



Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Impacts of a high-discharge submarine sewage outfall on water quality in the coastal zone of Salvador (Bahia, Brazil)

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ARTICLE INFO

Article history:

Received 29 December 2015

Received in revised form 15 March 2016

Accepted 20 March 2016

Available online xxxxx

Keywords:

Marine pollution

Oceanic Disposal System of Rio Vermelho

Carbon and nitrogen stable isotopes

Biological oxygen demand

Shelf hydrodynamics

Health threat

ABSTRACT

Carbon and nitrogen stable isotopic signatures of suspended particulate organic matter and seawater biological oxygen demand (BOD) were measured along a coastal transect during summer 2015 to investigate pollution impacts of a high-discharge submarine sewage outfall close to Salvador, Brazil. Impacts of untreated sewage discharge were evident at the outfall site by depleted $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ signatures and 4-fold increased BOD rates. Pollution effects of a sewage plume were detectable for more than 6 km downstream from the outfall site, as seasonal wind- and tide-driven shelf hydrodynamics facilitated its advective transport into near-shore waters. There, sewage pollution was detectable at recreational beaches by depleted stable isotope signatures and elevated BOD rates at high tides, suggesting high bacterial activity and increased infection risk by human pathogens. These findings indicate the urgent necessity for appropriate wastewater treatment in Salvador to achieve acceptable standards for released effluents and coastal zone water quality.

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1. Introduction

The discharge of municipal sewage into coastal waters represents a major cause of marine and estuarine pollution in many countries around the world (Rahaman and Varis, 2005; Lotze et al., 2006; Mara, 2013). In particular, the common dumping of sewage without primary treatment is of great concern, as these effluents not only contain high concentrations of suspended solids and nutrients, but often also carry substantial amounts of human organic waste products (e.g. feces) (Ramírez-Álvarez et al., 2007; Law et al., 2013). The discharge of untreated sewage alters the physico-chemical properties of coastal waters and may cause severe contamination of the marine environment (Teodoro et al., 2010; Lapointe et al., 2011), often characterized by high microbial loads, including human pathogens (Lyon et al., 2005; Despland et al., 2012; Wang et al., 2014). Visitors of recreational areas such as bathing beaches affected by sewage outfalls (i.e. swimmers and bathers) are at increased risk for various types of diseases and

infections, most commonly gastroenteritis or skin infections, posing a serious threat to human health in urbanized coastal regions (Griffin et al., 2001; Betancourt et al., 2014; Cheung et al., 2015).

Particularly in Latin America, the demographic explosion is absorbed by coastal or estuarine megacities (Cepal, 2000). With some of the highest coastal population densities in South America, most Brazilian cities are not equipped with facilities to collect, treat and dispose sewage in an environmentally sustainable manner (Abessa et al., 2005). In 2007, only 42% of all sewage in Brazil was collected, whereas only 32.5% of the collected volume was eventually treated (SINS, 2009). The common practice of draining raw sewage in the nearest body of water prevails (Salas, 2000), and in larger cities this is often implemented by means of a submarine sewage outfall. One of Brazil's largest submarine sewage outfalls belongs to the Oceanic Disposal System of Rio Vermelho (ODSRV) located off the coast of Salvador (Bahia), a city with ca. 2.7 million inhabitants (IBGE, 2010). The ODSRV was part of a state sanitation development program in the late 1970s, which involved the installation of a 2.35 km long concrete-steel pipeline with an inner diameter of 1.75 m. The Rio Vermelho outfall discharges $8.3 \text{ m}^3 \text{ s}^{-1}$ of pre-filtered (10–30% of suspended solids removed), but otherwise untreated (Feitosa, 2007; de Souza, 2011), municipal sewage into shallow waters (27 m depth) close to the inlet of Todos os Santos Bay. Nearly four decades after its first operation, Rio Vermelho still ranks among the largest submarine sewage outfalls in the world (Feitosa, 2007),

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and the extent of its plume can even be observed by open-access time series satellite imagery (Google Earth, 2015).

