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Research paper

Longitudinal study of N compounds during flood neap and spring tides in the Terengganu River estuary, Malaysia

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Abstract

This study was performed in the Terengganu River estuary, which is connected to the southern part of the South China Sea, to determine the concentrations of surface water nitrogen (N) compounds during a longitudinal survey. In addition, measurements of chlorophyll-a (chl-a) and the fractionation of total DON were also carried out. In general, higher concentrations of N compounds were recorded in lower salinity regions with many anthropogenic activities. The spring tide showed a significant role in controlling the N compounds as higher concentrations were found under this condition. This was probably due to strong currents which led to the resuspension of bottom sediment, thus releasing N compounds. Most of the N compounds showed addition behaviour along the salinity gradient which was more pronounced during the spring tide. A higher percentage of low molecular weight (LMW) total DON was recorded during the neap tide. The LMW fraction correlated well with chl-a concentrations suggesting that the source of this fraction is through direct release from phytoplankton. This longitudinal study shows the importance of measuring N compounds under different tidal conditions for a better understanding of the distribution of N compounds in the estuarine environment.

Keywords: *Surface Water; Nitrogen Compounds; Longitudinal; Behaviour; Fractionation.*

1. Introduction

Estuaries are unique regions that have a combination of both freshwater and saltwater and are the major conduits between the land and sea. Rivers influence the transport of soluble and particulate materials derived from the catchment area to coastal waters through estuaries, and tides are a significant agent of transport of these loads in most coastal environments [1]. Spring and neap tides have been recognized as playing a predominant role in controlling the distribution and transport of nutrients in estuaries. This has been documented for the Bonny estuary [2], the Zuari estuary [3], and the Terengganu River estuary [4]. The spring tide is associated with strong turbulent energy compared to the neap tide, as the amplitudes of spring tidal currents are approximately twice those of neap tides [5]. Spring tides strengthen the mixing of seawater between inside and outside of estuaries and cause stronger bottom stirring which results in the release of high levels of nutrients from bottom sediments to the surface water [3,4]. Therefore, it is necessary to understand the hydrodynamical effects induced by tides in order to explore the distribution of nutrients in estuaries.

Nutrient loads to estuaries have increased considerably as a result of anthropogenic activities and this deteriorates the water quality of the surrounding area [6].These threats affect freshwater sources and coastal areas through eutrophication and may lead to hypoxia event intensification. Thus, efforts to control estuarine eutrophication require a thorough understanding of the sources of nutrients, whether from inland, within the estuary or from the sea. The sources and sinks of nutrients within estuaries have been traditionally detected and quantified by a theoretical conservative mixing model proposed by Liss et al. [7]. The model is the plots of nutrients as a function of salinity to demonstrate the conservative or non-conservative behaviour of nutrients in an estuary from a mixing line of freshwater and seawater [8]. Nitrogen (N) compounds have been chosen in this study since they are often identified as limiting nutrients to phytoplankton growth in the marine environment. Additionally, most eutrophication is driven by N [2].

Our previous works have shown that tides play a major role in controlling the distribution of N compounds in the Terengganu river estuary during ebb tide conditions and have also demonstrated non-conservative behaviour of No compounds with an addition tendency in this estuary [6], and the goal here is not to repeat but to complement this work. The main focuses of this study are (1) to investigate the variation of N compounds i.e. nitrate, ammonia, dissolved organic nitrogen (DON) and particulate organic nitrogen (PON) along the surface water of the Terengganu river estuary, focusing on the effects of spring and neap tides during flood flow conditions, (2) to demonstrate the behaviour of the parameters measured during estuarine mixing using the model proposed by Liss et al. [7] and (3) to identify the concentrations of size-fractionated DON in order to further understand its possible sources.

