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SPATIO-TEMPORAL DISTRIBUTION OF DISSOLVED ORGANIC MATTER IN A TROPICAL FRESH WATER LAKE: INDIA

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INTRODUCTION

Dissolved organic carbon (DOC) is the second largest pool of carbon in the ocean, having approximately the same magnitude (approx. 685Gt C) as the pool of atmospheric CO₂ (Hansell and Carlson, 1998). With regard to absorption and exchange of CO₂ between ocean and atmosphere, understanding the dynamics of the dissolved organic carbon pool is essential for modeling the global carbon cycle.DOC is gradually recognized as a critical property of aquatic ecosystems. DOC influences a wide variety of physical, chemical, and biological processes. Light absorption by DOC has a strong influence on the penetration of ultraviolet and photosynthetically active radiation (Morris et al., 1995) as well as the mixing and thermal properties of lakes (Fee et al. 1996; Snucins and Gunn 2000). DOC interacts with dissolved nutrients and metals and thereby influences their concentrations and availability (Perdue 1998; Shaw et al., 2000). DOC mainly comprises acids and, therefore, affects pH and in some acidic waters acts as a buffer (Driscoll et al. 1994). DOC also represents a large reservoir of carbon in many systems, including the ocean (Benner et al. 1992). Small changes in DOC concentrations can be of considerable significance for carbon cycling and ecosystem metabolism because the pool is solarge (e.g., Cole et al., 2000). Globally, the concentration of DOC varies from, 100 to 4,000 µmol L in freshwaters (Perdue and Gjessing, 1990).

Kolleru lake, the largest freshwater lake in India was studied for the assessment of dissolved organic matter distribution and the influence on the lake hydro chemical characteristics. The study revealed the significant influence of freshwater inputs and their control in the distribution of dissolved organic matter (DOM) throughout the lake. Correlation analysis indicated that the DOM mineralization decreased the DO concentration during the monsoon and post-monsoon, making the lake under saturated (upto 7% DO saturation) whereas, during pre-monsoon DO in the lake was not controlled by DOM. Mixing plots of DOC, DON and DOP versus salinity suggested that DON and DOP acted as sink during all the seasons, whereas DOC acted as a source due to domination of production over mineralization. This is the baseline information regarding the dissolved organic matter and will be useful for further intensive study on biogeochemistry of the lake.

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Although DOC concentrations can vary dynamically in small lakes with short water residence times and high allochthonous loading (e.g., Hessen et al., 1997), DOC in systems with larger, longer residence time can appear to have low seasonal and inter-annual variance. This low variance reflects the slow dynamics of a large and relatively recalcitrant pool of organic matter (Wetzel, 1995). Despite the apparent slow dynamics and low variability, some observations reveal that DOC can change substantially in lakes. With regard to absorption and exchange of CO₂ between ocean and atmosphere, understanding the dynamics of the dissolved organic carbon pool is essential for modeling the global carbon cycle. Several studies have been carried out to investigate the distribution and biogeochemical cycling of DOM, focusing on DOC (Hama and Handa, 1983; Fukushima et al., 1996; Parks and Barker 1997).Several studies have observed conservative behavior of DOC in estuaries (e.g. Alvarez-Salgado and Miller, 1998; Abril et al., 2002). Some studies observed a significant degradation of DOC supplied by tributary rivers within the estuary (Raymond and Bauer, 2001). In addition, some studies identified the important internal sources of DOC within the estuary. But a very few studies are reported in the tropical freshwater lakes especially in Kolleru lake. In the present study we mainly focused on the biogeochemical processes of organic matter through spatial and temporal variation of dissolved organic matter.