To date, information on potential impacts by the Rio Vermelho outfall on water quality in adjacent coastal environments is scarce and difficult to access. This study is the first to analytically address concerns raised by previous oceanographic and hydrographic studies that tidal currents may transport sewage-derived organic matter associated with high microbial loads into near-shore waters and close to popular recreational beaches (Cirano and Lessa, 2007; de Souza, 2011). These concerns are based on knowledge of shelf hydrodynamics around the sewage outfall site that are affected both by the local wind field and tides (Cirano and Lessa, 2007; Amorim et al., 2012). While southerly winds induce water flow to the northeast during winter, northeasterly winds cause southwestward coastal currents during summer. Close to the inlet of Todos os Santos Bay these coastal currents become highly modulated by rising tides (Cirano and Lessa, 2007), which during summer promote rapid westward water flows and a potential transport of discharged sewage into near-shore waters (Fig. 1). Under these conditions, water flow close to the outfall site may reach up to 0.5 m s^{-1}

(Cirano and Lessa, 2007). A recent numerical model has simulated shelf hydrodynamics around the Rio Vermelho outfall projecting the presumed transport of sewage, in particular fecal bacteria, from the outfall site towards the inlet of Todos os Santos Bay (de Souza, 2011). Although this model demonstrated that the Rio Vermelho sewage plume reaches the city beaches of Salvador, reliable model confirmation based on essential in situ water quality measurements is still lacking.

Therefore, this study investigated a set of water quality parameters, including suspended particulate organic carbon and nitrogen stable isotope signatures ($\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$), dissolved oxygen (DO) concentrations and biological oxygen demand (BOD) of seawater at potentially impacted sites within the coastal zone of Salvador (Chapman and World Health, 1996; Rožič et al., 2014; Mancinelli and Vizzini, 2015). Sampling was conducted on a long-shore transect across the Rio Vermelho sewage outfall towards the inlet of Todos os Santos Bay and aimed at answering the following research questions: (1) Are impacts of untreated sewage discharge detectable directly at the Rio Vermelho outfall site? (2) Are these impacts traceable along the coastal transect

