

ลักษณะของสารอินทรีย์ละลายน้ำและดัชนีความขุ่นในน้ำทิ้งที่ผ่านการกรองของชุมชนริมทะเลสาบรอบลุ่มน้ำทะเลสาบสงขลา ประเทศไทย

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บทคัดย่อ

ลักษณะของสารอินทรีย์ละลายน้ำ (DOM) ในน้ำทิ้งที่ผ่านการกรองของชุมชนริมทะเลสาบรอบลุ่มน้ำทะเลสาบสงขลา (SLB) สามารถประเมินโดยใช้การวิเคราะห์สารอินทรีย์คาร์บอนละลายน้ำ (DOC) การดูดซับรังสียูวีที่ความยาวคลื่น 254 นาโนเมตร (UV254) และฟลูออเรสเซนซ์เอ็กไซเทชัน-อิมิสชัน เมทริกซ์ (FEEM) สเปกโทรสโกปี โดยดัชนีความขุ่น (HIX) ใช้จำแนกที่มาและขอบเขตของการก่อตัวของฮิวมิกใน DOM ในชุมชนริมทะเลสาบ เช่น ทะเลน้อย ต้นน้ำและกลางน้ำ มีส่วนประกอบ DOM เป็นสารฮิวมิกและกรดฟุลวิกจากตะกอนดิน สารคล้ายทรีปโตเฟนถูกพบในตัวอย่างน้ำทิ้งทั้งหมดของชุมชนริมทะเลสาบ ค่า HIX สูงขึ้นในระยะกักพักยาวนานของทะเลสาบ เช่น ทะเลน้อย ในขณะที่ค่าการดูดกลืนของโปรตีนมีความเข้มข้นในปากน้ำ SLB และ SLB ปลายน้ำ ซึ่งมีเวลากักพักสั้นลง ผลการวิเคราะห์ DOM สามารถสรุปน้ำเสียชุมชนริมทะเลสาบเป็นปัญหาสำคัญของ DOM ที่เกิดขึ้นใน SLB การพัฒนาความเชี่ยวชาญในพื้นที่ SLB นี้จะช่วยยกระดับการกำหนดนโยบายในการควบคุมกิจกรรมของมนุษย์และการบำบัดน้ำเสียชุมชนในพื้นที่ SLB

คำสำคัญ: สารอินทรีย์ละลายน้ำ ฟลูออเรสเซนซ์เอ็กไซเทชัน-อิมิสชัน เมทริกซ์ ดัชนีความขุ่น
ชุมชนริมทะเลสาบ ลุ่มน้ำทะเลสาบสงขลา

Characterization of Dissolved Organic Matter and Humification Index in filtered Effluent Water of Lakeside Communities around Songkhla Lake Basin, Thailand

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Abstract

The characterization of dissolved organic matter (DOM) in the filtered effluent of lakeside communities around the Lake Songkhla Basin (SLB) can be investigated using the analysis of dissolved carbon organic matter (DOC) and UV absorption at a wavelength of 254 nm (UV254) and fluorescence excitation–emission matrix (FEEM) spectroscopy. The humification index (HIX) was used to differentiate the origin and extent of formation of humic substances in DOM. In a lakeside community as Thale Noi, the upstream and midstream DOM contents were humic and fulvic acid–like substances from soil sediment. Tryptophan–like substances showed in all water samples of lakeside communities, HIX was higher in the long residence time of the lagoon as Thale Noi while protein–like fluorescence was stronger in downstream SLB and SLB estuary, where has a shorter residence time. The DOM results could conclude lakeside community wastewater was the major problem of DOM occurring in SLB. Advancing expertise in this SLB area would enhance policymakers to control human activities and community wastewater treatment along SLB.