ABSTRACT

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MATERIALS AND METHODS

Study Area

Kolleru Lake is the largest freshwater lake in India, situated between $16^{\circ}30' - 16^{\circ}45'$ latitude and $81^{\circ}05' - 81^{\circ}20'$ longitude, along the east coast of India (245 km² as in topographic maps of 1930s). Although the lake is about 35 km inland from the present coast, it was a coastal lagoon in the geological past, believed to have been formed around 6000 years BP. Kolleru lake was designated as a RAMSAR site, considering that the lake functions as a flood-balancing reservoir between the Krishna and Godavari deltas and that it supports vulnerable species like grey pelican as well as water fowl, including a variety of resident and migratory birds. As a number of rivulets such as Tammileru, Budimeru and several other smaller ones draining a total catchment of about 5400 km² are decanting their waters into it. Kolleru still maintains its connection with the Bay of Bengal through a 60 km long, intricately meandering tidal channel at Upputeru, a typical characteristic of coastal lagoons (Figure 1). About 42% of the 245 km² lake area was encroached for aquaculture and 8.5% more area was occupied for agriculture, while the rest of the lake is either beingdried out by reclamation or is infested with weed (Nageswara Rao et al., 2004). It experiences brackish water conditions in the south eastern region during summer and the salinity reaches upto mid of the lake, when the inflow of the fresh water into the lake is low. The saline water reaches up to the middle regions of the lake, while the northern most parts remains freshwater. Lake was predominantly covered with submerged (Otteliaa lismoides, Vallisneria spiralis, Ceratophyllum) and floating (Ipomea aquatic and Eichhornia crassipes) macrophytes. Nymphaea nouchali, Nymphaea stellata, Nymphoides hydrophylla and Salvinia cucullata are moderately distributed in many parts of the lake.

Sampling and Analytical methodology

Sub surface water samples were collected in three seasons (May, August and November) of 2010 at the stations indicated in Table1 to understand the biogeochemical cycling of organic matter in the lake. Altogether 10 hydrographically differing GPS fixed locations(Table1), representing the Kolleru lake were chosen for sampling detailed analysis of biogeochemical variables. At every time sampling was completed within the 2days fieldwork, a mechanized boat was used to reach the individual sites. A GPS (Garmin III, USA) was employed during navigation.

Sample collection and preservation

The water samples were collected using 5L Niskin sampler.2L of water was collected for organic nutrients such as Dissolved Organic Nitrogen (DON) Dissolved Organic Phosphorous (DOP), Chlorophyll. Samples were preserved at 4 °C, brought to the laboratory at the earliest. For dissolved oxygen (DO) samples were collected in a 250ml air tight glass bottle and fixed with Winkler's reagents. For dissolved organic carbon (DOC) the water sample was collected in 100ml pre-cleaned borosilicate bottles and stored at 4 °C in dark. Following the collection, a known volume of sample was filtered in duplicate within the 5hrs of the sampling through Whatman GF/F glass

fiber filters(nominal size cutoff : 0.7 μ m pore size; precombusted at 450 °C for 4h) using pre-cleaned filtration flask. Pre-cleaning of glassware performed by acid washing (10% HCL and copious rinsing with Milli-Q water), followed by combustion at 450°C for 1h. The filtrate obtained was collected and stored in pre-combusted, 25ml glass vials, acidified with 10% phosphoric acid, sealed with teflon-lined caps and frozen until the DOC analysis was performed.

Physical parameters

Temperature was measured using a portable conductivity, temperature probe (Hydrolab). Salinity was measured by the argentometric method (Knudsen scale), pH was measured using a Metrohm pH meter. The pH meter was calibrated on the NBS scale as described by Frankignoulle and Borges (2001). Analytical precision of pH for freshwater samples was ± 0.005 and for the rest ± 0.003 .

Dissolved oxygen, dissolved organic nutrients and chlorophyll analysis

Dissolved oxygen in the samples was fixed and analyzed using modified the methodology of Winkler (1888) iodometric titration by Carrit and Carpenter (1966). The analytical precision, expressed as standard deviation, was ±0.07% for DO.A dissolved organic nutrient (Dissolved Organic Nitrogen, Dissolved Organic Phosphorous) was performed on the samples filtered through 0.22 µ Millipore membrane filters and was analyzed by wet per sulfate oxidation method (Grasshoff et al., 1999) with a relative error of about 3%.A known volume of water was filtered through GF/F paper (Whatman) for the analysis of chlorophyll. Chlorophyll on the filters was first extracted with 90% acetone, at 4 °C in dark for 24 h, and then the pigment concentration was measured using a Shimadzu spectrophotometer (Model - UV 1700, Shimadzu)at different wave lengths 750, 664, 647, and 630 nm. Each extinction was corrected for a small turbidity blank by subtracting the 750 nm from the 664, 647 and 630 nm.(Parsons et al., 1984). The analytical precision for Chlorophyll-a (Chl-a) analysis was ±4%.All these analyses were completed within 12 h of sampling at the shore-based laboratory set-up at INS-Chilika, Orissa.

