vast difference in DOP percentage contribution at PRA and M3, the DOP concentrations were very close to each other at 0.054 mg/L and 0.046 mg/L respectively. However, the concentration of DIP at M3 was considerably higher than the stations before it.

Increased human activities are often correlated with an increase in DIP loading that originates from point sources rather than non-point ones such as sewage and industrial effluents introduced into the water (Harrison *et al.* 2010). This phenomenon was more likely to occur at station M3 of our study. The basin at M3 is more densely populated compared to stations located upstream and there are more industries including a wastewater treatment facility and a cement factory nearby. Besides, station M3 is also close to a tributary of Langat River namely Balak River which itself flows through the industrial town of Balakong before merging at the confluence in M3.

In terms of DOP percentage of contribution, the highest was found at station PRA which was the first upstream station and the least polluted although the total dissolved phosphorus is one of the lowest throughout the river (Class I physicochemical parameters). The changes observed in the contribution of DOP to TDP are associated with the land use surrounding the stations. The land surrounding station PRA is characterized by forests, villages and small-scale agriculture. In areas with near-pristine surroundings where orthophosphate is low, DOP is used as a source of bioavailable phosphorus (Baldwin 2013). The increased DOP downstream, on the other hand, may be linked to the increase in human population and its activities.

From station M4 onwards, there was a pronounced increase in the concentration of DOP. At stations M4, M5, D1 and D2, the DOP concentrations were 0.19 mg/L, 0.25 mg/L, 0.22 mg/L and 0.36 mg/L, respectively, which were all either close to the NWQS limit or exceed it. The DOP concentrations also were much higher than the DIP concentrations. Station M4 was located within the area of a major city in the state of Selangor where many industries and even a university is located. Food and beverage industries have been noted to be an important polluter and source of organic phosphorus in Langat River's waters however unethical dumping of food wastes from residential areas may contribute a significant amount that is often overlooked (Basheer *et al.* 2017). After M4, the landscape of Langat Basin becomes dominated with agriculture and its associated industries. At station D2 where agriculture is the main land use type, the DOP concentration is significantly higher than the other station (Table 2). According to Selamat *et al.* (2023), agriculture is the dominant land use (59.65%) in the Langat Basin because the land is flat downstream making it more suitable for large-scale plantations (Lim *et al.* 2023).

These represent a sizable portion of potentially bioavailable phosphorus in Langat River. Previous studies have pointed out that DOP tend to make up larger percentages of TDP in the lower reaches of rivers (Hee *et al.* 2018; Ni *et al.* 2016; Mudryk 2004). The nutrient availability downstream is higher since rivers have lower water retention capacity making the downstream a sink for allochthonous sources. Unlike unreactive chemical species that are more likely to reach downstream, reactive substances have a more complex relationship as they can participate in ecological processes (Teurlincx *et al.* 2019). DOP is made of a variety of chemical species and not all are part of the bioavailable phosphorus pool and thus take part in ecological processes. Those that can be hydrolyzed by microbial enzymes include labile monoester, diester and phytate-like phosphorus (Ji *et al.* 2017).

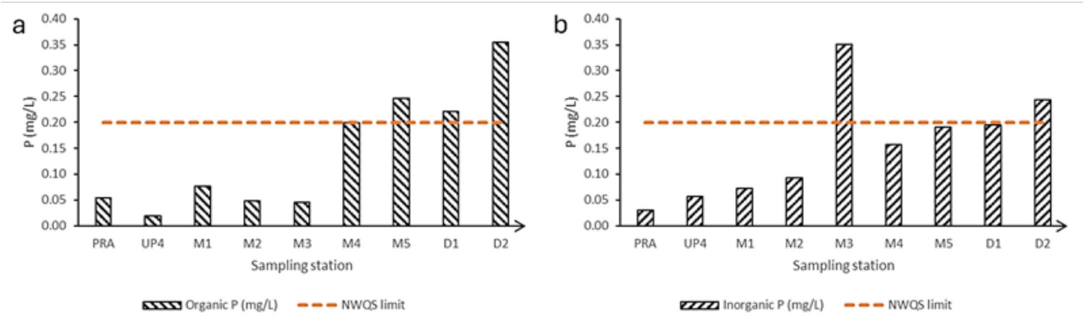

Figure 3 (a) Dissolved organic phosphorus (DOP) and (b) Dissolved inorganic phosphorus (DIP) (orthophosphate) concentration in all sampling stations

Notes: The dotted line marks the threshold limit for P pollution according to National Water Quality Standard Malaysia (NWQS).