Keywords: Dissolved organic matter, Fluorescence excitation–emission matrix, Humification index, Lakeside communities, Songkhla lake basin

Introduction

The Songkhla Lake Basin (SLB) has an area of 8,584.35 km² and includes 26 districts,

147 sub–districts/municipalities and 1,247 villages comprising areas of Phatthalung, Songkhla and Nakhon Si–Thammarat Provinces. The

SLB is characterized by three environments as freshwater, brackish water, and brine. The boundaries of these environments change according to the season and the estuarine tides. The SLB has the geological features of a lagoon (Tippayawong and Somboonsuke, 2013). The SLB is an essential water resource for local fisheries, agriculture and transport but is now facing problems of water quality deterioration. Wastewater ingress from industry, farms and lakeside communities has resulted in high levels of pollution. Degraded water quality in streams and lakes can be measured and characterized as organic loading and organic waste, for example, the U–Tapao Canal, which is a branch canal in SLB found doses of the dissolved organic matter (DOM) and humic substances throughout the year (Inthanuchit *et al.*, 2018). The DOM as compound and heterogeneous varieties of aromatic and aliphatic organic compounds with various functional groups and molecular structures (Bastidas *et al.*, 2012; Burpee *et al.*, 2016; Song *et al.*, 2018) was measured in water from lakeside communities around the SLB and reported as dissolved carbon organic matter (DOC) provides the predominant material that absorbs UV, and rapid attenuation occurs with moderate dissolved organic carbon concentrations (Thurman, 1985) and UV absorption at a wavelength of 254 nm (UV254) provides a type of the concentration of organic matter, such as humic substances, are aromatic and occur in high

concentrations in surface water (Gerrity *et al.*, 2012; Weishaar *et al.*, 2003; Wittmer *et al.*, 2015). Then, Fluorescence excitation–emission matrix (FEEM) spectroscopy was applied to determine the origin and composition of the DOM at ordinary concentrations without isolation and chemical reagent preparation before analysis (He *et al.*, 2016; Sgroi *et al.*, 2016; Wu *et al.*, 2011). FEEM spectroscopy quantified the shift of emission spectra towards longer wavelengths with increasing humification. The HIX values are essential factors in controlling the sorption of DOM to mineral surfaces. Amendment with materials that release DOM of molecular weight and humification cause enhanced initial sorption of DOM to soil solids, contributing to increased soil organic matter (Hansen *et al.*, 2016).

The aim of this study was therefore to characterization of dissolved organic matter and humification index in filtered water of lakeside communities around SLB in term of seasonal (dry and rainy season) and business and administrative practices (holiday and working day). DOC and UV–254 analysis could classify the major discharged locations of DOM into SLB. FEEM analysis could encourage to provide a better understanding of the variation of complex compositions of DOM along SLB. The HIX values can differentiate describe the origin and transformation of DOM from various sources in watersheds. The data achieved would be helpful for definite identification of watersheds contamination source and the effective

SLB water quality management.

Research Methodology

Lakeside Communities and sample collection

In this study, the SLB was divided into three large-scale areas according to the characteristics of lagoon physicality (Somboonsuke, 2013; Srichaichana *et al.*, 2019;

Tippayawong and). It also studied physical size of SLB lakeside communities and sewage system. St₁ was at Thale Noi, St₂–St₇ were located in the upstream SLB, St₈–St₁₀ were located in the midstream SLB and St₁₁–St₂₀ were located in the downstream SLB. St₁₂ represented the Hatyai Municipal Wastewater Treatment Plant (Hatyai MWTP) as shown in figure 1.