Dissolved organic carbon analysis

Samples were collected in the 25ml vials and used for the determination of Dissolved organic carbon (DOC) analysis at ICMAM laboratory at Chennai, within 15 days. DOC samples were analyzed by means of high temperature catalytic oxidation using a Shimadzu TOC-VCPH analyzer equipped with a platinum catalyst on quartz wool and IR detector. High temperature catalytic oxidation is generally considered to be the most appropriate technique for determining DOC concentrations in surface waters (Spyres et al., 2000). Before analysis, the TOC-VCPH was conditioned by repeated injections with Milli-Q water until the blank was low and stable. The system blank was determined by injecting condensed pyrolysed water in the furnace. This system blank was usually below 0.1 mg C 1^{-1} and was therefore not subtracted from the DOC measurements. Before injection into the furnace, the acidified sample/blank was decarbonated with

carbon-free oxygen at a flow of 150 ml min⁻¹ for 4 min. Therefore, our DOC measurements do not include volatile DOC. Three to five replicate 50 μ l subsamples were injected until the coefficient of variation for these replicate injections was below 2%. Calibration was carried out using potassium hydrogen phthalate dissolved in Milli-Q water. A 5-point calibration series in the range of 0 to 20 mg C l⁻¹ was made during each run. The regression line for this calibration always had an r²>0.99. No significant differences were observed for slopes of calibration curves based on Milli-Q or artificial seawater. The accuracy of DOC measurements was checked once in every ten samples with Certified Reference Material (CRM) (from Hansell Laboratory, University of Miami). The analytical precision of the analysis was usually <2% with CRM over time.

RESULTS AND DISCUSSIONS

Hydrographical Characteristics

The mean surface water temperature in the lake was 28.89°C, 31.32°C and 29.33°C during pre-monsoon, monsoon and postmonsoon respectively. Lowest temperature was recorded in the northern part and highest in the south eastern part of the lake during the entire study period. Salinity varied significantly between pre-monsoon and monsoon, whereas it was not found between monsoon and post-monsoon. Salinity varied between 1.6-34.4 during pre-monsoon whereas, it was restricted to 0.2 for the rest of the period (Figure 2c), indicated that the system was dominated with fresh water except in premonsoon. pH was found to be lowest(7.44) in the monsoon and highest was observed in pre-monsoon(8.34). A significant positive correlation was found between pH and salinity during pre-monsoon indicates that the seawater intrusion increases the pH (Figure 2b). Turbidity was high during pre-monsoon at the high saline region due to the high wind, shallowness and the tide induced churning of the water column leads to the resuspension of bottom sediments (Figure 2a). Similar phenomenon was also observed in Chilika, a shallow brackish water lagoon (Muduli et al., 2012).

Cluster analysis and multi-dimensional scale (MDS) analysis was performed to understand the similarities among the stations. Cluster analysis has been used as a very useful tool for discriminating similar stations according to different hydro chemical characteristics in coastal waters (Muduli et al., 2011). During the study period, discriminant behavior of different stations can be observed from the cluster and MDS analysis. Stations 1 to 6 are fresh water dominated, whereas the stations from 7 to 10 are towards the sea and higher saline. Stations 1 to 6 are vulnerable to domestic sewage and also river discharges which makes this part of the lake more polluted as compared to the part connected to the sea. During all the seasons, stations maintained the discriminant characteristics (Figure 3). One-way analysis of variance (ANOVA) was performed to observe if any significant variation in the results of different seasons. ANOVA showed except water temperature and turbidity all parameters have

