2018-06-08

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Research Paper

Distribution of organic carbon: possible causes and impacts in the Pangani River Basin ecosystem, Tanzania

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Environmental context. Understanding the sources of organic carbon and its spatial and seasonal variation is essential for implementing measures to control water pollution. There is, however, only limited information about organic carbon in east African rivers. This study reports the distribution of dissolved and particulate organic carbon in the Pangani River Basin, using isotopes to trace sources of carbon to the basin and its flux to the Indian Ocean.

Abstract. There is limited information on organic carbon in African rivers, especially from the eastern side. Here, we report distribution and impacts of total suspended matter (TSM), and dissolved and particulate organic carbon (DOC & POC) in the Pangani River Basin (PRB) ecosystem together with their fluxes to the Indian Ocean. $\delta^{13}C$ was also used to trace sources of carbon in the basin. Results showed that the basin is supplied with carbon from allochthonous sources dominated by C_3 plants, with higher levels of TSM and DOC in the wet season than in the dry season. Several factors, including altitude, temperature, rainfall, lithology and anthropogenic activities, have a significant influence on the seasonal and spatial distribution of organic carbon in the basin. High discharge in the wet season mobilised terrestrial organic carbon to elevate concentrations of DOC, POC and TSM. Mean concentrations of DOC, dissolved inorganic carbon (DIC), POC and TSM in PRB were in ranges comparable to that in other tropical rivers but their fluxes were lower than in most tropical rivers around the world. Diverting water from the river for irrigation and hydroelectric power production was one of the factors that reduced the flux of carbon. Observed hypoxic conditions in the reservoir indicates that the quality of water for human and aquatic ecosystem health is possibly threatened by a high level of organic carbon; furthermore, the trends of increasing population, deforestation, temperature and rainfall will likely increase the concentration of organic carbon in the future. Better management of waste, afforestation and reforestation are recommended to restore degraded natural forest, so as to reduce uptake of organic carbon from the terrestrial environment.

Additional keywords: water chemistry.

Received 22 October 2017, accepted 26 January 2018, published online 8 June 2018

Introduction

Poor water quality is a major cause of disease and death in most developing countries (Muhammad et al. 2017). Diarrhoea, cholera, hepatitis A and lead poisoning are some of the more common diseases from contaminated water. Since organic carbon is a pollutant on its own, but also carrier of pollutants, understanding the sources, forms and concentration of organic carbon is an important step in managing water quality (Balakrishna and Probst 2005; Huser et al. 2011). In rivers, organic carbon originates from autochthonous and allochthonous sources. Autochthonous sources cover organic carbon from biomass of riverine plankton, whereas allochthonous source are organic carbon from terrestrial sources including soil, terrestrial plants, industrial and domestic wastes (Gao et al. 2002; Kubo and Kanda 2017). Allochthonous organic carbon gets into the river via leaching and surface runoff (Lee and Choi 2009). The organic carbon can be transported either as dissolved (DOC) or particulate organic carbon (POC). Likewise, the concentration of total suspended matter (TSM), which covers organic and inorganic materials, is another concern influencing water quality for drinking as well as for aquatic ecosystem health. High levels of TSM in rivers can increase treatment costs and also reduce light penetration, which interferes with primary productivity and fish growth (Zhang et al. 2017). Concentrations of

TSM in river basins are influenced by basin area, slope, temperature, runoff, lithology and human activities (Tamooh et al. 2014).

On the one hand, organic carbon is among the important components in aquatic ecosystems for buffering water pH, absorbing ultraviolet radiation and providing energy for aquatic organisms (Zigah et al. 2017). On the other hand, a high content of organic carbon can result in a brownish tint or dark colouration of the water, and can influence the taste and odour of the water (Ledesma et al. 2012). In addition, colouration can reduce light penetration, which interferes with aquatic life (Worrall and Burt 2010). There is a link between the concentration of organic matter and formation of carcinogenic disinfectant by-products, including trihalomethanes and haloacetic acids. These result from the chlorination process when treating drinking water with a high content of organic carbon (Aschermann et al. 2016). Other impacts of organic carbon are their high affinity to trace metals, which can lead to the formation and transportation of toxic metal complexes (Balakrishna and Probst 2005); while photochemical and microbial breakdown of organic carbon releases CO_2 and methane to the atmosphere, which accelerate global warming (Cory et al. 2013). Thus, organic carbon can have both positive and negative effects on the environment, and on human and aquatic ecosystems, depending on the amount present in the water.

Population increase, urbanisation, elevated CO_2, temperature increase and deforestation are some of the factors which influence carbon stock, production and transport from land to ocean (Freeman et al. 2004; Tian et al. 2013; Noacco et al. 2017). Recent impacts of increasing carbon content have been reported from the Mackenzie River in Canada and São Paulo State in Brazil (Cunha et al. 2016; Tank et al. 2016). The increase in organic carbon reduced the quality of water in São Paulo State, thereby increasing the cost of treating drinking water; while the increase in organic carbon in the Mackenzie River has changed coastal processes such as bacterial metabolism and food-web structure (Cunha et al. 2016; Tank et al. 2016).

An empirical model estimated that global rivers transport ~390 Tg $year^{-1}$ of organic carbon to the ocean; of which 15 % is transported by rivers draining the tropical coast to the western Indian Ocean (Ludwig et al. 1996*a*). It is clear that a global model gives reasonable information on global use but the estimate does not provide sufficient information about regional use, including pollution control at local level (Dai et al. 2017). Beyond model estimates, field measurement is the best way of estimating riverine carbon flux and pollution control at a regional level (Li et al. 2017). Several field studies have been conducted on large tropical African rivers discharging to the Atlantic Ocean such as in Congo, Niger, and Senegal (Martins and Probst 1991; Spencer et al. 2016). However, less is known about the concentration and contribution of organic carbon from the Rufiji, Ruvuma, Ruvu and Wami rivers to the Indian Ocean (Bouillon et al. 2007). Some of the few available studies conducted in the Tana and Zambezi rivers reveal significant input of organic carbon to the Indian Ocean (Bouillon et al. 2007; Teodoru et al. 2015). Therefore, to bridge the gap between west and east Africa, this study was conducted to broaden the dataset of organic carbon flowing into the Indian Ocean. It is one of the first studies of organic carbon including surface and groundwater in the Pangani River Basin (PRB) as an important tropical basin in East Africa. The aims of the present research were (i) to study concentration and factors regulating spatial and seasonal variability of DOC, POC and TSM, together with their possible impacts on human and aquatic-ecosystem health and (ii) to give quantitative estimates of DOC, POC and TSM fluxes, so as to quantify the contribution from the PRB to the Indian Ocean. In addition, by using $\delta^{13}C$, possible sources of organic carbon were identified in the basin. Locally, understanding the sources, and the spatial and seasonal variation of organic carbon can help water-resource managers in planning and implementing effective measures to control water pollution. Knowledge of carbon fluxes from the PRB to the Indian Ocean coast will improve regional and global estimates for the carbon budget. This research reveals spatial and seasonal variation of TSM and organic carbon in the basin. The significance of that variation was tested with a paired Student's *t*-test.

Experimental

Study site

Details of the study area have been previously described (Selemani et al. 2017*a*, 2017*b*). Briefly, the PRB can be separated into two parts: Pangani headwater, consisting of streams originating from mountain ranges of Meru, Kilimanjaro, Pare and Usambara. Mount Kilimanjaro and Mount Meru are composed of young volcanic rocks; therefore, upstream is characterised by volcanic soils, whereas downstream, together with Mount Pare and Mount Usambara, are composed of Precambrian rocks (Selemani et al. 2017*b*). Streams from Mount Kilimanjaro and Mount Meru feed directly into the reservoir, whereas streams from Mount Usambara feed directly into the main Pangani River (Fig. 1). Geologically, the basin is composed of volcanic rocks in the upstream and sedimentary rocks along the river mouth, while a large part is covered by Precambrian metamorphic rocks (Selemani et al. 2017*b*) (Fig. 1b). Highlands are characterised by fertile soils with rainfall ranging from 1000 to 2000 mm per year (PBWB/IUCN 2008). As a result of these conditions, the highlands consist of dense forest rich in biodiversity and agricultural activities, and there is a high population-growth rate >4 % (Pamba et al. 2016). Lowlands consist of the main Pangani River extending 500 km from the reservoir to the Indian Ocean. The lowlands consist of grasslands and open forest vegetation, receiving less rainfall than the highlands.