2. Methodology

The study area is the Terengganu River estuary (TRE), situated next to the southern waters of the South China Sea (Figure 1). Generally, this estuary is categorized as a small estuary with a total area of about 8 km^2 [9]. It receives run-off from the Terengganu and Nerus Rivers. Tides in the TRE are typically semi-diurnal with tidal ranges during neap and spring tides of about 0.7-2.0 m and 0.5-2.5 m, respectively. This study was carried out during a neap (2/4/2012) and spring tide (10/6/2012) under similar tidal conditions (flood flow) along the TRE. A total of 24 stations were selected, covering the upstream and downstream areas. The surface water samples were collected at approximately 0.5 m depth using a Van Dorn sampler and placed directly into acid-washed polyethylene bottles. These were immediately kept in an icebox and transported to the laboratory. Samples were filtered through pre-combusted (450 ºC for 5 h) Whatman GF/F filters (0.7 µm) immediately upon arrival at the laboratory. The dissolved samples were stored frozen (-20 ºC) and loaded filters for chl-a analysis were folded and placed in a polyethylene centrifuge tube wrapped with aluminium foil and also frozen for further analysis. In addition, the particulate matter retained on the filters was kept in a desiccator overnight and then subjected to PON analysis. Pre-filtered 100 mL water samples from eight selected stations were filtered again through tangential flow ultrafiltration vivaflow 200 (Sartorius stedim) with a molecular weight cut-off of <10 kDa.

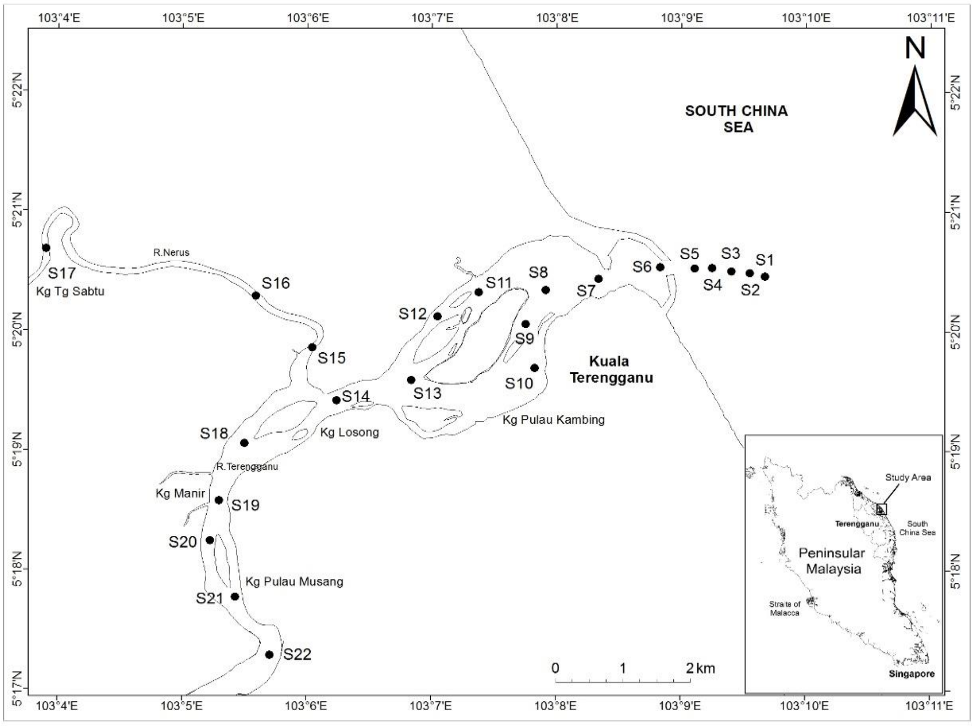


Fig. 1: Map Showing the Sampling Stations in the Study Area.

In laboratory analyses, dissolved inorganic nitrogen (DIN), i.e. nitrate and ammonia, was determined colourimetrically, where concentrations of nitrate were analysed using the Cu-Cd column reduction method and ammonia with indophenol blue colour formation and detected spectrophotometrically as a coloured complex with a continuous flow analyser (Skalar San plus) as followed by Kirkwood [10]. Dissolved organic N (DON) was determined by measuring TDN and subtracting the DIN concentrations. TDN was measured based on the high-temperature catalytic oxidation (HTCO) method with a Shimadzu TOC-VCPH total organic carbon analyser equipped with a TNM-1 total nitrogen measuring unit as described in detail by Suratman et al. [11]. Weight fractionation of DON was carried out using an ultrafiltration technique. The ultrafiltrate from this technique was collected and the concentration of TDN in this fraction was measured accordingly and then the DIN concentrations were taken from this. The DON in this ultrafiltrate is then referred to as low molecular weight (LMW) [11]. In addition, chl-a analysis was undertaken according to Parson et al. [12], and the concentration in the extracted filtrate mixed with 90% acetone for 24 h was determined using a Shimadzu 1201 spectrophotometer.