Table 1. Latitude and longitude of the sampling stations in Kolleru Lake

St.No	Latitude N	Longitude E
1	16°37'44.5"	81°12'34.3"
2	16°37'00.6"	81°14'31.8"
3	16°36'57.5"	81°16'31.8"
4	16°36'49.5"	81°18'33.1"
5	16°36'29.7"	81°20'30.3"
6	16°34'36.9"	81°20'43.9"
7	16°31'53.3"	81°20'34.2"
8	16°30'21.7"	81°21'18.1"
9	16°26'00.1"	81°23'55.8"
10	16°23'51.5"	81°27'57.7"

Table 2. Analysis of variance (ANOVA) of studied parameters in different seasons

_		Sum of Squares	Mean Square	F	Sig.
WT	Between Seasons	29.03	14.52	2.81	0.08
	Within Seasons	113.76	5.17		
	Total	142.79			
Turb	Between Seasons	820.70	410.35	2.44	0.11
	Within Seasons	3700.46	168.20		
	Total	4521.16			
pН	Between Seasons	0.78	0.39	22.98	0.00
	Within Seasons	0.37	0.02		
	Total	1.15			
Salinit	yBetween Seasons	1290.66	645.33	12.70	0.00
	Within Seasons	1117.59	50.80		
	Total	2408.24			
DO	Between Seasons	207.25	103.62	69.70	0.00
	Within Seasons	32.71	1.49		
	Total	239.95			
DO%	Between Seasons	44706.26	22353.13	62.24	0.00
	Within Seasons	7901.59	359.16		
	Total	52607.85			
Chl-a	Between Seasons	3699.68	1849.84	48.04	0.00
	Within Seasons	847.06	38.50		
	Total	4546.74			
DOC	Between Seasons	460015.19	230007.59	10.59	0.00
	Within Seasons	477673.03	21712.41		
	Total	937688.22			
DON	Between Seasons	18543.16	9271.58	30.34	0.00
	Within Seasons	6722.87	305.59		
	Total	25266.03			
DOP	Between Seasons	175.56	87.78	69.48	0.00
	Within Seasons	27.79	1.26		
	Total	203.36			

p < 0.05, indicates all parameters varied significantly with respect to different seasons (Table 2).

Dissolved oxygen and Chlorophyll

Maximum oxygen saturation was observed during premonsoon 154.48% and it was decreased to minimum of 7.15% during monsoon. The average DO saturation during premonsoon was ~5 and 3 times higher than the monsoon and post-monsoon respectively (Figure 2d), associated with peak rainfall events in monsoon. DO saturation fell from over saturation to under saturation suggesting either a shift in biological regime or enhanced community respiration. Irrespective of the season Chl-a concentration seems to be high in the south eastern part of the lake. The average Chl-a concentration was low (< 4 mg m⁻³) during post-monsoon and

	Pre-monsoon	Monsoon	Post-Monsoon
Turb	3.22-56.4 (17.24±19.73)	1.01-46.7 (7.66±13.92)	0.65-3.65 (2.16±1.24)
pН	7.93-8.34 (8.13±0.17)	7.44-7.95 (7.69±0.16)	7.64-7.81 (7.75±0.06)
Salinity	1.67-34.46 (16.98±14.96)	0.04-0.07 (0.05±0.01)	0.23-0.34 (0.27±0.04)
WT	24.4-33.2 (28.89±4.7)	31-32 (31.32±0.42)	28.9-30 (29.33±0.51)
DO	5.98-10.98 (8.99±1.7)	0.54-4.14 (1.79±1.1)	1.63-4.81 (3±0.97)
DO%	72.17-154.48 (129.14±30.13)	7.15-56.58 24.21±15.08)	21.18-63.74 39.21±12.85)
Chlorophyll-a	22.95-44.95 (32.55±7.87)	16.74-35.51 (26.58±7.47)	1.07-7.9 (3.94±2.13)
DOC	232.84-950.84 (668.75±298.23)	664.92-824.59 (740.49±55.73)	406.84-486.17 (436.98±25.09)
DON	47.32-74.99 (62.09±10.43)	84.97-103.38 (90.71±6.46)	3.09-76.92 (28.15±26.94)
DOP	0.56-3.62 (1.28±1.17)	3.84-8.76 (6.53±1.35)	0.22-2.28 (1.02±0.77)