Materials and methods

Water was sampled in two cruises; dry season (October 2014) and wet season (May 2015). Pre-cleaned 1 L plastic bottles and a 5 L plastic bucket were used to sample water. A bucket attached to a string was used to sample from the top of the bridges. At other stations, local boats were used to sample in the middle of the river or lake so as to avoid anthropogenic impacts along the river banks. Before sampling, the sampling devices were rinsed at least three times with the water to be sampled so as to minimise contamination. Water samples from 39 stations, including surface- and groundwater (geographical locations of the stations are presented in Table 1) were collected for DOC, whereas samples from 33 stations covering only surface water were collected for POC and TSM. Many stations are located in the upstream because there are many streams and groundwater feeding the reservoir (Fig. 1). We aimed to identify the contribution of each stream before mixing in the reservoir in order to be able to control the sources of pollution.

Samples for TSM were obtained by filtering a known volume of water through pre-cleaned and pre-weighed cellulose acetate filters of pore size 0.45 μm. Similarly, samples for POC were

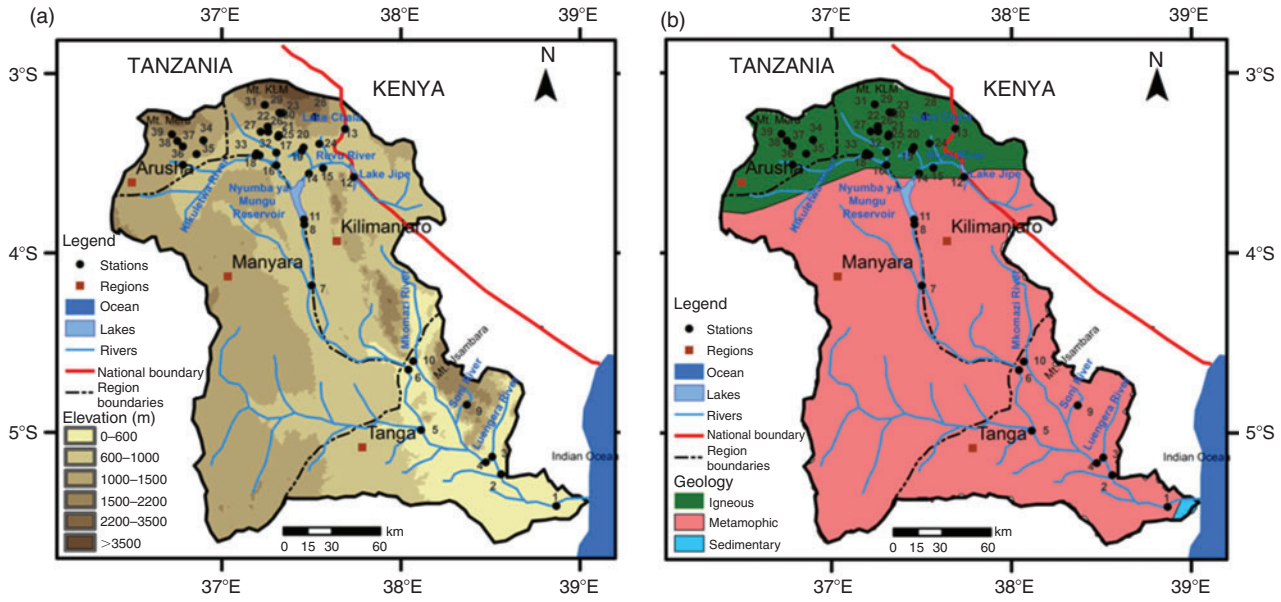

Fig. 1. Study area, showing sampling stations, regions forming the basin and (a) elevation, (b) geology. The station numbers presented in the map are the same as those presented in Table 1.

obtained by filtering a known volume of water through pre-combusted and pre-weighed 47 mm Whatman GF/F filters of pore size 0.7 µm. After filtration, the GF/F and cellulose acetate filters containing particulate matter were stored at −20 °C. For DOC, syringe filters (nylon membrane of 0.45 µm pore size), were used to filter water samples. The syringe was rinsed three times, then the first filtered 5–10 mL of water sample was used to flush the filter and rinse the pre-combusted (450 °C for 5 h) glass tubes twice. Filtered samples were then kept in glass tubes sealed with Teflon-coated screw caps. Phosphoric acid was used to preserve the samples at pH ≤ 2, which were then stored at −20 °C.

Soil and sediment samples were collected to estimate the concentrations of carbon and nitrogen in the soil and sediment. After sampling, the samples were dried at 40 °C in the oven. Similarly, leaves from C_3 (rice leaves and grasses) and C_4 (maize and sugarcane) plants were also collected to determine sources of carbon in the basin. After the cruise, all samples were packed in an ice box and transferred to State Key Laboratory of Estuarine and Coastal Research of East China Normal University for further analysis.

In the laboratory, the cellulose acetate filters were dried at 55 °C and re-weighed to determine the TSM. The sample preparation and analysis of POC, particulate nitrogen (PN), $\delta^{13}C$ and $\delta^{15}N$ followed the methods described by Zhang et al. (2007). In this case, the frozen Whatman GF/F filters for POC were dried at 55 °C and reweighed. Thereafter, one quarter of the GF/F sample was weighed and decarbonised by using HCl vapours for 48 h (Wu et al. 2007). The sample was then dried at 55 °C and packed in a tin boat. The remaining three quarters of the GF/F was prepared for PN analysis and packed in tin boats ready for measurement. POC from soil and sediment samples was treated with acid, whereas PN from soils, sediments and leaves was not treated with acid. Carbon and nitrogen isotopes were analysed by using a Finnigan EA 1112 elemental analyser interfaced with a Finnigan Delta *plus XP* continuous-flow isotope mass spectrometer (CF-IRMS). Intra-laboratory standards of IAEA-600 caffeine, IAEA CH3 cellulose and IAEA-N2 ammonium sulfate were analysed several times to correct $\delta^{13}C$ and $\delta^{15}N$ values of samples; the calculated precision was ±0.1‰ and ±0.2‰ for $\delta^{13}C$ and $\delta^{15}N$, respectively. POC and PN were also analysed by using an elemental analyser, with intra-laboratory standards of IVA99995 soil standard. The estimated precision was <4 % and <6 % for POC and PN respectively. POC (mg L^{-1}) was calculated from POC %, whereby POC % is the percentage of POC per dry weight (POC % = POC (mg)/total particulates (mg) × 100) and POC (mg L^{-1}) is weight per volume of water. DOC was analysed by using a Shimadzu Total Organic Carbon (TOC) analyser (model: TOC L-CPH) following the method described by Wu et al. (2007). DOC data are the mean of three to five replicate injections, for which the precision was ±2–3 %. Inorganic carbon (HCO_3^-) and electrical conductivity (EC) data were measured by using the method published in Selemani et al. (2017*b*). In brief, water samples for HCO_3^{-1} were filtered through cellulose acetate filters and analysed by titration. Diluted HCl of known concentration was used as a titrant, while phenolphthalein and methyl orange were used as colour indicators to indicate the desired pH. Measurements were made in duplicate or triplicate, and the calculated precision was <10 %. Electrical conductivity (EC) was measured *in situ* by multi-parameter probe (Multi 350i Set 5 model from WTW, Weilheim, Germany). Standard solutions of pH 4.01 and 7.00 were used to calibrate the pH meter before measurement. The calculated precision of the instrument from repeating measurements of some selected stations was <5 %. Dissolved silicate (DSi) measurements were made following the previously published method from Selemani et al. (2017*a*) by using a Skalar SAN^{plus} continuous flow autoanalyser. Data quality was checked by repeating some selected measurements, and the calculated precision was <5 %.