3. Results and discussion

3.1. Longitudinal distribution

The distributions of N compounds in the TRE for neap and spring tides are given in Figure 2. The nitrate concentrations ranged between 1.2 and 4.7 µM and the two-way ANOVA test showed nitrate concentrations were significantly different ($p<0.05$) between sampling trips and sampling stations. When comparing the range concentrations of nitrate between the neap and spring tides, it was seen that nitrate in the surface waters was higher in the spring tide (1.2-4.7 µM) compared to the neap tide (1.4-4.0 µM). During the spring tide, stronger water turbulence with a high tidal range may result in direct contact between the surface and bottom waters, leading to stronger bottom mixing. Stronger bottom mixing tends to increase the nitrate released from the resuspended bottom sediment to the surface water. Similarly, a study by Sharples at el. [5] obtained higher nitrate during spring tide compared to neap tide and indicated that the strong turbulence during the spring

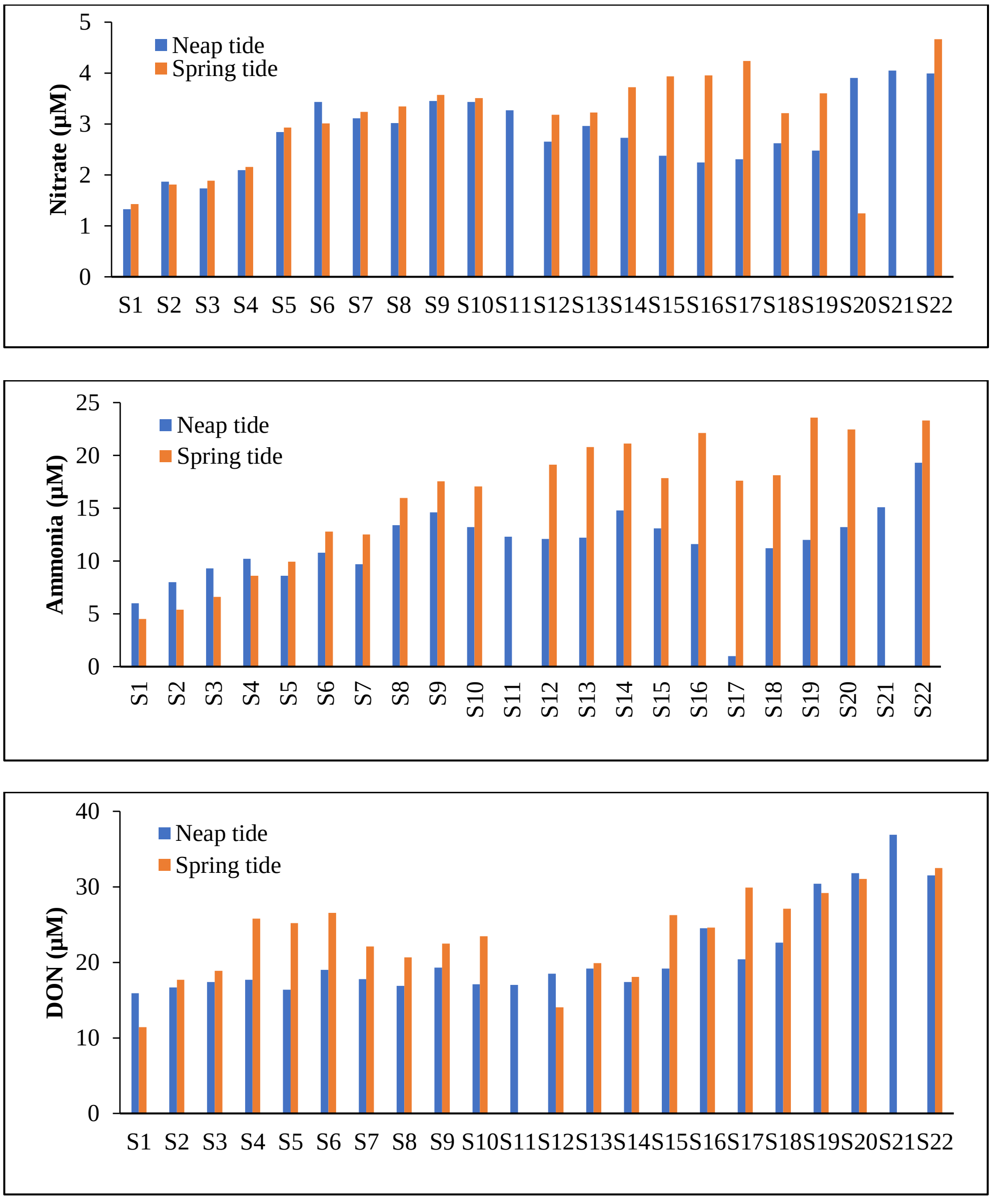


Fig. 2: Longitudinal Distribution of N Compounds in Terengganu River Estuary.

Tide affected sediment resuspension and in response caused high nitrate release. A similar explanation was given by Anand et al. [3] who recorded a higher range of nitrate concentrations during the spring (12.7-16.2 µM) compared to the neap tide (9.5-10.8 µM) due to strong turbulence. Lallu et al. [13] found that the variation of nutrients depends on tidal and freshwater flow – high tidal range and increased freshwater flow during the spring tide influenced the higher nitrate concentrations observed at all sampling station in the Cochin estuary, India.