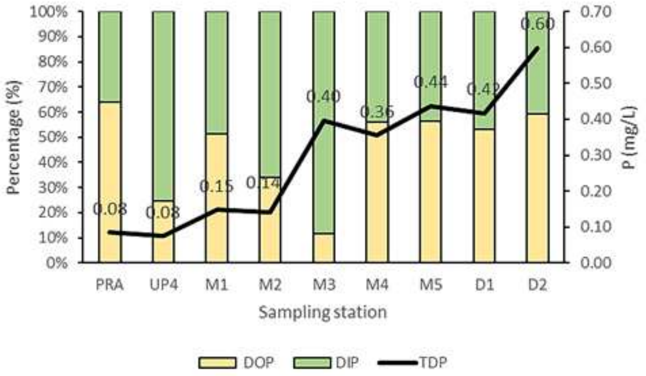

Figure 4 Percentage of DOP and DIP contribution (refer primary y-axis) at each of the nine sampling stations along Langat River

Notes: The total dissolved phosphorus of each station is represented by the line graph and the corresponding values are mentioned in the graph.

Sediment Organic Phosphorus and its Contribution to the Langat River

Fractionation separates the organic phosphorus in the sediments into either labile, moderately labile or non-labile fractions. According to Wan *et al.* (2020), the labile and moderately labile fractions are the most bioavailable forms. The relative contribution of organic phosphorus in the sediments is presented in Figure 5 with the most total organic phosphorus recorded at station D2 (105.68 mg/kg P), while the least was recorded at station M5 (35.20 mg/kg P). The highest non-labile contribution was found in station D2 (74.1 mg/kg P) and the least recorded value was at station M5 (24.36 mg/kg P). The highest labile contribution to sediment was found at station D2 (11.41 mg/kg P), whereas station UP4 only recorded 3.09 mg/kg P. Regarding the moderately labile fraction, the highest load was discovered at station D2 (20.17 mg/kg P) as station M5 had the least (3.40 mg/kg P). In general, station D2 records the highest organic phosphorus load for every fraction extracted from sediments of Langat River. Station D2 is situated at the lower stream in the town of Labohan Dagang where the land use is dominated by agricultural plantations.

One of the environmental issues found occurring at riverbanks of Langat River at Labohan Dagang is erosion where 65.5% was reported to be gully erosion (Abidin *et al.* 2017). Erosion events are exacerbated after rainfall and sedimentation of suspended solids that include eroded agricultural soils from the vicinity and organic matter as well as other organic phosphorus particulates flushed from the upstream accumulate at the downstream during sediment deposition as the flow rate decreases (Ruttenberg 2014).

The non-labile fraction dominated the total organic phosphorus extracted from all sampling stations and comprised extracts of humic acid and residual organic phosphorus. According to Ahlgren *et al.* (2006), smaller contributions by labile and moderately labile forms of organic phosphorus indicate a greater degree of pollution due to active microbial mineralisation. In this study, the labile fraction and DOP are very strongly correlated (r = 0.931, P < 0.01). However, the moderately labile fraction is not significantly associated with DOP.

The strong linkage between the labile fraction and DOP could signify the detachment of sediment-bound organic phosphorus into the river water and is also supported by the strong positive correlation with electric conductivity and a strong negative correlation between pH and dissolved oxygen to DOP and the labile fraction (Table 3). According to Gardolinski *et al.* (2004) and Søndergaard *et al.* (2013), sediments release phosphorus when there is a decrease in pH and dissolved oxygen concentration, such as downstream of the Langat River. In addition, sediment-released organic phosphorus is also determined by intense biological and chemical activity at the sediment-water interface and could be mediated by microorganisms present on the sediment (Huo *et al.* 2011).

The large contribution of non-labile organic phosphorus in the sediment is commonly attributed to organic matter and agricultural runoff as phytate, a major component of the non-labile fraction, accumulates in sediments where pH is low and the clay content is high (Watson *et al.* 2018; Huo *et al.* 2011). Most sampling sites in this study were not in the vicinity of agriculture except M5, D1 and D2, sites with high non-labile contributions, indicating that the sediments of Langat River have considerable abilities to retain and sequester organic phosphorus which is dependent on its sorption capacity.

The maximum sorption capacity (Qmax) of sediments from each sampling station with station M3 recording the lowest Qmax value (83.3 mg/kg P), while station D2 recorded the highest (597.7 mg/kg P) (Fig. 6). A study performed by Mahmud *et al.* (2022) at Langat River reported that the particle size of sediments decreased toward the downstream. Sediment phosphorus sorption can be influenced by sediment properties such as the particle size where decreasing particle size is associated with the increase in sorption capacity. However, other factors that require further investigation may also play a significant role including organic matter content and minerals present in sediments (Wang *et al.* 2006).