Results

Spatial and seasonal variability of organic and suspended matter

Results are presented in Table 2 along with data from rivers around the world for comparison. Figure 2 shows concentration of parameters against altitude of the stations in the wet (May 2015) and dry (October 2014) seasons. Generally, from upstream

Table 1. Sampling stations, their geographical location and elevation
Station numbers presented here are the same as in Fig. 1

Name of the river	Latitude (°S)	Longitude (°E)	Number	Elevation (m)
Pangani River @ Maseko	5.40958	38.86875	1	6
Pangani River @ Mnyuzi	5.23361	38.56018	2	293
Luengera River @ the bridge	5.13515	38.50959	3	296
Pangani River @ Korogwe	5.16615	38.47371	4	287
Pangani River @ Mkalamo	4.98639	38.11254	5	489
Pangani River @ Buiko	4.64937	38.04159	6	533
Pangani River @ Naururu	4.18012	37.50136	7	639
Pangani River D/S Nyumba ya Mungu reservoir	3.84022	37.46001	8	665
Soni River @ Soni	4.84554	38.36876	9	1179
Mkomazi River @ Bendera	4.60216	38.06852	10	470
Nyumba ya Mungu reservoir	3.8128	37.45856	11	694
Lake Jipe @ Makuyuni	3.57702	37.73659	12	718
Lake Chala @ Safari lodge	3.30827	37.68885	13	847
Ruvu River @ Tingatinga	3.55712	37.48665	14	695
Ruvu River @ Kifaru	3.52601	37.56544	15	701
kikuletwa River @ TPC	3.51039	37.30484	16	712
Karanga River @ TPC	3.44025	37.30453	17	746
Chemka spring	3.44418	37.19363	18	845
Miwaleni spring	3.43086	37.44586	19	723
Miwaleni Borehole	3.43086	37.44586	20	721
chekereni/ weruweru spring	3.35182	37.31507	21	872
Nsere springs	3.29528	37.25655	22	1023
Mwenge borehole	3.21793	37.32146	23	1039
Himo River @ the bridge	3.39046	37.54489	24	841
Karanga River @ the bridge	3.34118	37.31783	25	888
Weruweru River @ the bridge	3.3244	37.2589	26	957
Kikafu River @ the bridge	3.32416	37.21686	27	976
Marawee stream @ Marangu	3.24098	37.52092	28	1845
Sungu River @ Singandoo	3.21793	37.32334	29	1542
Mweka stream @ Mweka gate	3.21967	37.34151	30	1643
Machame stream @ Machame gate	3.17448	37.2396	31	1789
Maji ya Chai River	3.37073	36.8969	34	1224
Kikuletwa River @ Karangai	3.44816	36.85841	35	1020
Kikuletwa @ kambi ya Chokaa	3.45762	37.18767	32	842
kikuletwa @ power station	3.45488	37.21064	33	834
Themi River @ Lokii mnadani	3.50879	36.78243	36	1029
Nduruma River @ NM-AIST road	3.40522	36.78165	37	1206
Nduruma River @ the bridge	3.37569	36.75114	38	1340
Themi River @ Olesha Olgilai	3.33858	36.72075	39	1569

Table 2. Concentration of DOC, TSM and POC (mean ± s.d.) in PRB and in other rivers across the world
All parameters are given in mg L^{-1} except POC (%) which is the percentage of organic carbon in dry weight of particulate and carbon isotope given in ‰

River name	DOC	TSM	POC	POC (%)	$\delta^{13}C$ (‰)	References
Dry season	2.5 ± 3.5	16.0 ± 13.2	1.2 ± 1.2	16.9 ± 16.6	−25.5 ± 2.5	This study
Wet season	2.7 ± 3.7	22.9 ± 16.0	1.6 ± 0.9	9.0 ± 6.4	−25.0 ± 2.4	This study
Zaire/Congo	7.2–11.8	15.5–27.2	1.01–1.97			[Spencer et al. 2016]
Nile	2.23–11.3					[Badr 2016]
Niger	3.5	127	2.59			[Martins and Probst 1991]
Orange	2.3		0.9			[Martins and Probst 1991]
Gambia River	2.39					[Lesack et al.1984]
Heihe River	1.39					[Hu et al. 2016]
Songhua River	7.4		152.5			[Sun et al. 2017]
Tana	2.7	802 ± 464	8.6 ± 4.5			[Bouillon et al. 2007]
Tapti	8.59					[Krishna et al. 2015]
Narmada	4.24					[Krishna et al. 2015]

to the river mouth, DOC, POC and TSM varied widely from 0.11 to 18.7, 0.04 to 5.63 and 1.8 to 63.0 mg L^{-1} respectively. The lowest DOC concentration (0.11 mg L^{-1}) was measured at Chemka Spring (station number 18 (Fig. 1), at 845 m above sea level (asl) (Fig. 2a), whereas the highest DOC of 18.7 mg L^{-1} was measured in Lake Jipe (station number 12, at 718 m asl). The

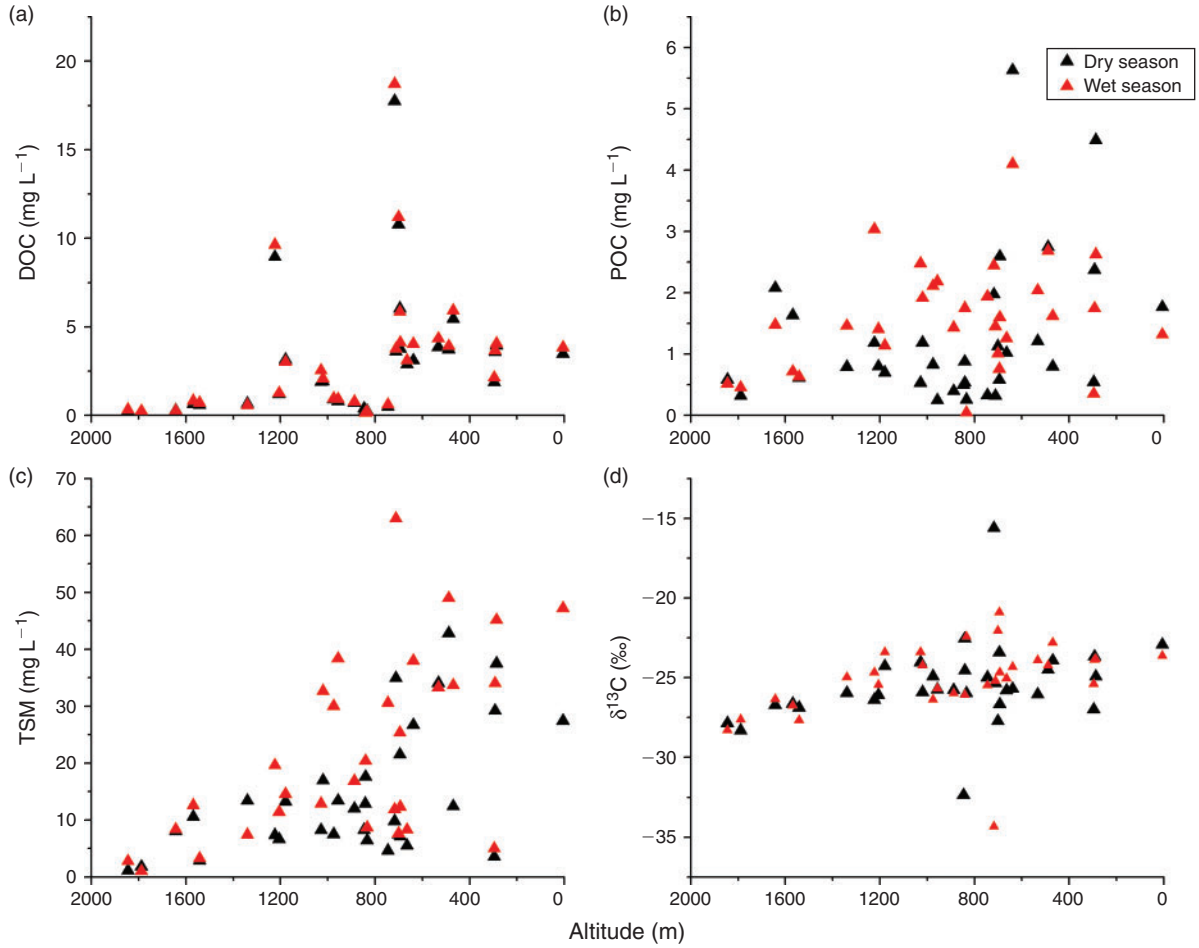

Fig. 2. Variation of organic carbon with elevation: (a) DOC in mg L^{-1} (b) POC in mg L^{-1}, (c) variation of TSM in mg L^{-1} and (d) variation of δ^{13}C. Note the increasing trend from upstream to the river mouth with the increase in DOC, POC, TSM and δ^{13}C in wet season compared with dry season.