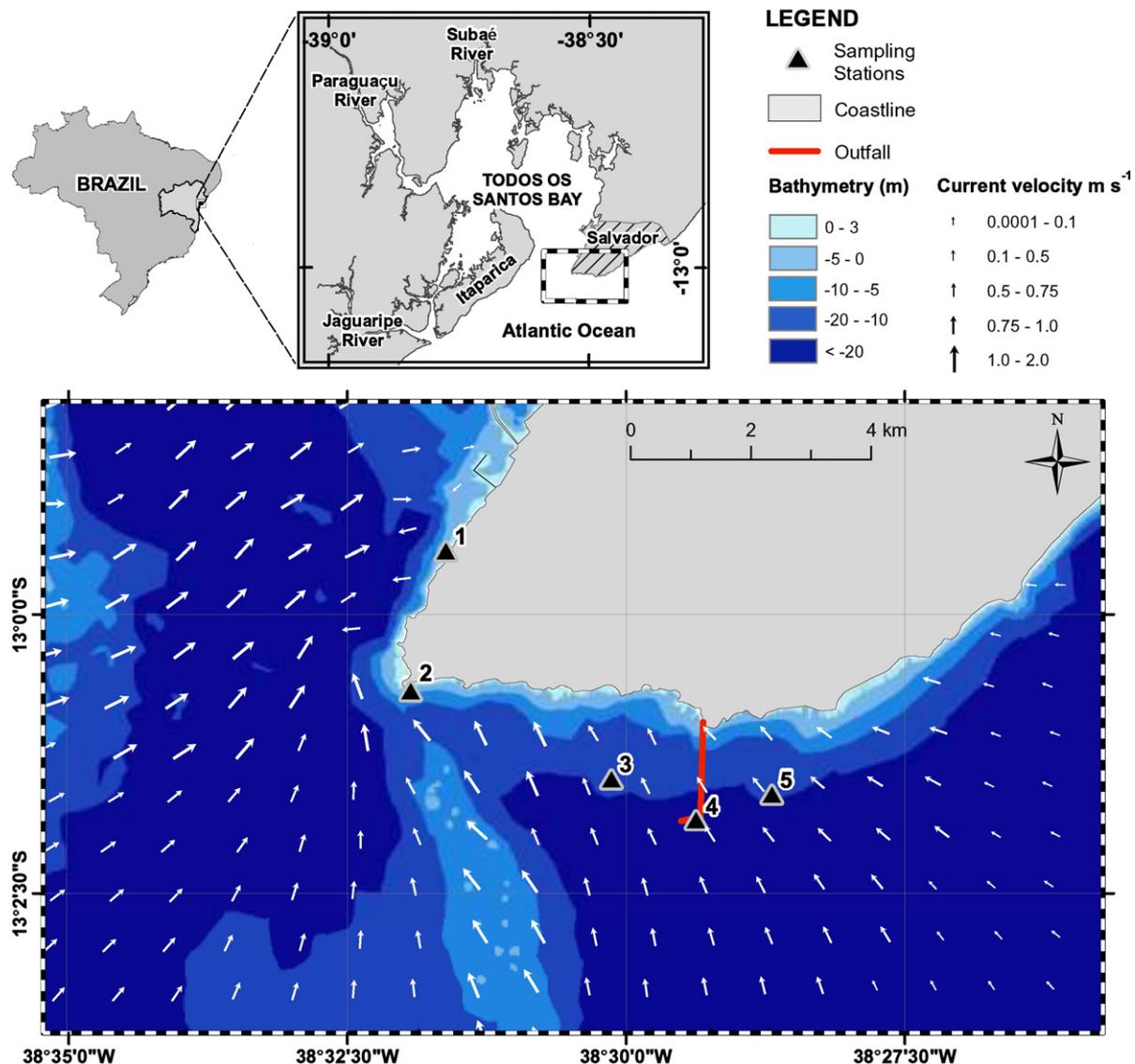


Fig. 1. Bathymetric map of the study site in the coastal zone of Salvador indicating the sampling stations, the Rio Vermelho sewage outfall site and the tidal current profile during high tide. Current vectors are derived from a numerical simulation using the Regional Ocean Modeling System (ROMS) (500 m grid resolution), projecting maximum flood currents during a typical summer spring tide. For the model simulation, temperature boundary conditions were obtained from a Moderate Resolution Imaging Spectroradiometer (MODIS) 2003–2012 sea surface temperature series, while water level, external currents and salinity boundary conditions were extracted from HYCOM (Hybrid Coordinate Ocean Model). Calculated averages encompassed the years 2003–2009 (HYCOM, 2011).

up to the city beaches of Salvador? (3) Which processes are controlling the transport of the discharged sewage?

2. Material and methods

2.1. Study sites and sampling design

This study was carried out throughout March 2015 (southern hemisphere summer) along the western shore of the Todos os Santos Bay inlet (Bahia State, Brazil) within the municipal boundaries of the city of Salvador. The study site experienced typical summer conditions throughout the sampling period with respect to wind direction and velocity data recorded at the Salvador Airport meteorological station (Fig. S1) (Cirano and Lessa, 2007).

A monitoring site (station 1; Fig. 1) was established at the pier of Hotel Sol Victoria Marina (12°59'26"S 38°31'37"W), a recreational area well-frequented by hotel guests and beach visitors. Close to the pier, a buoy was deployed and anchored to the sandy ocean floor (water depth: 10–15 m) and a data logger (Onset HOBO Pendant UA-002-64) was mounted at 5 m water depth to continuously (30 s interval) record water temperature. At station 1, sampling of surface seawater was conducted twice per week, whereby each sampling event comprised the sampling of incoming and outgoing tides during slack water (i.e. stagnant period between tides) conditions.

Four additional sampling sites (stations 2–5) were established along a coastal transect between the monitoring site (station 1) and a reference site (station 5; 13°01'37"S 38°28'41"W) (Fig. 1). Stations 2–4 were chosen to sample effluents in seawater directly above the Rio Vermelho outfall site (station 4; 13°01'51"S 38°29'22"W), and to track the high tide westward flow of the sewage plume towards Salvador's city beaches (stations 3 and 2; 13°01'29"S 38°30'08"W and 13°00'41"S 38°31'56"W). Station 5 located eastwards beyond the outfall site served as a reference site for oceanic water non-affected by the sewage plume (i.e. upstream) during high tide. All seawater samples of the coastal transect were taken on 20 March 2015 during high tide slack water in order to sample oceanic reference seawater at station 5 together with the identical sewage plume passing stations 4 through 1 (i.e. downstream). Additional information on sampling dates, time and tidal ranges are presented in Table S2.