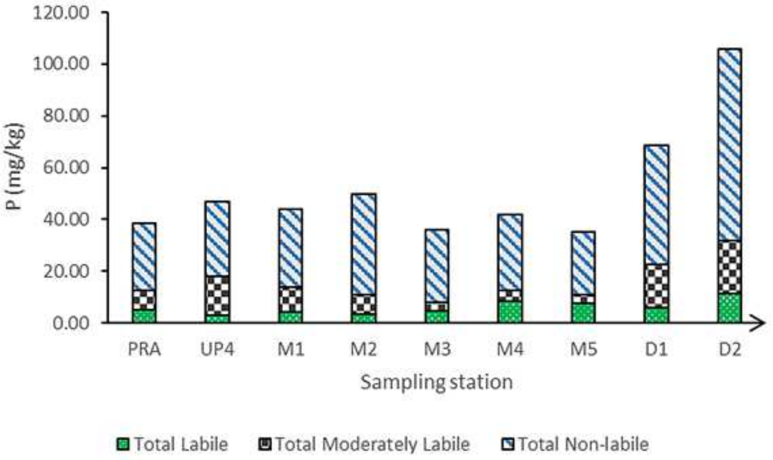

Figure 5 The total organic phosphorus extracted and contributions of each fraction for all sampling stations

Table 3 Correlation matrix of physicochemical parameters, DOP concentration and sediment organic phosphorus fractions separated into labile, moderately labile and non-labile fractions.

	DOP (mg/L)	**pH**	**EC (µS/cm)**	**DO (mg/L)**	**COD (mg/L)**	**BOD (mg/L)**	**Labile (mg/kg)**	**Non-labile (mg/kg)**	**Moderately labile (mg/kg)**
DOP (mg/L)	1								
pH	-0.707*	1							
EC (µS/cm)	0.855**	-0.705*	1						
DO (mg/L)	-0.876**	0.737*	-0.945**	1					
COD (mg/L)	0.444	-0.555	0.734*	-0.585	1				
BOD (mg/L)	0.608	-0.767*	0.812**	-0.688*	0.795*	1			
Labile (mg/kg)	0.931**	-0.544	0.837**	-0.797*	0.491	0.522	1		
Non-labile (mg/kg)	0.667*	-0.691*	0.656	-0.702*	0.511	0.351	0.642	1	
Moderately labile (mg/kg)	0.375	-0.540	0.195	-0.335	0.224	0.056	0.266	0.774*	1

Notes: DOP = Dissolved organic phosphorus; EC = Electric conductivity; DO = Dissolved oxygen; COD = Chemical oxygen demand; BOD = Biological oxygen demand; * = Significant correlation at the 0.05 level (2-tailed); ** = Significant correlation at the 0.01 level (2-tailed).

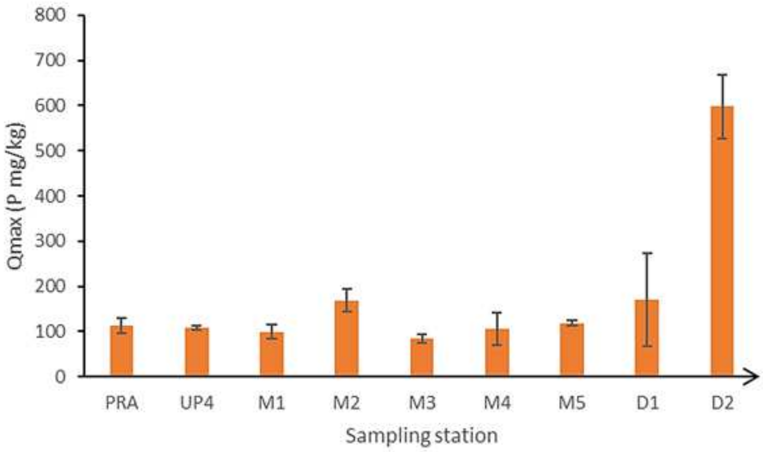

Figure 6 The maximum P sorption capacity (Qmax) based on the Phosphorus Sorption Index (PSI)

Note: The error bars indicate standard deviation.

Comparing the total organic phosphorus extracted through fractionation the Qmax, total organic phosphorus accounts for 17.6% to 44.3% of the calculated Qmax. In phosphorus-stressed environments, such as eutrophic rivers, anoxia and harmful algal blooms often occur (Aristi *et al.* 2015) but these signs were neither observed at Langat River during data collection nor reported previously. This suggests that at present, the organic phosphorus sources in the river subsidize the ecosystem and the phosphorus retention capability of sediments prevent rapid release to the water column. Nevertheless, high sediment preservation of organic phosphorus can be a potential source of internal loading if external sources are reduced and thus any planned mitigation efforts to prevent any serious consequences in the future must seriously consider the non-labile fraction.

CONCLUSION

The water quality of Langat River decreases from upstream to downstream, as evidenced by the degradation of all the water physicochemical parameters tested in this study. Dissolved oxygen, biological oxygen demand and chemical oxygen demand were identified to have significant deterioration and can affect the ecological functions of Langat River. Similarly, organic phosphorus load both dissolved in the water and the sediments also increase towards the downstream. Dissolved organic phosphorus constitutes a significant portion of the total dissolved organic phosphorus and is a possible significant source of bioavailable phosphorus for aquatic photosynthesizing organisms and microorganisms. Sediment deposition at the downstream from allochthonous sources, sediment sorption capacity, matter introduced from tributaries and land use types including agriculture affect the distribution of organic phosphorus in the Langat River. The non-labile fraction of the sediment presents a source of internal loading and a probable eutrophication risk if loading is not controlled.

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