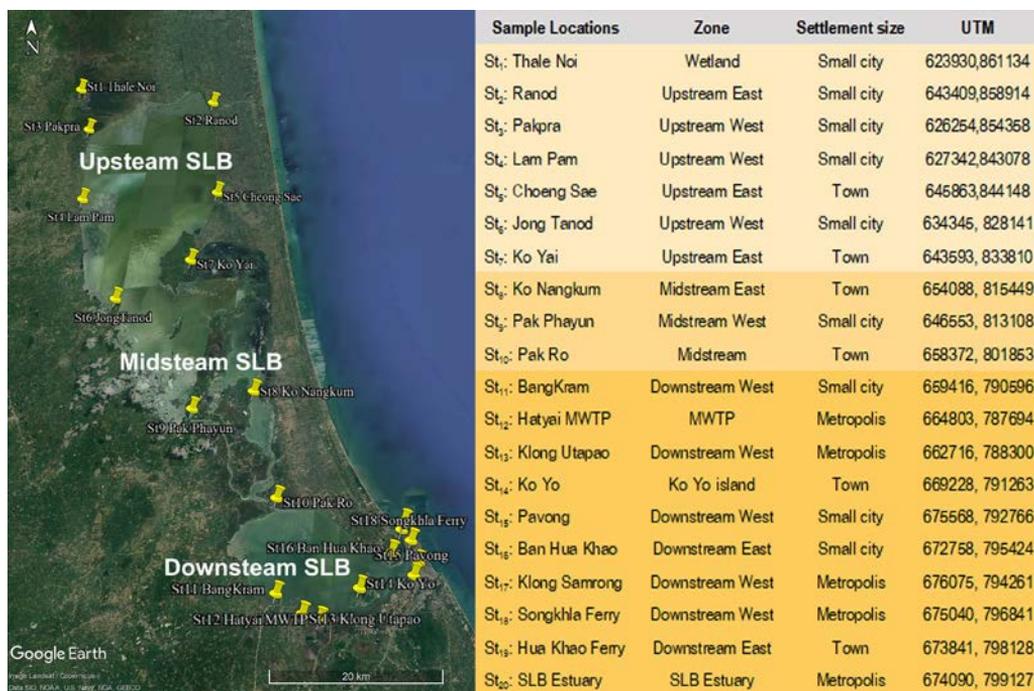


Figure 1 Sample collection point of lakeside communities around the SLB (adapted from Google Earth Pro)

Analytical methods

All sampling along the lakeside communities around the SLB was conducted in term of seasonal in Songkhla like February to May (dry season) and June to January (rainy season) (Climate Center, 2018) and business and administrative procedures (holiday and

working day). Sample waters were stored on 8–9 March and 15–16 November 2018 and 15–16 February and 18–19 October 2019. All samples were collected as surface water (1–2 m depth) by one sampling location per site from 20 sites on the same day to represent outflow of residential, community, industrial, agricultural

catchments, and Channel outlet. The statistics analysis of data was using Pearson's correlation (r) test at $p < 0.01$ to identify the association between pair of variables measure significant differences seasonal and business and administrative procedures along SLB.

The collected water samples were filtered through a Whatman GF/F membrane (pore size 0.7 μm) prior to store at 4°C in the dark, add H_2SO_4 to pH < 2 in accordance with the standard method 1060B and immediately analyzed to establish DOCs were analyzed in accordance with the standard method 5310D using (the Wet-Oxidation Method) UV-254 was analyzed in accordance with the standard method 5910 B using a UV/VIS spectrometer, Jasco V-350 UV/VIS spectrophotometer at 253.7 nm, with matched quartz cells providing a path length of 10 mm (American Public Health Association (APHA), 2017). FEEM spectroscopy was

measured using Jasco FP-6200 and FP-750 Spectrofluorometers with a wavelength range of 220 nm to 600 nm for excitation and emission. FEEM spectra of all water samples were subtracted by the FEEM spectra of Milli-Q water and converted to quinine sulphate units (QSUs). Ten QSUs are equal to the fluorescence spectra of 10 $\mu\text{g/L}$ quinine sulphate solution at 450 nm with an excitation wavelength of 345 nm. FEEM data were discarded when the excitation wavelength (Ex) was greater or equal to the emission wavelength (Em) or $\text{Ex} \times 2$ was less than or equal to Em to eliminate the influence of primary and secondary scattered fluorescence and highlight the targeted peaks. Rayleigh and Raman scattering at peak $\text{Em} \pm 10\text{--}15$ nm of each Ex was also separated from the FEEM spectra. The humification index (HIX) value was calculated from the Equation (1).

$$\text{HIX} = \frac{\sum \text{FL}_{435 \rightarrow 480}}{\sum \text{FL}_{300 \rightarrow 345} + \sum \text{FL}_{435 \rightarrow 480}} \quad \text{--- (1)}$$

where FL represents the fluorescence intensity at each wavelength (Ohno, 2002). The emission spectrum was measured at an excitation wavelength of 254 nm, which suggests humification or degradation of DOM.