lowest concentrations of POC and TSM were measured at Kikuletwa power station (station number 33, 834 m asl; Fig. 2b) and stream at Machame gate (station number 31 at 1789 m asl; Fig. 2c), respectively. The highest concentrations of POC and TSM were measured in Pangani River at Naururu (station number 6 at 639 m asl; Fig. 2b) located in Kirua swamp and Kikuletwa River at TPC (station number 18 at 712 m asl; Fig. 2c) located in the sugar plantation, respectively. The concentrations of DOC, POC, TSM and δ^{13}C tended to increase from high altitude to river mouth and also from dry season to wet season (Fig. 2, Table 2). PN was, however, not reported because the concentration of nitrogen was below the instrument detection limits.

The basin was divided into three main tributaries and the main Pangani River from the reservoir to river mouth. For example, streams from Mount Kilimanjaro were grouped as one tributary, and a similar case applied to Mount Meru and Mount Usambara (Fig. 1). The sums of organic carbon (POC + DOC) for the main Pangani River, Usambara, Meru and Kilimanjaro tributaries were 6.18, 5.44, 3.76 and 1.19 mg L^{-1} respectively, while the POC/DOC ratios were 0.43, 0.27, 1.1 and 2.71 respectively. The sums of organic carbon for Lake Jipe, Challa and Nyumba ya Mungu reservoir were 20.44, 0.88 and 6.03 mg L^{-1}, respectively. For groundwater samples, only DOC was measured, which ranged from 0.11 to 0.26 mg L^{-1} (mean 0.16 mg L^{-1}). These results show that the highest level of organic carbon was measured in the main river, while the lowest level was measured in tributaries from Mt. Kilimanjaro. For POC/DOC, the lowest ratio occurred in the main river compared with tributaries in the upstream. Results from the lakes show that the highest level of organic carbon was measured in Lake Jipe and the lowest in Lake Challa. The sums of TSM were 32.72, 18.45, 12.48 and 3.65 mg L^{-1} for the main river, Usambara, Meru and Kilimanjaro tributaries, respectively.

The concentration of organic carbon in the soil varied depending on the location of the sampling sites. Samples collected near the forest reserve of Mount Kilimanjaro, with a mean percentage of 22 %, had higher percentages of organic carbon than other stations. The lowest percentage of organic carbon was measured in bare land, where the mean percentage of organic carbon was 0.37 %. The high percentage of organic carbon in the soils of the Kilimanjaro forest reserve reflects the volcanic nature of the soil..

A positive correlation was found between POC (mg L^{-1}) and TSM in both seasons (Fig. 3b, Table 3), whereas POC % decreased with increasing TSM in all seasons (Fig. 3c, Table 3). There was a negative correlation between TSM and altitude in both seasons (Fig. 2).

Nyumba ya Mungu reservoir (694 m asl) collects water and pollutants from upstream, while its output is through the main Pangani River on the way to the river mouth. The content of DOC, POC and TSM entering the Nyumba Ya Mungu reservoir comprised of input from stations number 14 and 16 (Fig. 1). In

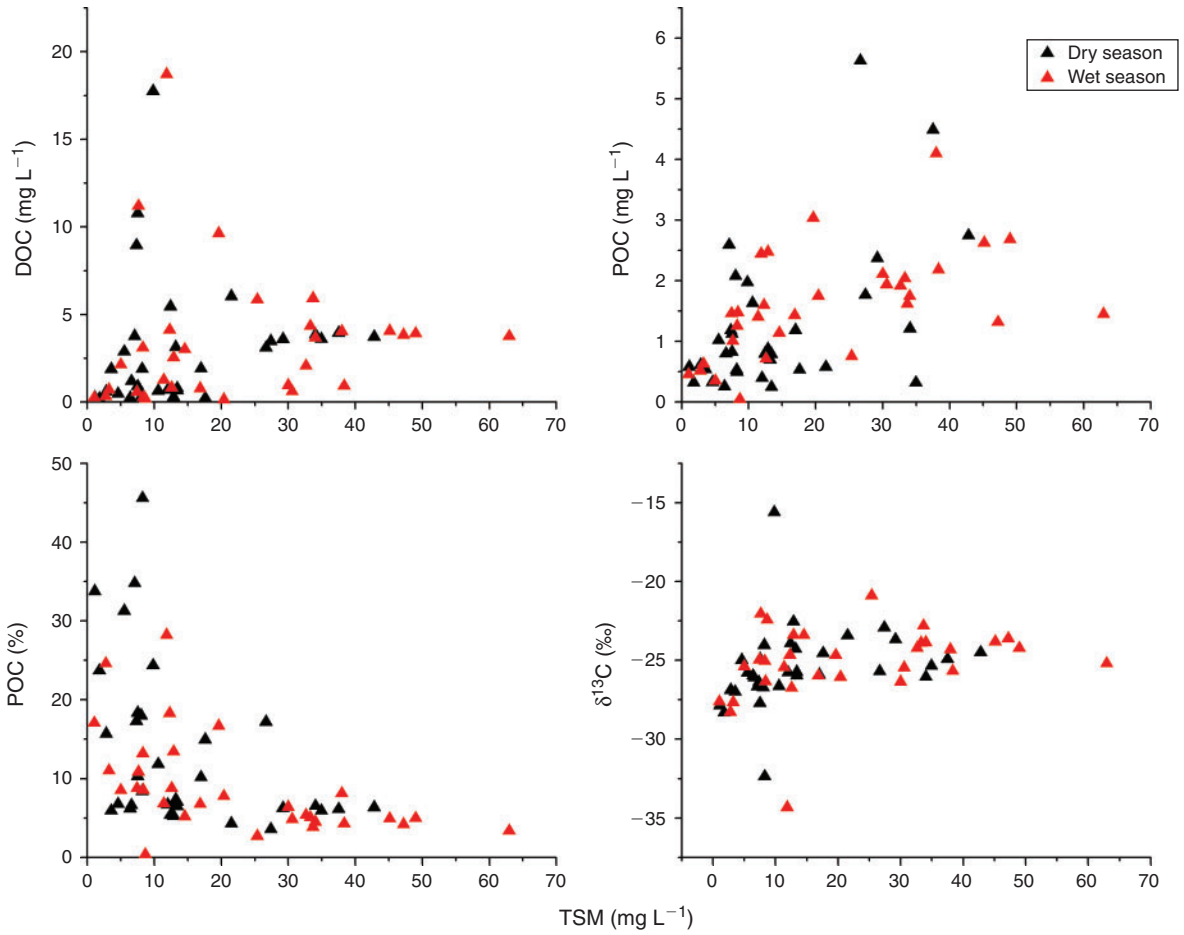

Fig. 3. Variation of DOC, POC (mg L^{-1}), POC % and δ^{13}C against TSM in dry and wet seasons. Note: POC (mg L^{-1}) increases with TSM but POC % decrease with the increase of TSM in both seasons.