In this study, the ammonia concentrations varied from 4.5 to 23.6 µM. The two-way ANOVA test showed the ammonia concentrations were significantly different ($p<0.05$) between sampling surveys, but not significantly different ($p>0.05$) between sampling stations. A higher range of ammonia concentrations was found during the spring tide (4.5-23.6 µM) compared to the neap tide (17.6-23.3). Similar to nitrate, the large tidal range generated a strong current which leads to resuspension of bottom sediments during the spring tide. The ammonia in the bottom sediment was released, leading to an increase of ammonia in the surface water. The reverse of this observation was seen during the neap tide, with low ammonia concentrations. The same observation was reported by Gilbert et al. [14] in the Columbia River estuary, where higher ammonia concentrations were recorded during the spring tide rather than the neap tide. The authors indicated that the strong current and well-mixed water column during the spring tide resulted in a more turbid estuary. As a result, the disturbed sediment caused increased ammonia to be released. Similarly, Anand et al. [3] also found higher ammonia concentrations in the Zuari estuary, India during the spring tide and attributed the higher concentrations to the higher tidal range during the spring tide which enhanced the water turbulence and facilitated the ammonia release from the bottom sediments. In addition, Tanaka & Choo [15] reported significantly higher ammonia concentrations during the spring tide in the Matang Mangrove estuary, Malaysia which was due to the effect of tidal mixing caused by the spring tide. The authors indicated that the water was vertically well-mixed during the spring tide and this resulted in the resuspension of mud flat sediment and lead to the release of a high amount of ammonia.