Results and Discussion

Variation of DOC and UV254 in water samples from lakeside communities around the SLB

Figure 2 shows average DOC during

the dry season of 2018 to 2019 as 1.7 mg/L from St₁ Thale Noi. Average DOC value increased to 2.5 mg/L at St₆ Jong Tanod (upstream SLB) and was recorded at 3.4 mg/L at St₉ Pak Phayun (midstream SLB), while an average DOC value of 3.7 mg/L was identified in St₁₂ Hatyai MWTP. The downstream location at St₁₇ Klong Samrong had an average DOC value of 4.3 mg/L. In the SLB estuary, an average DOC value of 2.0 mg/L was measured. During the rainy season of 2018 to 2019, the

average DOC value at the upstream area (St₆) was 2.5 mg/L. This declined along with midstream locations from 3.3 mg/L at St₉ to 1.9 mg/L at St₁₁. An average DOC value of 3.7 mg/L was detected at St₁₂ Hatyai MWTP. For the downstream area (St₁₇), an average DOC value of 4.2 mg/L was observed. The SLB estuary (St₂₀) gave an average DOC value of 2.0 mg/L.

The UV254 values of water samples from the SLB during the rainy and dry seasons of 2018 to 2019 were highly variable. Average UV254 during the dry season was 0.063 cm⁻¹ at St₁ Thale Noi, while average UV254 value increased to 0.193 cm⁻¹ at the upstream area (St₆) and also increased to 0.262 cm⁻¹ at the midstream location (St₉), whereas an average UV254 value of 0.263 cm⁻¹ was observed in the downstream area (St₁₇). In the rainy season, average UV254 value of water in Thale Noi was 0.059 cm⁻¹, average UV254 value at an upstream

SLB (St₆) was 0.192 cm⁻¹ and increased along with midstream locations from 0.230 cm⁻¹ at St₉ to 0.297 cm⁻¹ at the downstream SLB (St₁₇). For the SLB estuary (St₂₀), an average UV254 value of 0.212 cm⁻¹ was observed.

The DOM statistics analysis by Pearson's correlation (*r*) test at *p* < 0.01 showed that quantity of DOM in seasonal compare 2018–2019 showed a moderate positive correlation of UV254 (*r* = 0.668), DOC showed a strong positive correlation (*r* = 0.997) and business and administrative procedures along SLB in all seasonal of 2018–2019 was a strong positive correlation (*r* > 0.998). The DOM showed a moderate positive correlation DOC with UV 254 (*r* = 0.694).

Based on land used type as report by Somboonsuke (2013), Srichaichana *et al.* (2019) and Tippayawong, at an upstream SLB, land used type was wetland and agricultural land because it comprises many types of acti-