Table 3. Correlation table of different parameters in dry and wet seasons
* indicates correlation is significant at $P \leq 0.05$; ** indicates correlation is significant at $P \leq 0.01$

Parameter	Altitude	TSM	DOC	POC	δ^{13}C	%OC	Temp	DSi	DO	EC	Salinity	pH
Altitude												
TSM	−0.59**											
DOC	−0.32	0.14										
POC	−0.33	0.55**	0.33									
δ^{13}C	−0.31	−0.01	−0.04	−0.16								
%OC	0.39*	−0.52**	0.26	0.02	−0.22							
Temp	−0.75**	0.52**	0.51**	0.45**	−0.23	−0.21						
DSi	−0.15	0.09	0.49**	0.08	0.05	−0.02	0.37*					
DO	0.55**	−0.02	−0.48**	0.06	−0.49**	0.06	−0.28	−0.23				
EC	−0.56**	0.35*	0.67**	0.43*	0.17	0.01	0.68**	0.44**	−0.48**			
Salinity	−0.54**	0.29	0.63**	0.40*	0.17	0.03	0.63**	0.40*	−0.46**	0.98**		
pH	−0.31	0.43*	0.36*	0.52**	−0.61**	−0.09	0.76**	0.19	0.17	0.37*	0.30	

both seasons, the concentrations of DOC, POC and TSM entering the reservoir were higher than in the outflow from the reservoir (Fig. 2a–c). The average annual fluxes of DOC, POC and TSM into the Nyumba Ya Mungu reservoir were 4.58×10^6, 0.74×10^6 and 34.47×10^6 kg year^{-1}, respectively, while the corresponding outflows from Nyumba Ya Mungu reservoir were 1.42×10^6, 0.54×10^6 and 3.28×10^6 kg year^{-1}.

The contribution of PRB to the Indian Ocean was estimated as the combined fluxes of DOC, POC, DIC and TSM measured at station number 1 (Fig. 1). The flux of DOC ranged from 1.65×10^6 to 1.83×10^6 kg year^{-1}, POC ranged from 0.63×10^6 to 0.84×10^6 kg year^{-1}, dissolved inorganic carbon (DIC) ranged from 147.8×10^6 to 177.8×10^6 kg year^{-1} and TSM ranged from 13.05×10^6 to 22.48×10^6 kg year^{-1}. The fluxes were all higher in the wet season than in the dry season (Table 4).

The measured stable carbon isotopic signature from particulate carbon (δ^{13}C) ranged from −34.3‰ to −15.6‰. Seasonal

Table 4. Carbon flux and yield from the PRB and other rivers

River	Country	Area (10^3 km^2)	Annual flux					Annual yield			References
			Discharge (M^3 s^{-1})	TSM (kg year^{-1})	POC (kg year^{-1})	DOC (kg year^{-1})	DIC (kg year^{-1})	TSM (kg km^{-2} year^{-1})	POC (kg km^{-2} year^{-1})	DOC (kg km^{-2} year^{-1})	
PRB	Tanzania	39.8	15.1	17.76×10^6	0.73×10^6	1.74×10^6	162.8×10^6	446.28	18.44	43.72	This study
Congo	Republic of Congo	3500	41134	29.21×10^9	1.96×10^9	12.5×10^9			560	3571.43	[Spencer et al. 2016]
Niger	Niger	1200	4886		0.66×10^9	0.53×10^9			500	440	[Martins and Probst 1991]
Orange	South Africa	1020	360		0.01×10^9	0.03×10^9			10	29.4	[Martins and Probst 1991]
Songhua	China	567	2406.8		0.08×10^9	0.17×10^9			140	300	[Sun et al. 2017]
Wanquan	China	3.69	164.9		2.51×10^3	8.49×10^3			0.68	2.3	[Wu et al. 2013]
Godavari	India	300	2830		650×10^6	130×10^6			2166.7	433.33	[Sarin et al. 2002]
Tapti	India	65	471.5			0.128×10^9				1969.2	[Krishna et al. 2015]
Narmada	India	99	1446			0.193×10^9				1949.5	[Krishna et al. 2015]
Cauvery	India	88	676.6			0.191×10^9				2170.5	[Krishna et al. 2015]
Tana	Kenya	120	126.8	3.21×10^9	3.44×10^7	1.19×10^7	9.4×10^7		286.7	99.17	[Bouillon et al. 2007]
Global average									1627	1926	[Ludwig et al. 1996*b*]

and spatial variation showed that the $\delta^{13}C$ signature was enriched in the wet season relative to the dry season. It was also enriched downstream relative to the upstream (Fig. 2d). Analysed plant leaves showed that $\delta^{13}C$ for C_4 leaves ranged from −11.9‰ to −10.8‰ (mean -11.5 ± 0.54‰), whereas for C_3 leaves it ranged from −29.2 to −28.6‰ (mean -28.9 ± 0.43‰).

A cluster analysis was also carried out to determine similar stations based on the spatial variation of DOC, POC, TSM and $\delta^{13}C$. Two main clusters were found, with six subclusters (Fig. 4). The first subcluster included stations 1, 2, 4, 5, 6, 7, 8, 11 and 16, which were located between the Nyumba Ya Mungu reservoir and river mouth. The second subcluster included stations 3, 9, 10, 35, 36, the first three of which (3, 9 and 10) were from Precambrian metamorphic rocks, whereas stations 35 and 36 were from volcanic rocks of Mount Meru. One subcluster contained two stations (15 and 34), which both collect water from swamps although they are located far from each other. Station 15 is located after Kirua swamp, while station 34 located after Maji ya Chai swamp.

To test the hypothesis that the spatial and seasonal variation of TSM and organic carbon in the basin is significant, a paired Student's *t*-test was used to test seasonal variation; the results showed that there was a significant increase of TSM ($r^2 = 0.69$, $P < 0.001$) and DOC ($r^2 = 0.99$, $P < 0.001$) in the wet season relative to the dry season. On the other hand, the variation of POC and $\delta^{13}C$ were not statistically significant.

Discussion

Possible sources of organic carbon in the basin

Most of the C_4 plants found in this basin are maize and sugarcane while C_3 plants are rice, beans, tomatoes and other vegetables. Those plants are grown near the river banks. Human settlements are also located along the river banks with domestic wastes being poured into the river. The increase of POC in the wet season suggests that most of the carbon is from allochthonous sources (soil, plants and anthropogenic waste), since rain water mobilises the carbon from allochthonous sources to the river. This result reveals the dominant role played by allochthonous organic carbon relative to that from autochthonous sources (Sun et al. 2017). The increase of DOC and POC in the wet season may also be influenced by seasonal farming of maize, rice and other vegetables. Farming practices and rainfall enhance leaching of DOC from the soils, while POC increases due to erosion of soil particles carrying organic carbon (Zuijdgeest et al. 2015). Positive correlation between TSM and POC (Table 3) suggests that TSM and POC share common sources, as flowing water mobilises particles (through mechanical erosion) as well as organic carbon stored in soil particles. In addition, positive correlation between DOC and electrical conductivity in both seasons (Table 3) may also reflect the allochthonous source of DOC, since chemical weathering increases both DOC and electrical conductivity.