Table 3. Descriptive analysis of hydrochemical parameters during the pre-monsoon, monsoon and post-monsoon of 2010, parenthesis indicates the average and standard deviation

Table 4a. Correlation of organic matter and hydrochemical parameters during pre-monsoon 2010

	WT	Turb	pН	Salinity	DO	DO%	Chl-a	DOC	DON	DOP
WT	1.00									
Turb	0.59	1.00								
pН	0.93	0.76	1.00							
Salinity	0.98	0.70	0.94	1.00						
DO	0.02	-0.01	-0.21	0.09	1.00					
DO%	0.70	0.45	0.50	0.75	0.73	1.00				
Chl-a	-0.03	-0.43	-0.12	-0.18	-0.48	-0.43	1.00			
DOC	-0.94	-0.82	-0.95	-0.98	-0.08	-0.72	0.27	1.00		
DON	-0.86	-0.84	-0.94	-0.87	0.22	-0.44	-0.02	0.91	1.00	
DOP	-0.44	-0.29	-0.47	-0.48	0.17	-0.21	0.23	0.46	0.33	1.00

Table 4b. Correlation of organic matter and hydrochemical parameters considering monsoon & post-monsoon data set of 2010

	WT	Turb	pН	Salinity	DO	DO%	Chl-a	DOC	DON	DOP
WT	1.00									
Turb	0.17	1.00								
pН	-0.13	-0.39	1.00							
Salinity	-0.86	-0.27	0.30	1.00						
DO	-0.30	-0.24	0.17	0.62	1.00					
DO%	-0.26	-0.23	0.16	0.59	1.00	1.00				
Chl-a	0.87	0.46	-0.33	-0.89	-0.39	-0.36	1.00			
DOC	0.91	0.29	-0.26	-0.94	-0.74	-0.42	0.96	1.00		
DON	0.83	0.30	-0.20	-0.82	-0.36	-0.33	0.79	0.84	1.00	
DOP	0.84	0.33	-0.19	-0.92	-0.50	-0.48	0.89	0.91	0.85	1.00



Figure 1. Map showing the location of Kolleru lake



Figure 2. Station wise variation of studied parameters during pre-monsoon, monsoon and post-monsoon

increased to maximum 32.55 mgm^{-3} during pre-monsoon (Figure2a). The mean Chl-a concentrations of >8 mgm⁻³was observed during both pre-monsoon and monsoon (Table 3, Figure 2e). The present data, thus, suggest that the peak Chl-a in Kolleru lake occurs annually in the pre-monsoon. A significant positive correlation was found between Chl-a and water temperature during monsoon and post-monsoon indicating the importance of water temperature on the occurrence of Chl-a.

Spatio-temporal variations of DOM

During monsoon, average DOC concentration was found to be maximum (740.48 \pm 55.72 μ M, range: 664.92-824.58 μ M) followed by pre-monsoon (668.75 \pm 298.22 μ M, 232.83-950.83 μ M) and post monsoon (436.97 \pm 25.09 μ M, 406.83-486.17 μ M). The highest DOC concentrations during monsoon

could be attributed by DOC carried by the riverine discharge during the heavy rain fall(August). There was a significant variation in DOC concentration throughout the lake. The DOC concentrations were very high as compared to lagoons and other estuarine environments in India, as reported by in earlier studies viz. Godavari (100 - 300 µM; Sarinet al., 2002; Bouillon et al., 2003), Ganges and Brahmaputra (260 - 380 µM; Spitzy and Leenheer, 1991), and Chilika (<500µM; our unpublished data). Even though the average DOC concentration during the monsoon was high, the DOC concentration in stations 3, 4 and 5 during pre-monsoon were higher which could be due to higher primary production (Figure 2f).During Premonsoon there was no significant correlation between DO and DOC where as a significant correlation of -0.737 was observed considering the monsoon and post monsoon together, indicating the consumption of DO for remineralisation of organic matter.