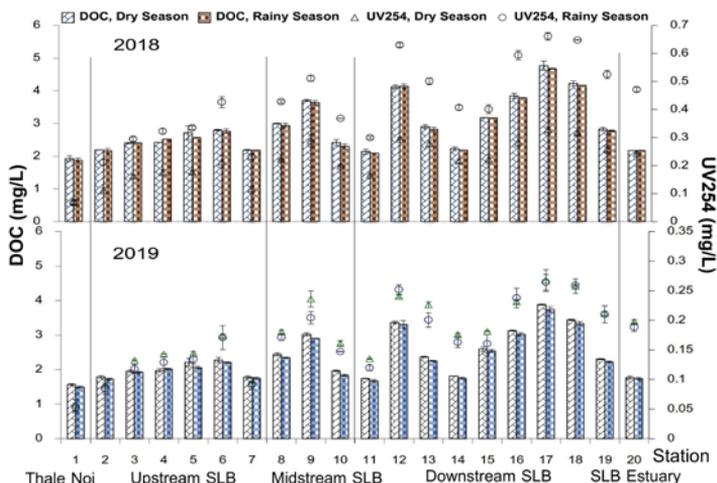


Figure 2 Variation of DOC and UV254 in water samples from lakeside communities around the SLB

vities. The major land used type for midstream SLB was miscellaneous, followed by urban and built-up land. With more detail consideration, it could originate sources of contamination from non-point sources, including agricultural and community activities and point sources, including industrial and community activities. The data could consider the DOM level at midstream SLB like St₉ Pak Phayun relatively to be mentioned. With downstream SLB, land used types were agricultural and urban and built-up land. It could originate sources of contamination from and point sources, including industrial and community activities and non-point sources, including agricultural and community activities. St₁₂ Hatyai MWTP and St₁₇ Klong Samrong are on a downstream SLB. Therefore, point sources and non-points sources contaminants could be generated from this site. The DOC and UV₂₅₄ levels sharply increased in upstream SLB areas after Thale Noi and changed in midstream to downstream SLB areas. the DOM origin was reduced though the distance along the SLB, for example St₆–St₇, St₉–St₁₁ and St₁₇–St₂₀, because watershed restoration occurs, based on land used of each station related the low rate of organic load from community activities after DOM origin into SLB ecosystems.

Characteristics of FEEM spectroscopy in water sampling of the SLB

Figure 3 presents three results of

FEEM spectroscopy from lakeside communities around the SLB during the rainy and dry seasons of 2018 to 2019 at contour intervals of 10 QSU. The fluorescent excitation–emission wavelengths (Ex/Em) demonstrated major fluorescent emission intensities, divided as fluorescence peaks, as represented in Figure 3. Peak positions A, B, C and D of FEEM spectroscopy showed conditions of natural organic matter and organic matter from non-point source pollution in the water samples. Chen *et al.* (2003) and He *et al.* (2013) characterized three major fluorescence peaks in water samples as (i) humic/fulvic-like organic matter (Ex/Em = 330–350/420–480 nm), (ii) humic-like organic matter peak (Ex/Em = 250–260/420–480 nm) and (iii) protein-like organic matter (Ex/Em = 250–260/250–280 nm). Results of FEEM spectroscopy showed fulvic acid-like substances at 280 nm_{Ex}/410 nm_{Em} (C) and 340 nm_{Ex}/410 nm_{Em} (D) with tryptophan-like substances at 240 nm_{Ex}/360 nm_{Em} (A) and 260 nm_{Ex}/360 nm_{Em} (B) also recognized. Peaks in the FEEM spectroscopy investigation were used to produce a report on the putative origin of fluorescent organic matter. Musikavong *et al.* (2013) classified the putative fluorescent organic matter into three subgroups as tyrosine-like, tryptophan-like and humic and fulvic acid-like substances. This research complemented the studies of Musikavong *et al.* (2013) who showed that fluorescence peaks of industrial estate wastewater and treated wastewater had six major peaks

as tyrosine-like substances, tryptophan-like substances and humic and fulvic acid-like substances. For tryptophan-like substances, all peaks were close to the previous study; the point and non-point pollutions could be sus-

picious sources that discharge DOM from the lakeside communities to the SLB. The putative FEEM spectroscopy peaks A and B are tryptophan-like substances, while peaks C and D are humic and fulvic acid-like substances.