2.2. Water quality parameters

Surface seawater samples ($n = 4$) were collected at each of the stations (1–5) at 5 m water depth using a 5 L Niskin water sampler. Temperature and salinity were measured immediately after sampling using a TetraCon 925 WTW sensor (accuracy $\pm 0.5\%$ of the measured value), before samples were transferred into pre-rinsed (sample seawater) high-density polyethylene canisters (5 L) and processed for subsequent analyses as described below.

BOD rates were determined by dark incubation of one seawater subsample from each canister ($n = 4$ per station). To this end, subsamples (50 mL) were transferred directly after sampling into clean Winkler glass bottles and dark incubations were started on site and kept at simulated in situ temperature for 12–16 h. DO concentrations ($\text{mg O}_2 \text{ L}^{-1}$) in the incubation medium were measured at the start and end of each incubation period using an optical FDO 925 WTW dissolved O_2 sensor (accuracy $\pm 1.5\%$ of the effective range). Measured DO concentration changes were used to calculate BOD rates normalized by the volume of incubated seawater and incubation period.

Subsamples for suspended particulate organic carbon and nitrogen stable isotope signatures ($^{13}\text{C}_{\text{org}}$ and ^{15}N) were filtered from each canister (1000 mL per parameter) onto pre-combusted GF/F filters (diameter: 25 mm, nominal particle retention: 0.7 μm) within 1 h after sampling. Filters were dried at 40 °C for 48 h and stored dry in Eppendorf vial. For $^{13}\text{C}_{\text{org}}$ analysis, filters were decalcified using fuming HCl, re-dried at 40 °C for 24 h and transferred into silver cups

pending analysis. Filters were analyzed for $^{13}\text{C}_{\text{org}}$ and ^{15}N signatures using a Thermo/Finnigan Flash 1112 elemental analyzer coupled to a Thermo/Finnigan Delta plus isotope ratio mass spectrometer. Repeated measurements of standard material (peptone; $\delta^{13}\text{C}$: $-22.0 \pm 0.1\%$, $\delta^{15}\text{N}$: $5.8 \pm 0.1\%$) revealed standard deviations of $<0.2\%$. C and N stable isotope ratios were expressed by the delta (δ) notation in units per mil (‰) and calculated as: $\delta^{13}\text{C}$ or $\delta^{15}\text{N} = (\text{R}_{\text{sample}} / \text{R}_{\text{ref}} - 1) \times 1000$, where R_{sample} is the ratio of $^{13}\text{C}/^{12}\text{C}$ or $^{15}\text{N}/^{14}\text{N}$ in the sample and R_{ref} is the heavy/light isotope ratio of the reference material (C: $\text{R}_{\text{ref}} = 0.01118$, Vienna Pee Dee Belemnite; N: $\text{R}_{\text{ref}} = 0.00368$, atmospheric N_2).

2.3. Statistical analyses

Statistical analyses were performed using SPSS (IBM) and SigmaPlot (Systat) software packages. Data were tested for normal distribution with probability plots (Q–Q-plot) and/or the Shapiro–Wilk-Test. Tests for equal variances were passed in all cases. Differences in $\delta^{13}\text{C}_{\text{org}}$, $\delta^{15}\text{N}$ and BOD were analyzed using 2-factorial analysis of variance (ANOVA) with *tide* (high and low tide) and *time* (8 sampling events) as fixed factors for the monitoring site (station 1). Correlation of tidal range and BOD at station 1 was assessed by linear regression analysis. Differences in water quality parameters between two individual stations were analyzed by t-tests, whereas more than two stations were compared using 2-factorial ANOVA with *distance* (stations 1–5) as fixed factor. Holm–Sidak tests were used for post-hoc pairwise comparisons.

3. Results

3.1. Physico-chemical parameters

At the monitoring site (station 1), surface seawater temperature ranged from 27.9–29.5 °C (at 6:00 a.m. and 1:00 p.m., respectively) throughout the study period, while salinity was stable at 37.5 ± 0.1 . Significant temperature fluctuations were only detectable with respect to time of day ($p = 0.02$), without any effect of tides. At the sewage outfall site (station 4), surface seawater temperature (27.1 ± 0.1 °C) and salinity (37.2 ± 0.0) were significantly decreased ($p = 0.004$ and $p = 0.02$, respectively) compared to reference station 5 (28.1 ± 0.2 °C and 37.5 ± 0.1). These temperature and salinity differences were already no longer detectable at station 3 (distance: 1.5 km downstream, Fig. 1). Values measured at stations 1–3 were again similar to reference station 5, ranging from 27.9–28.1 °C and 37.4–37.5 salinity units.