The $\delta^{13}C$ end member for C_4 plants was −11.4‰ but for C_3 plants it was −28.9‰. These values are within the proposed regional end member of −12‰ and −26‰ for C_4 and C_3 plants, respectively (Muzuka 1999). Most of the stations in PRB had $\delta^{13}C$ values ranging from −28.3 to −23.4‰ (Fig. 3d); suggesting a dominant supply of carbon from C_3 plants. The values are comparable to tropical rivers dominated by C_3 carbon sources, including Tana ranging from −19.9‰ to −31.8‰ and Congo ranging from −28.0‰ to −26.1‰ (Tamooh et al. 2012; Spencer et al. 2016). Additionally, a downward increase of $\delta^{13}C$ (Fig. 3d) is opposite to the pattern observed in the

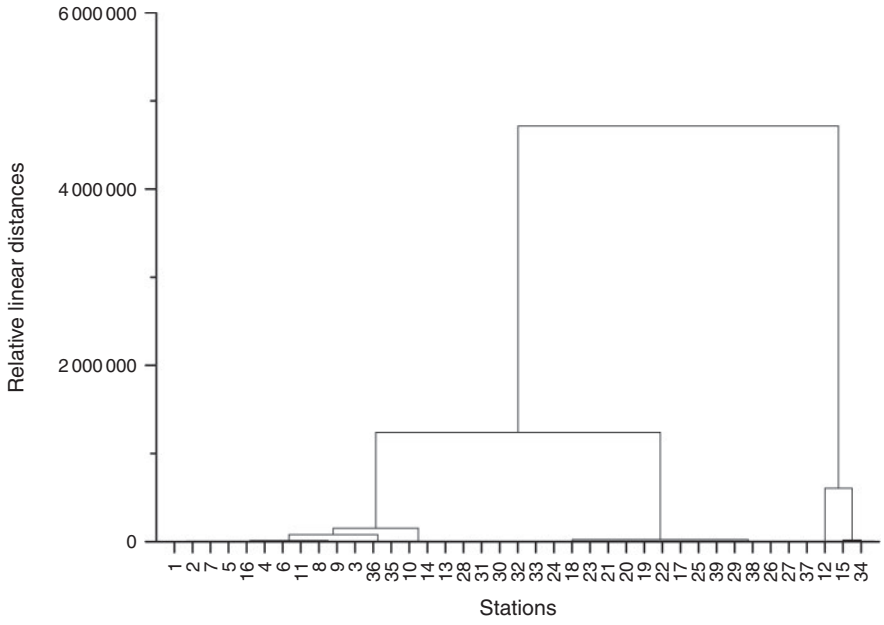

Fig. 4. Cluster analysis grouping stations with similar characteristics.

tropical Amazon River but similar to the tropical Tana River. A possible cause of the downward increasing $\delta^{13}C$ is decreasing rainfall, since rainfall in PRB decreases downward, which can increase the $\delta^{13}C$ value of C_3 plants (Tamooh et al. 2012).

Lake Jipe (station number 12, Fig. 1) had an exceptional signature of $\delta^{13}C$ values ranging from −15.61 to −34.3‰ in dry and wet seasons, respectively. Similar values ranging from −18.9‰ to −16.8‰ were observed in sediment from Lake Victoria, which was dominated by *Cyperus papyrus* with a $\delta^{13}C$ value of −13.5‰ (Gichuki et al. 2005). Lake Jipe is also dominated by *Cyperus papyrus*, which raises the possibility that $\delta^{13}C$ originates from C_4 *Cyperus papyrus* in the dry season. The depleted $\delta^{13}C$ value of −34.3‰ in the wet season was in the same range as the value from Lake Challa; −32.4‰ (station number 13, Fig. 1). Previously published analyses of $\delta^{13}C$ from diatom samples of Lake Challa ranged from −27.3‰ to −36.4‰ (Barker et al. 2013). The measured organic carbon from Lake Challa may be from diatoms, since it is a tropical crater lake. It is likely that even the organic carbon with depleted $\delta^{13}C$ value in Lake Jipe may also originate from diatoms. Analysed terrestrial C_3 plant leaves from Tanzania ranged from −26.7‰ to −31.9‰ (Muzuka 1999). The depleted $\delta^{13}C$ value in Lake Jipe measured in the wet season suggests that runoff rainwater collected some organic carbon from allochthonous sources. It is also possible that, in the season studied, carbon in Lake Jipe was a mixture of carbon from diatoms and allochthonous sources (Muzuka 1999). Future work should define sources of carbon in Lake Jipe, especially in the wet season.

Sources of organic carbon in other tropical African rivers varied widely depending on vegetation cover, relief and anthropogenic activities (Martins and Probst 1991; Badr 2016; Spencer et al. 2016). For example, most organic carbon came from allochthonous sources with C_3 as the major source of DOC in the Tana River (Bouillon et al. 2007). Vascular plants dominated by C_3 were also a dominant source of organic carbon in the Congo River (Spencer et al. 2016). Allochthonous sources dominated the organic carbon in the Niger River (Martins and Probst 1991), while anthropogenic wastes were the major source of DOC for the Nile River (Badr 2016). From those examples, it is clear that most tropical African Rivers are dominated by a supply of allochthonous organic carbon, whereas in lakes and reservoirs there is contribution from autochthonous sources (Barker et al. 2013; Lambert et al. 2015).

High levels of organic carbon (% OC) in the particulate matter were observed in Lake Jipe, Challa and Nyumba ya Mungu reservoir, which were 24.3, 45.6 and 34.8 % respectively. The % OC was in comparable to that seen in the tropical Tana River (ranging from 1.1 % to 49.8 %) but higher than subtropical Yangtze River (ranging from 0.5 % to 2.5 %: Wu et al. 2007; Tamooh et al. 2012). The PRB, like other basins in developing countries, reflects poor management of domestic and industrial wastes. Observed high % OC in the PRB, especially in lakes, might have been accumulated from soil, plant residues, and decomposed domestic and industrial waste. We did not analyse isotopic data from DOC to ascertain the source of DOC; nonetheless, the increase in DOC with rainfall, and knowledge of cultivation season suggest that it comes from allochthonous sources, including anthropogenic wastes. A similar pattern was also observed in tropical African rivers including Congo, Tana, Zambezi and Niger (Martins and Probst 1991; Lambert et al. 2015; Spencer et al. 2016). Therefore, organic carbon in the PRB originates from allochthonous sources dominated by C_3 plants, whereas in lakes, there is also a contribution from C_4 plants and autochthonous sources.

Factors regulating suspended matter and organic carbon in the PRB

Loading of suspended and dissolved matter in any river is a function of climate, lithology, vegetation cover, topography, discharge and human activities of the area (da Costa et al. 2017). The trend of increasing TSM, POC and DOC in the wet season (Table 2, Fig. 2) has been contributed to by hydrology and anthropogenic activities. The PRB has a population of ~6.8 million people, with a mean annual growth rate of 2.4 % (National Bureau of Statistics 2013). Increase in population goes hand in hand with increase in sewage discharge and the

conversion of natural forest into agricultural land. It was estimated that, from 1952 to 1982, ~41 km^2 of natural forest in PRB was converted to agricultural and human settlement (Yanda and Shishira 2001). Such forest removal enhances erosion, which takes particulates and organic carbon from terrestrial environment to the river, increasing the concentration of TSM and organic carbon transported by river.

Under normal conditions, most of the measured organic carbon in the dry season was eroded from riverbed, riverbank, *in situ* produced carbon and carbon from domestic and industrial effluents discharged into the river. Rainfall increases surface runoff, which enhances uptake of organic carbon away from the river, including domestic wastes. Increasing organic carbon with discharge has also been reported in the arid basin of Heihe River (Hu et al. 2016), and the tropical basins of the Wanquan, Maji Ya Chai, Niger, Tana and Congo rivers (Martins and Probst 1991; Bouillon et al. 2007; Wu et al. 2013; Aschermann et al. 2016; Spencer et al. 2016). In those rivers, agricultural activities, deforestation, domestic and industrial effluents have been causes of concern.