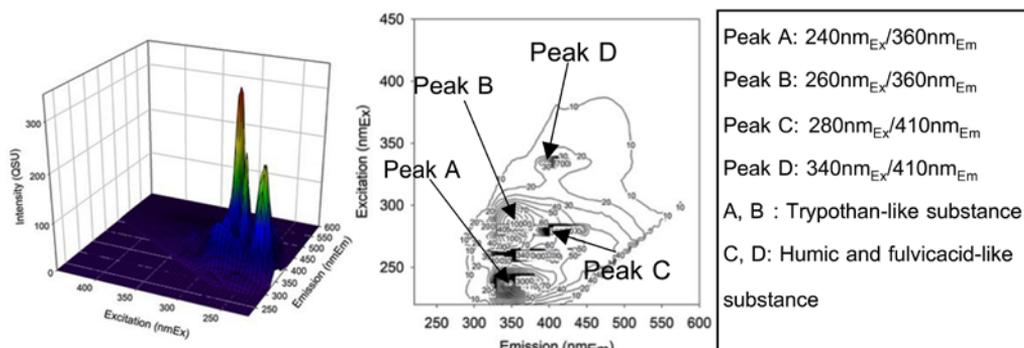


Figure 3 Peak positions A, B, C and D of FEEM spectroscopy water sample results from SLB

Thale Noi (St_1) during the rainy and dry seasons of 2018 to 2019 showed peak D with average fluorescence intensity of 36 and 27 QSU and peak C with average fluorescence intensity of 42 and 33 QSU. Humic and fulvic acid-like substances, such as humic acid and fulvic acid are predominantly present in surface water, protein-like substances (Barker and Stuckey, 1999) could result from the initial DOM in water from Thale Noi in the dry and rainy seasons. Peaks C and D in this study were the same as determined by Musikavong *et al.* (2013) and expected as the fluorescence peak positions in reservoir waters in Thailand, this research proposed that humic and fulvic acid-like substances could result from the initial DOM in wetlands upstream of the SLB. Peaks C and D were stable after complexing produced

by humic-like substances (Zhang *et al.*, 2016).

Peaks A, B, C and D from lakeside communities around the SLB are illustrated in Figures 4 and 5. Fluorescence peak position intensities of water samples collected on holidays and working days were similar. Peaks A, B, C and D of upstream SLB (St_6) in the dry and rainy seasons of 2018–2019 showed average fluorescence intensities of 207, 236, 322 and 45 QSU. Peaks A, B, C and D in the dry and rainy seasons of 2018 to 2019 decreased at St_7 with average fluorescence intensities of 130, 163, 265 and 34 QSU. Fluorescence intensities of all peaks increased when compared with Thale Noi; therefore, tryptophan-like substances and humic and fulvic acid-like substances at all peaks were generated from discharged water into the SLB, like St_2 – St_6 in the

upstream SLB area.

For midstream SLB, the tyrosine-like and tryptophan-like substances as residues primarily contributed to protein-like fluorescence in wastewater (Mohapatra *et al.*, 2021) were defined as indicators for measuring stream water quality (Zhao *et al.*, 2016). The fluorescence

intensity of tryptophan-like substances at peaks A and B were high at Pak Phayun (St₈) and fluorescence intensity of humic and fulvic acid-like substances at peaks C and D increased. However, the process was incapable of reducing the tryptophan-like substances at Bang-Kram (St₁₁).