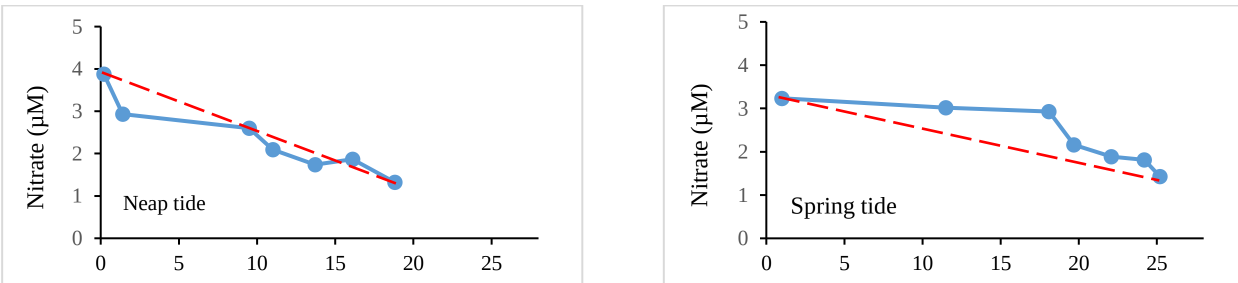
The concentrations of DON found in this study ranged from 11.4 to 36.9 µM. The two-way ANOVA test showed the DON concentrations were significantly different ($p<0.05$) between sampling surveys, but not significantly different ($p>0.05$) between sampling stations. Comparisons of DON levels between neap and spring tides indicated that the range of concentrations of DON was higher during the spring tide (11.4-32.5 µM) compared to the neap tide (15.9-36.9 µM). Other studies have recorded a reverse trend, with a high range of concentrations of DON during the neap tide in comparison to the spring tide. For example, a study by Liu et al. [16] in Jiaozhou Bay, China suggested that DON derived from land sources is higher during the neap tide due to low water turbulence which disables the dispersion of nutrients in the surface water and results in the high DON concentrations observed during the neap tide. In addition, Kaiser et al. [1] also found a higher value of DON during the neap tide instead of the spring tide for both the spring and summer seasons. The authors suggested that DON derived from livestock waste, and possibly low water turbulence during the neap tide, led to higher DON availability in the surface water. However, this was not observed in the TRE as a lower range of DON concentrations was found during the neap tide. This was probably related to strong turbulence that resulted in the resuspension of bottom sediments and vertical mixing which was induced by strong tidal currents during the spring tide. As a result, these processes accelerated the release of high levels of DON. The trend observed was similar to the studies by Sharples et al. [5] and Anand et al. [4], which showed that the spring tide affected (strengthened) sediment resuspension and vertical mixing of the water column. In addition, a contribution of DON from the resuspension of organic matter from bottom sediments was observed by Suratman et al. [4] and Zhang et al. [17]. Both of these studies recorded increases in DON concentrations in the water column as a result of mixing driven by storms and wind.

Throughout the study, the PON concentrations ranged between 6.0 and 18.8 µM and the two-way ANOVA test showed PON concentrations were not significantly different ($p>0.05$) between sampling trips or sampling stations. Comparison of PON levels between the neap and spring tides indicated that the range of concentrations of PON was lower during the neap tide (10.5-18.8 µM) compared to the spring tide (6.0-18.3 µM). This high range of concentrations in the spring tide compared to neap tide was similar with the ranges found for other N compounds. Kristensen & Suraswadi [18] also recorded that PON was higher during the spring tide rather than the neap tide and indicated that fast water currents during the spring tide resulted in higher turbidity, which may increase PON availability.

3.2. Behaviour

The N compounds–salinity relationship in the TRE during neap and spring tide surveys are shown in Figure 3. Generally, the nitrate concentrations were higher at the freshwater end-member than the coastal seawater end-member. These results revealed that the source of nitrate was the freshwater region. Nitrate concentrations behaved non-conservatively, with removal and addition recorded during the neap and spring tides, respectively. The removal of nitrate was observed in the middle salinity region (0-15 ppt). This was possibly due to a chemical denitrification process process which takes place in the estuary. Generally, the denitrification process favours low oxygen conditions in the water column where nitrate will be reduced to ammonia, which eventually leads to high ammonia concentrations. This observation is supported by the removal of DO that was observed by Salum et al. [19] in the TRE during the same survey. In addition, this was consistent with the ammonia value, which was found to behave non-conservatively with addition processes. This result provides a clear indication that denitrification processes may take place in the estuary and low concentrations of nitrate may lead to high ammonia concentrations due to the influence of low DO concentrations. The denitrification process has been widely cited as a possible mechanism for nitrate removal during estuarine mixing. Examples include the Wenjiaohe estuary, China [20] and the Columbia River estuary, USA [14]. In contrast, the addition of nitrate was observed in the estuarine area, especially in the higher salinity region. The additional source of nitrate could be associated with anthropogenic activities in the TRE. Previous studies by Suratman et al. [21] indicated that several activities in the TRE such as Pasar Payang, fish landing at the jetty, boating activities, tourism, and direct domestic sewage discharge may lead to high nitrate concentrations in surface water. In addition, Jickells et al. [6] also found that the TRE was impacted by a secondary source of nutrients (sewage input) which may encourage the non-conservative behaviour of nutrients.