The construction of the Nyumba ya Mungu reservoir is another human influence on suspended and organic matter in PRB. The reservoir has an area of ~150 m^2, having been constructed in 1965 for flood control, production of electricity and stored water for irrigation. The reservoir has previously been found to be a nutrient sink (Selemani et al. 2017*a*), while we have also found that the reservoir is a TSM and organic carbon sink, which has significant influence on the distribution of TSM and organic carbon in the basin. Trapping materials usually limit normal flow and reduce TSM and organic carbon downstream. Therefore, there must be several factors that have played roles to increase TSM and organic carbon downstream of the reservoir (Fig. 2). One of the factors is input downstream of the reservoir from tributaries, including Soni, Mkomanzi and Luengera, which would be expected to contribute organic carbon and TSM to the main river (Fig. 1). However, examination of the input from tributaries revealed that they do not significantly increase TSM or organic carbon downstream. In addition, just as the geology of the PRB varies from upstream to downstream, so too do the concentration of organic carbon in the soil, and anthropogenic input differ from upstream to downstream. For example, a study by Hellar-Kihampa et al. (2013) found high content of organic carbon from sediment collected in Precambrian metamorphic rocks located downstream of the basin. Their finding concurs with the study from the Yellow River, where organic-matter-rich soil in the Qinghai-Tibetan Plateau raised the content of DOC in the upper reach (Zhang et al. 2013). In addition, a study of tropical Indian rivers also showed a linear relationship between DOC fluxes and the content of organic carbon in the sediment (Krishna et al. 2015). An effect of lithology and relief was also observed in the tropical Gambia River, where a high concentration of organic carbon was measured downstream relative to upstream (Lesack et al. 1984; Lambert et al. 2015).

The influence of temperature and salinity on chemical weathering can be seen in the correlation of these two factors with DOC levels (Table 3). This is consistent with global observations that tropical rivers have higher concentration of organic carbon than temperate and arctic rivers (Spencer et al. 2012). Therefore, the higher temperature and salinity downstream of the PRB than upstream may enhance leaching of organic carbon from Precambrian rocks and soils, leading to an increased concentration of DOC downstream.

Stations were grouped based on similar characteristics in the cluster analysis, which grouped most of the stations downstream into one sub-cluster (Fig. 4). Since the river flows through various lithology, the spatial variation of TSM shows that physical weathering differs for different rocks. A high rate of weathering occurs in Precambrian rocks, elevating the level of TSM downstream of the basin, whereas the low rate of weathering in young volcanic rocks reduces the level of TSM upstream (Selemani et al. 2017*b*). The positive correlation between TSM and temperature may reflect an increase in the rate of erosion with temperature. Furthermore, the temperature increases as the elevation decreases. High levels of TSM downstream may also have resulted from higher temperatures. The strong negative correlation between altitude and TSM is consistent with an increase of TSM with decreasing altitude (Table 3). A similar trend of increasing TSM downstream was also observed in Tana River, which was reported to arise from weathering downstream and input from tributaries downstream (Tamooh et al. 2012). Therefore, the high content of organic carbon and TSM downstream of the reservoir may result from the Precambrian rocks, rate of weathering and input from tributaries.

In the PRB, there was a positive correlation between DOC and dissolved silicates (DSi) (Table 3). Since dissolved silicates arise mainly from weathering, the positive correlation with DOC suggests a relationship between weathering and leaching of DOC. Because DOC results from the degradation of organic matter, the positive correlation of temperature with DSi and DOC may reflect that both weathering and degradation of organic matter increase with temperature. Figure 5a shows that, in the main Pangani River (from the reservoir to the estuary), leaching of DOC increases with DSi in a downslope direction. The mean ratio of DOC/DSi was 0.4, showing that leaching of DOC from the soil/rocks was almost half due to weathering. Since DOC and DSi depend on temperature, the expected future increase in temperature due to global warming will likely increase weathering of DSi and leaching of DOC at a ratio of 0.4.

For most of the tropical rivers, DOC/DSi increased linearly with runoff (Ramesh et al. 1995; Gaillardet et al. 1999; Badr 2016) (Fig. 5b). On one hand, for rivers with low runoff such as the Tana, the Nile and the PRB their DOC/DSi increases with runoff. On the other hand, in rivers with high runoff, like the Amazon and the Caura, the DOC/DSi will no longer increase with runoff. Instead, the ratio can either remain the same or start to decrease (Fig. 5b).

The mean concentration of DOC from the PRB was within the range of tropical rivers, ranging from 2.04 to 15 mg L^{-1} (Martins and Probst 1991), and lower than the global average of 5.29 mg L^{-1} (Dai et al. 2012). The mean concentration of DOC in the PRB was higher than in the Orange and Heihe rivers, but was comparable to that in the Niger, Nile and Tana rivers (Table 2). The higher concentration of DOC in the PRB than in the Orange and Heihe rivers is consistent with the trend of increasing organic carbon from semiarid to tropical rain forest (Meybeck 1982). High vegetation cover, rainfall and temperature enhance chemical weathering to elevate the level of dissolved organic carbon in the tropical region (Martins and Probst 1991). The concentration of DOC in the Congo River was twice as high as in the PRB. This is because the Congo River is located at the equator; most of its tributaries receive rainfall throughout the year, which mobilises much of the allochthonous organic carbon. In addition, soils in the Congo River basin are rich in organic carbon relative to the PRB (Spencer et al. 2016). High DOC and POC were also measured in the Songhua River

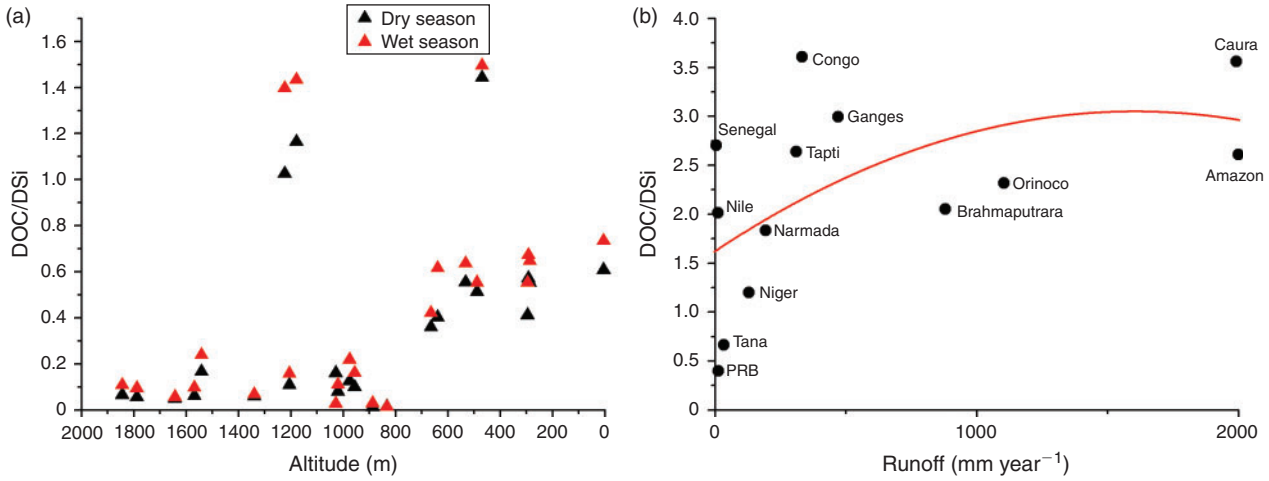

Fig. 5. How leaching of DOC and weathering of DSi varies with (a) altitude in the PRB and (b) runoff of global tropical rivers. In the main Pangani River, the DOC/DSi ratio increases with decreasing altitude (a) and there is a polynomial increase in the DOC/DSi ratio with increasing runoff (b).

from a temperate region with high soil organic carbon and intensive agricultural activities (Sun et al. 2017). The polluted rivers of Tapti and Narmada have higher concentrations of DOC than the PRB even though they have similar catchment areas to the PRB and are also located in tropical regions. Therefore, the concentration of DOC in the PRB is low compared with polluted rivers and rivers rich in soil organic carbon. The observed concentration of organic carbon in the PRB is a result of geographic location, geology, altitude, temperature, rainfall and also anthropogenic activities.

The negative correlation between POC % and TSM (Table 3) has also been observed in many rivers such as the Brahmaputra, Godavari and Zhenjiang rivers (Ludwig et al. 1996*b*; Balakrishna and Probst 2005; Gao et al. 2007). Normally, an increase in TSM comes from an increase in physical soil erosion, and that erosion can reach several soil horizons. A low rate of soil erosion can occur in surface soil, which is usually rich in organic carbon, but increased erosion can collect soil from deep horizons poor in organic matter (Zhang et al. 2009).