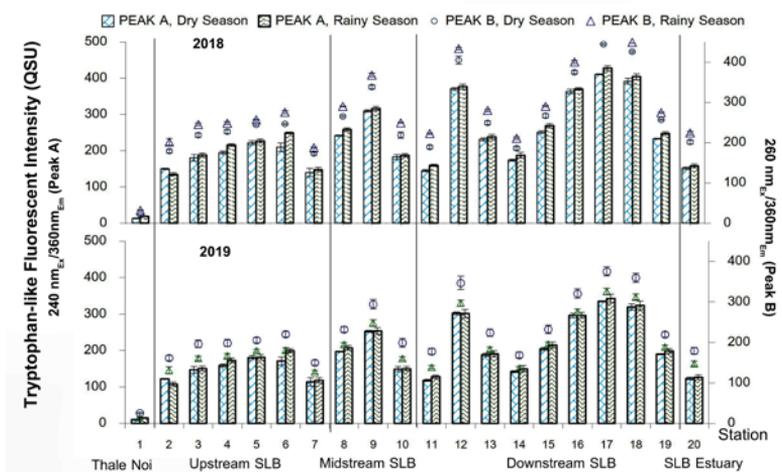


Figure 4 Variation in tryptophan-like fluorescence intensity at peaks A and B from lake-side communities around the SLB

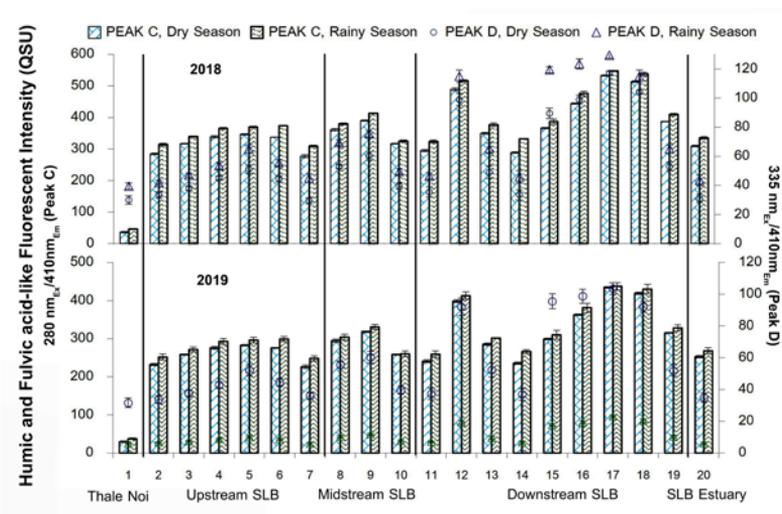


Figure 5 Variation in humic and fulvic acid-like fluorescent intensity at peaks C and D from lakeside communities round the SLB

At downstream SLB, the fluorescence intensity of tryptophan-like substances at peaks A and B and fluorescence intensity of humic and fulvic acid-like substances at peaks C and D were higher than at midstream SLB. Hatyai MWTP (St₁₂) was situated downstream SLB. An early investigation by Musikavong *et al.*, (2013) stated that dry and rainy seasons had humic and fulvic acid-like substances in water from Klong Utapao (St₁₃). This research found that all peaks were highest at Klong Samrong (St₁₇); however, the process was incapable of reducing the tryptophan-like substances at the SLB estuary. Thus, downstream SLB experienced increased DOM during the dry and rainy seasons since the tryptophan discharged into downstream SLB areas. From the FEEM spectroscopy results, the origins of DOM in water from Hatyai MWTP (St₁₂), Klong Utapao (St₁₃), Klong Samrong (St₁₇) and Songkhla ferry (St₁₈) were tryptophan-like and humic and fulvic acids-like substances.

The FEEM spectroscopy results indicated that water from upstream and midstream SLB emanated from community wastewater, industrial wastewater, and farm pollution discharge, while tryptophan-like substances were the dominant DOM from community wastewater discharged into the downstream SLB compared to humic and fulvic acid-like substances. This research could be concluded lakeside community wastewater was the main problem of DOM occurring in SLB, as shown

in St₆, St₉, St₁₂ and St₁₆–St₁₈, where were a large lakeside community along SLB.

The FEEM statistics analysis by Pearson's correlation (r) test at $p < 0.01$ showed that quantity of FEEM in seasonal compare 2018–2019 showed a strong positive correlation of tryptophan-like substances ($r = 0.999$), humic and fulvic acids-like substances showed a strong positive correlation ($r = 0.823$) and business and administrative procedures along SLB in all seasonal of 2018–2019 was a strong positive correlation ($r > 0.993$). The FEEM showed a strong positive correlation tryptophan-like substances with humic and fulvic acids-like substances ($r = 0.881$).