Similar to nitrate, higher concentrations of ammonia were also found in the freshwater compared to the seawater end-member, indicating the input of this particular nutrient was also from the freshwater area. Non-conservative behaviour with an addition tendency was observed for ammonia in both surveys. The maximum input of ammonia was more pronounced during the spring tide at salinities of 10-15 ppt. In the present study, significant removal of nitrate and DO content [19] was found during the neap tide, clearly showing that predominant denitrification processes inside the estuary were possible sources of the ammonia content in the water column. Similarly, Tahir et al. [8] found an additional source of ammonia in the Dungun River estuary that was attributed to the denitrification process which converts oxidised nitrogen species, e.g. nitrate and nitrite, to ammonia in water with low DO content. The observation made by Liu et al. [20] showed the addition of ammonia was more pronounced at low salinity regions, which was comparable to the present results. The authors suggested that the input of ammonia was derived from anthropogenic activities such as agricultural activities, i.e. extensive use of chemical fertilizer, degradation from organic matter, and natural denitrification processes within the estuaries. In addition, the additional source of ammonia can be attributed to the resuspension of bottom sediment. For example, Kadiri et al. [22] found an addition source of nutrient in the Severn estuary, UK and suggested the higher input of nutrients can be associated with internal sources such as resuspension from the estuary bed. The resuspension of bottom sediment will release nutrients into surface water and indirectly increase the nutrient concentrations there. The present study revealed that the concentrations of ammonia in the estuary were contributed to by the internal process of denitrification and external process of anthropogenic activities.