The POC/DOC ratio can be used to describe the transformation of POC to DOC within the river basin (Krishna et al. 2015). The ratio varies from river to river based on basin characteristics. In the PRB, the POC/DOC was higher in tributaries from Kilimanjaro Mountain (2.7 ± 2.4) than in the tributaries from Meru Mountain (1.1 ± 0.9). This reflects that the concentration of POC was higher than DOC, and the POC binds to mineral particles. The transformation of POC to DOC was therefore a slow process in the tributaries of Kilimanjaro and Meru. The POC/DOC for the main Pangani River located in Precambrian rocks was 0.43 ± 0.25. The ratio <1 indicates that biological and /or chemical dissolution of POC to DOC was a fast process (Wang et al. 2012). Globally, the POC/DOC varies from river to river based on the rate of transforming POC to DOC; the higher the rate of transformation, the lower the ratio. Most Asian rivers (e.g. the Yellow, Ganges and Yangtze Rivers) have low transforming rates and high sediment transport, and thus higher POC/DOC (Wu et al. 2007). Most of the modelled African rivers have POC/DOC ratios of around one due to poor soil organic matter (Martins and Probst 1991). In contrast, rivers like the Congo and the Tana from wet tropics have high DOC due to the surrounding wetlands and forest soils (Table 2). The Lena River, with its low sediment transport, and other estuaries of Vaigai and Ambalyaal dominated by carbon originated from autochthonous sources, also have low POC/DOC ratios (Krishna et al. 2015).

Water quality in the PRB

The high supply of DOC, POC and TSM into the Nyumba ya Mungu reservoir and the low outflows from the reservoir will impact on the ecosystem in the reservoir and downstream of the reservoir. This is because degradation of the remaining organic matter in the reservoir will decrease dissolved oxygen, which will impact the ecosystem in the reservoir (Hong et al. 2012). This effect has already been reported by Selemani et al. (2017*a*), who showed that surface dissolved oxygen in this reservoir was 7.82 mg L^{-1}, but measured dissolved oxygen from the bottom layer was 0.78 mg L^{-1}. Such a decrease in dissolved oxygen in the reservoir can have impacts on aquatic organisms. The negative correlation between DOC and dissolved oxygen may explain the negative impact of DOC on water for aquatic ecosystems (Table 3). On the other hand, the low supply of organic carbon downstream reduces the supply of food to aquatic organisms downstream.

Moreover, there is a plan to use water from the reservoir to supply the communities for drinking purposes, and possibly chlorine will be used to treat the water. The measured organic carbon in the reservoir was 6 mg L^{-1} a level that might have some human health impacts. According to the EPA Ireland (2012), chlorination of water is suitable for water with total organic carbon levels of <2 mg L^{-1}. Water with higher levels (>2 mg L^{-1}) can reduce residual chlorine and pose threat of bacterial contamination. Another possible health effect is formation of carcinogenic compounds, including organochlorides, as byproducts of the chlorination process (Aschermann et al. 2016). Total organic carbon of >2 mg L^{-1} was also measured in Lake Jipe, the main Pangani River, and in tributaries from Usambara and Meru; this is a call for management measures to reduce organic carbon levels for the well-being of both human communities and aquatic ecosystems. For groundwater samples, only DOC was measured and was shown to be lower than surface flowing water. This is because, in most cases, litter and humus occurs in the first centimetres of the soil, which increases the DOC to surface flowing water (da Costa et al. 2017). Low levels of organic carbon in the groundwater suggest suitability of water for chlorination processes and other domestic use.

The PRB is not isolated from possible climate-change impacts. A study from PWBO/IUCN (2010) predicts an increase in temperature of 1–3 °C accompanied by increased rainfall in the wet season. Another expected change is the conversion of natural forest into agricultural land and increase in anthropogenic wastes due to increasing population and economic activities. Deforestation will expose soil organic carbon, while increasing temperature can speed up decomposition of organic matter (Feng et al. 2008). Besides, the increasing rainfall will enhance soil erosion, solubility and transport of DOC, POC and TSM from allochthonous sources to the river. The increase in organic matter will possibly impact the composition, function and activity of microbial organisms, which will affect the entire aquatic ecosystem (Traving et al. 2017). The increase in organic carbon can also influence the quality of water for drinking purposes. A study from Njau and Mlay (2004) showed the effectiveness of using constructive wetland with Vetiver grass in removing nutrient pollutants; further research is needed to find the best method for removing organic carbon from the PRB.

TSM and organic carbon from the PRB to the Indian Ocean coast

The estimate of carbon flux from the river to the ocean is important for understanding how the river contributes to the local, regional and global carbon budgets (Moyer et al. 2015). The PRB contributes $\sim 165.2 \times 10^3$ t of carbon per year, of which 2.5×10^3 t is organic carbon (DOC +POC) and 162.8×10^3 t is dissolved inorganic carbon (DIC) (Table 4). Based on a previously published estimate, Ludwig et al. (1996*a*) the PRB contributes 4 % of organic carbon eroded by tropical rivers to the Indian Ocean. The results also show that more than 95 % of the carbon flux from the PRB is inorganic carbon. A high flux of inorganic carbon is common to basins rich in carbonate minerals, including the Yellow and Mississippi rivers (Wang et al. 2012). Therefore, a high flux of carbon from the PRB may be contributed by a high rate of carbonate weathering, whereas a low input of organic carbon reduces the flux of organic carbon. The flux of organic carbon was dominated by DOC; a higher flux of DOC than POC was also observed in the Congo River caused by the high rate of transforming POC to DOC and the higher rate of chemical weathering over mechanical weathering (Spencer et al. 2016). Generally, the flux of carbon from the PRB was higher than the Tana River draining the eastern coast of tropical Africa, while the flux of organic carbon was lower than in the Tana River. Similarly, the yield of organic carbon from the PRB was lower than the global average (Ludwig et al. 1996*b*). The low discharge from the PRB could be another factor reducing its organic carbon flux. For example, the mean DOC from the PRB was higher than for Orange River, but the flux from the PRB was almost 6 % compared with that of the Orange River. Diverting water from the river for irrigation and hydroelectric power production also reduces the discharge from the PRB.

The PRB pours 17.8×10^3 t per year of TSM into the Indian Ocean; the amount is lower than for the Tana and Congo Rivers. The presence of the Nyumba ya Mungu reservoir possibly reduces the flux of TSM and organic carbon from the PRB to the Indian Ocean.

Conclusions

This research investigated spatial and seasonal variation of DOC, POC and TSM in the PRB, revealing both seasonal and spatial variation of organic carbon and TSM in the basin. The organic carbon and TSM were higher in the wet season than in the dry; in addition to that, there was an increasing trend of DOC, POC and TSM from upstream to the river mouth.

Climatic factors such as temperature and rainfall, together with altitude, lithology and anthropogenic activities were some of the factors influencing level of organic carbon in the basin. Allochthonous was the main source of organic carbon in the rivers dominated by C_3 plants, whereas lakes and the reservoir get organic carbon from both allochthonous and autochthonous sources. Conversion of forest to agricultural land and some farming methods such as tillage expose soil organic carbon for photochemical or microbial reaction. Exposed organic carbon can be collected by surface flowing water, elevating DOC and POC in the river. Measures are needed to reduce deforestation and encourage best farming practices, including contour farming, which can reduce loss of soil organic carbon by surface flowing water. Proper management of anthropogenic wastes is another area in need of special attention.

It was observed that concentration of organic carbon in most of the sampling stations was higher than the level recommended water chlorination processes. Further research is also needed for investigating a suitable method to treat drinking water with high organic carbon.

Conflicts of interest

The authors declare that they have no conflicts of interest.

Acknowledgements

This study was supported by postgraduate school and State Key Laboratory of Estuarine and Coastal Research of East China Normal University under project number SKLEC-KF201502. The authors are grateful to the reviewers and editors for their comments which helped to improve the manuscript.

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Handling Editor: Ralf Ebinghaus