HIX values of water samples from lakeside communities around the SLB

The HIX values of DOM in surface lake water samples are shown in Figure 6. The HIX values of SLB showed seasonal variation. Ohno (2002) presented high HIX for soil fulvic acid. In Thale Noi, upstream and midstream SLB the waters have a long residence time and labile organic matters degrade within a few months. Origin, and chemical composition of Thale Noi, upstream and midstream SLB are controlled by autochthonous sources and dynamics of the SLB. The HIX of downstream SLB and the SLB estuary DOM are distributed since water mixing were lower than in upstream and midstream areas. The major contributor to downstream SLB DOM composition might be allochthonous sources such as elution

from Hatyai MWTP and wastewater discharge around the downstream SLB.

Comparison of HIX values in lakeside communities around the SLB showed that DOM at Thale Noi had the highest HIX value in each watershed that could have originated from soil fulvic acids (Ohno, 2002). DOM of the downstream SLB showed the lowest HIX value in this research. HIX were higher in the long residence time of the lagoon as Thale Noi (St₁) while protein-like fluorescence was stronger in downstream SLB and SLB estuary, where has a shorter residence time. According to Ohno (2002), DOM extracted from corn residue has a low value and is a primary SLB source of

non-humified, water-soluble material in soils. It has the effectiveness of a tryptophan/tyrosine-like substance like contributions from sewage and farm wastes. Downstream SLB like Hatyai MWTP (St₁₂) and Klong Utapao (St₁₃) has many streams along with paddy fields and households. Therefore, the DOM might comprise less humified, fresh and proteinous organic materials.

The HIX statistics analysis by Pearson's correlation (*r*) test at *p* < 0.01 in seasonal and business and administrative procedures along SLB in all seasonal of 2018–2019 was a strong positive correlation (*r* > 0.991).

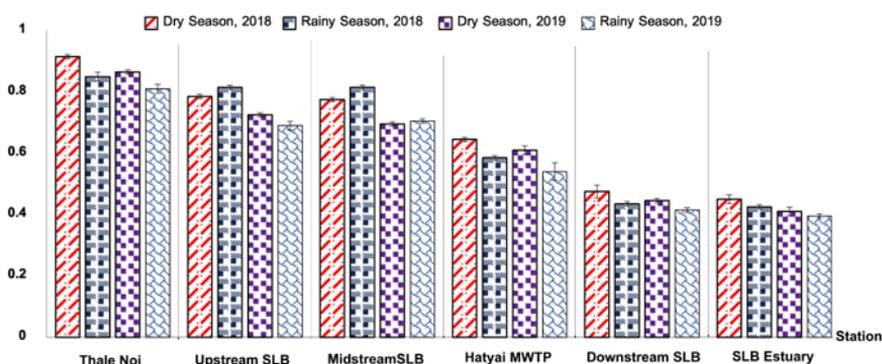


Figure 6 Seasonal variation in HIX values of DOM from lakeside communities around the SLB during the rainy and dry seasons of 2018 to 2019

Conclusions

The DOM levels sharply increased in upstream SLB areas after Thale Noi and changed in midstream to downstream SLB areas. The DOM was reduced where watershed restoration occurs, based on land used in each station related to the low rate of organic

load from community activities after DOM origin into SLB ecosystems. FEEM spectroscopy discovered humic and fulvic acid-like substances at peaks C and D and tryptophan-like substances at peaks A and B in all water samples along the SLB. HIX was higher in the long residence time of the lagoon as Thale Noi

while protein-like fluorescence was stronger in downstream SLB and SLB estuary, where has a shorter residence time. The DOM results could conclude lakeside community wastewater was the major problem of DOM occurring in SLB. Advancing expertise in this SLB area would enhance policymakers to control human activities and community wastewater treatment, which will not affect the environment and ecosystem.

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