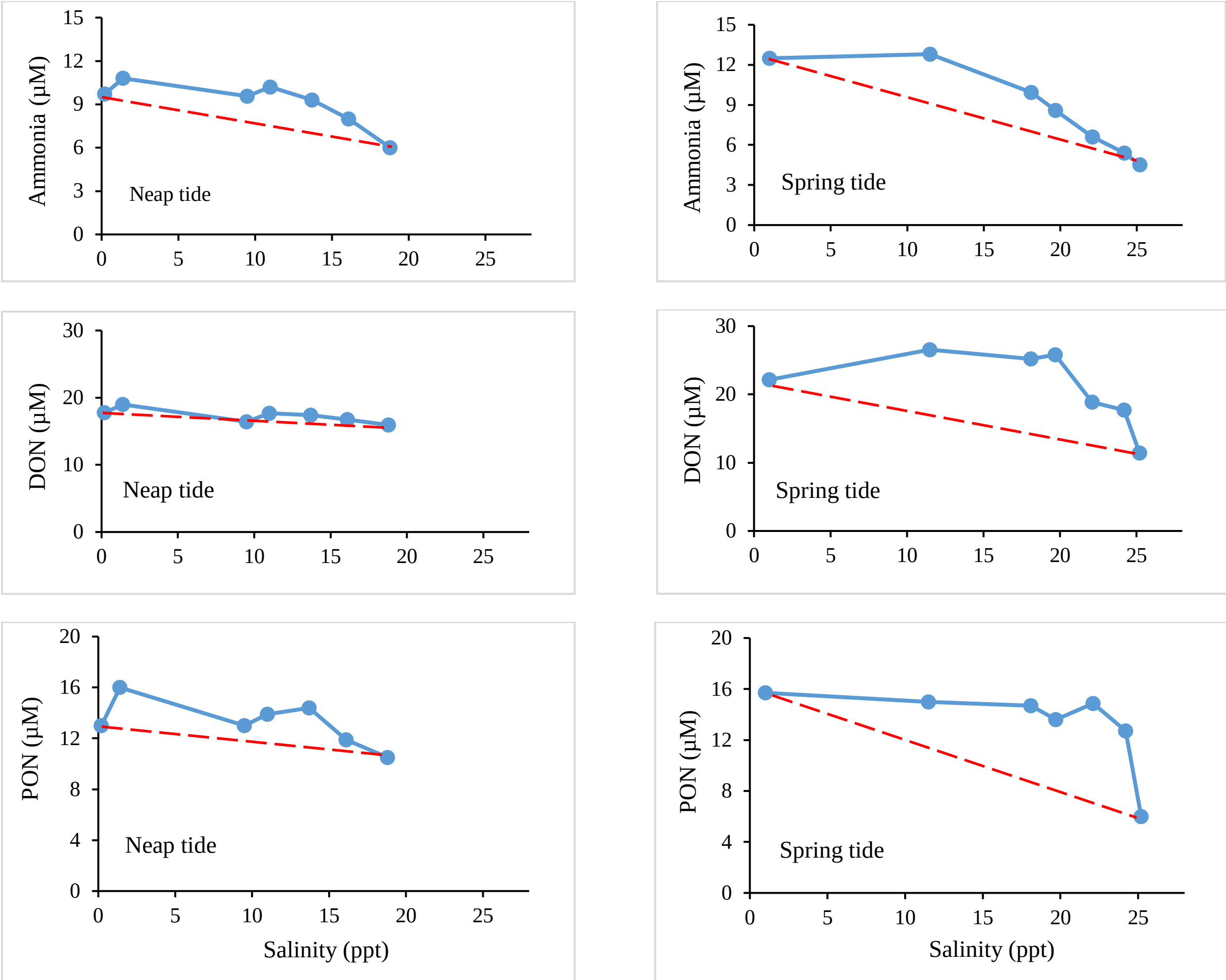


Fig. 3: Behaviour of N Compounds in Terengganu River Estuary.

The effect of mixing patterns between freshwater and coastal seawater on DON in the TRE showed that non-conservative behaviour was found for neap and spring tide surveys, with addition of DON. In general, the DON concentrations were more concentrated at the freshwater end-member compared to the coastal seawater end-member and gradual decreasing concentrations were seen as salinity increased. This indicated that the DON in this estuary was mostly from riverine input. An additional source of DON concentrations could be anthropogenic activities from upstream and the urban area of Kuala Terengganu town. Several studies have also found that anthropogenic activities contributed to the addition of DON in the water column. For example, Kaiser et al. [1] found the non-conservative behaviour of DON in the Nanliu estuary, China and indicated that an input of DON from livestock waste as a strong source of DON concentrations in the inner part of the estuary. Therefore, the addition of DON in the TRE was mainly due to the anthropogenic activities such as domestic sewage, economic activities, livestock farming, boat manufacturing, aquaculture activities and tourism along the Terengganu River which flows into the low and middle salinity regions.

The results obtained for both surveys demonstrated non-conservative behaviour with addition of PON. In general, the concentrations of PON decreased in higher salinities but exhibited a wider range of PON concentrations in the lower salinity reaches. This indicated that the PON in this estuary could be derived from riverine input. There were several sources which could explain the addition of PON in this area. Among the anthropogenic activities that could lead to an increase in PON concentrations are domestic sewage from residential areas and effluents from activities in both rural and urban areas that discharge directly into the water column. It is possible that the river transported mineral-rich material such as detritus from terrestrial plants and the resuspension of sediment correlated to the tidal conditions, water current and wind may release more PON to the water column. Furthermore, the addition of PON in the estuary was not only from land-derived and riverine input but might be influenced by other mechanisms such as autochthonous materials from the aquatic system (i.e. phytoplankton and bacteria) that contained a high level of organic matter which contributed to the concentrations of PON in the water column. Abril et al. [23] reported 70% of PON from various sources in the Gironde estuary, France was carried by the rivers and recycled by mineralization, especially in the turbid area of estuaries, because PON is part of suspended particulate matter (SPM). Thus, an addition of PON in the TRE corresponds to input from upstream and land-based activities.