Figure 3. Cluster analysis and Multi dimensionalscale(MDS) for the all relevant parameters studied in Koleru lake

DON concentrations showed a significant variation with respect to seasons (Table 2) and the concentration varied from 47.32 to 74.9, 84.9 to 103.3 and 3 to 76.9 µM during Premonsoon, monsoon and post monsoon respectively. The seasonal mean was highest during monsoon (90.71 \pm 6.46) could be attributed by riverine discharge, whereas lowest during post monsoon (28.15± 26.94). The variation of DON can be observed from figure 2g. The mean concentrations of dissolved organic phosphorous followed similar trend as DON, as the highest concentration was observed during monsoon and the concentration varied from 3.84 to 8.76 with an average of 6.53 ± 1.35 (Table 3). The variation of DOP can be observed from figure 2h. The main sources of DOM in the lake appear to be primary production within the lake and loading from the catchment. The quantitative importance of the two sources cannot be estimated precisely because of unavailability of total primary production data for the system and in particular how much of the primary production that ends up as DOM in the water column. Macrophytes like Zostera marina (L.), different macro algae species and in particular benthic microalgae are important primary producers (Sand-Jensen and Borum, 1991). All plants are probably

but in particular macro algae are known to produce large amounts (Hulatt et al., 2009). Thus, even in the lakes with small connection to the sea, autochthonous processes are the dominating processes to identify the sink source strengths of DOM and are therefore expected to govern its characteristics, except in close proximity to stream outlets. This has important implication for under standing the results from conservative mixing. Conservative mixing assumes theoretically that two end-members with constant concentrations are mixed and insitu processes within the lake are insignificant, except that an initial change between the freshwater runoff and the inner end-member can be estimated as in Figure 4.Significant deviations from a conservative mixing line or the absence of a relationship means that either end-member concentrations vary to an extent which controls the temporary variability due to production/remineralization processes dominates over mixing process. Alternatively, the absence of a relationship can occur if the uncertainty on measured concentrations is large. Mixing plot of DOC, DON and DOP versus salinity suggested during all the season DON and DOP sinks in the system, whereas DOC acts as a source during pre-monsoonas



Figure 4. Conservative mixing plot of a) DOC, b) DON and c) DOP versus salinity in Kolleru lake

the macro algal production dominates the mineralization (Figure 4a,b, c). Irrespective of the season, salinity showed a strong negative correlation with DOC, DON and DOP which indicates that the enormous DOM loadings during the fresh water discharge into the lake throughout the study period. During pre-monsoon, the dissolved oxygen concentration did not show any correlation with dissolved organic matter (Table 4a) indicated DO was not controlled by mineralization of organic matter, whereas a negatively significant correlation during monsoon and post-monsoon (Table 4b) indicated the mineralization of DOC, DON and DOP occurred using DO leading to anoxic condition in the lake. The expected mineralization could be due to fresh water influence, as evident from the positive correlation of salinity and dissolved oxygen. Hence from the correlation of DOM with salinity and DO, it can be expected that during low saline condition the organic matter gets decomposed or remineralized by oxygen consumption, making the lake highly undersaturated which might turn the system in to heterotrophic (Sarma *et al.*, 2009).

Conclusions

We found that the DOMconcentration in the lake was controlled by freshwater loadings and primary production. Highest DOM concentrations were found during the monsoon season. The end water mixing line model showed that among the DOM components DOC seemed to be produced during pre-monsoon, whereas irrespective of the season DON and DOP seemed to be mineralized in the lake. Correlation analysis revealed that the freshwater plays a major role in organic matter mineralization leads to decrease in oxygen concentration in the lake and made the system highly under saturated. The cycling of DOM in the fresh water bodies like Kolleru lake need to be studied further, as the under saturation observed in the lake might be the indication of high CO_2 flux to the atmosphere.

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