3.3. Fractionation

The percentage abundance of the LMW fraction in total DON for the longitudinal surveys is illustrated in Figure 4. Throughout the study, the percentage of LMW DON was in the range 58 to 93% (mean 78±13%) and 45 to 71% (56±11%) for neap and spring tides, respectively. However, the two-factor ANOVA test showed there was no significance difference ($p>0.05$) between sampling surveys and sampling stations. The LMW DON showed distinct variations between neap and spring tides as a higher percentage was recorded during the neap tide compared to the spring tide. These results revealed that the LMW DON fraction accounts for most of the total DON during neap tide surveys, but for spring tides, most of the DON fraction was in the form of HMW DON. This relationship was expected to be driven by the water current. In general, the stronger tidal currents during spring tides were able to increase the resuspension of bottom sediments which contain high HMW fractions, thus directly decreased the LMW DON fraction in the surface water [4,24]. This could be a response to the low percentage of LMW DON present in the surface water column during the spring tide.

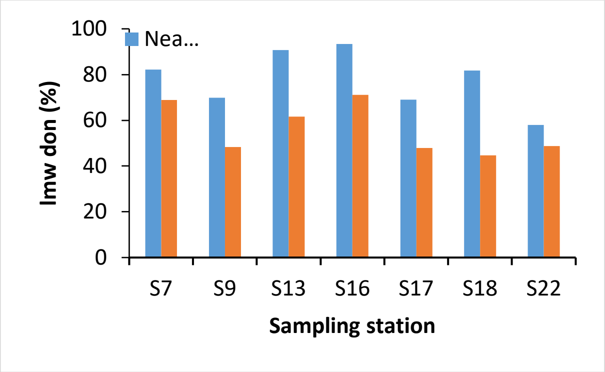


Fig. 4: Percentage Abundance of LMW DON at Selected Stations.

The relationship between LMW DON with chl-a was used in order to investigate the possible source of LMW DON in the surface water. Chl-a is often used as an indicator of phytoplankton biomass in the water column [24]. The linear regression between LMW DON and chl-a for this study is shown in Figure 5. A higher positive correlation (R^2=0.60, R=0.78, n=14) was recorded between LMW DON and phytoplankton. A high positive correlation indicates a higher percentage of LMW DON, which corresponds to a high concentration of chl-a. The LWM DON fraction was from biological origin, which is related to phytoplankton biomass that is associated with high chl-a concentration. Many studies have suggested that the source of LMW DON in the water column is attributed to an internal source, most likely direct release from the phytoplankton via excretion [11], [25]. In addition, Suratman et al. [11] recorded a higher percentage of LMW DON in the North Sea during autumn and spring and attributed those increases to high chl-a concentrations as corresponding to phytoplankton biomass. The authors suggested that that DON released by phytoplankton was likely to be the main source of LMW DON.

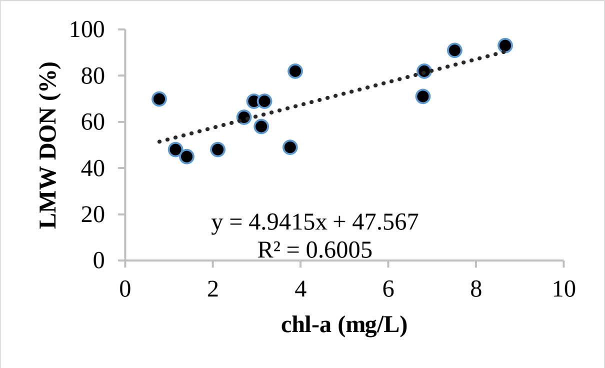


Fig. 5: Relationship between LMW DON and Chl-A for Terengganu River Estuary.

4. Conclusion

This study has shown that higher concentrations of N compounds were found at the freshwater end-member sampling point compared to the seawater end-member sampling point, suggesting that the main inputs of these nutrients to the TRE were mainly from upstream of the river. Nevertheless, anthropogenic activities within the estuary also contributed to the nutrient concentrations in this area. Spring and neap tides play a major role in controlling the concentrations as a higher range of N compounds were recorded during the spring tide and a lower range for the neap tide. This was due to the higher tidal range for the spring tide which created stronger currents and more turbulence. This affected the sediment suspension and released nutrients from the bottom to the surface water. There was a high positive correlation between LMW DON and chl-a and this was most likely through direct release from the phytoplankton via excretion.

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