3.2. Biological oxygen demand

At the sewage outfall site (station 4), DO concentrations were significantly depleted compared to reference station 5 ($p < 0.001$), while station 4 BOD rates showed a 4-fold increase (Fig. 2, A). Only at larger distance to the sewage outfall (i.e. 6 km downstream, station 1), DO concentrations significantly increased again ($p = 0.01$) and BOD rates declined by 50% ($p = 0.004$). However, neither parameter showed a return to reference levels measured at station 5 (Fig. 2A). At the monitoring site (station 1), BOD rates doubled during high tide compared to low tide throughout the study period ($p < 0.001$), and were positively correlated ($r^2 = 0.81$, $p < 0.001$) to the day-specific tidal range (Fig. 3). Further, a significant 2-fold BOD increase compared to the overall mean BOD at station 1 ($p = 0.001$) was observed during a spring tide event on 18 and 19 March 2015 (Fig. 3).

3.3. Carbon and nitrogen stable isotopic signatures

Suspended particulate organic matter at the sewage outfall was significantly depleted both in $\delta^{13}\text{C}_{\text{org}}$ ($-22.8 \pm 0.3\%$) and $\delta^{15}\text{N}$ ($3.4 \pm 0.2\%$) compared to the upstream reference station 5 ($\delta^{13}\text{C}_{\text{org}}$: $-20.4 \pm$

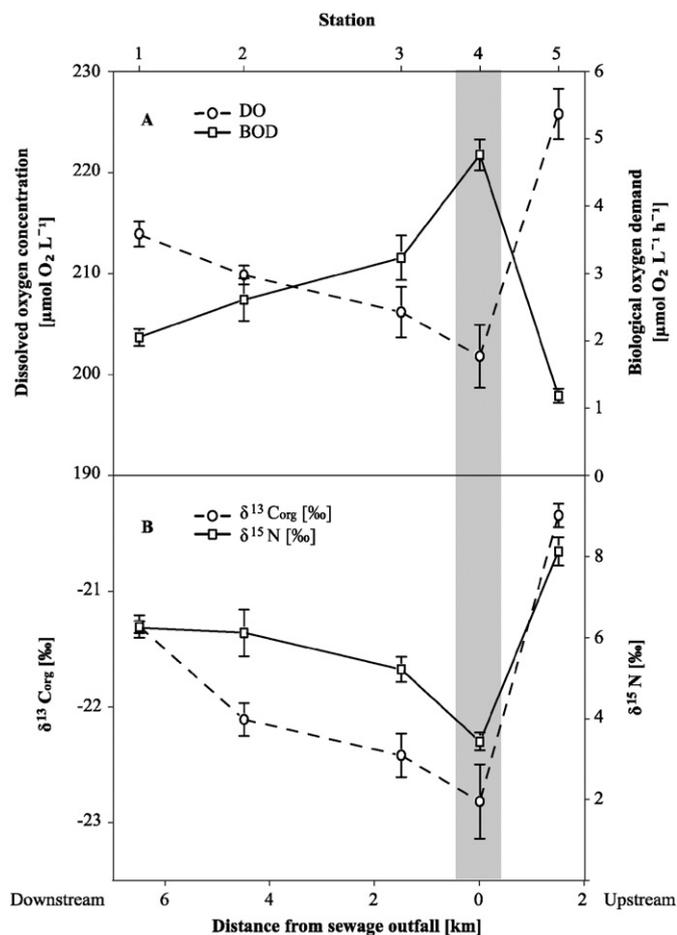


Fig. 2. Water quality parameters at the Rio Vermelho sewage outfall site and along the Salvador coastal transect. (A) Dissolved oxygen concentration and biological oxygen demand, (B) $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ stable isotope signatures of suspended particulate organic matter at stations 1–5 during high tide slack water on 20 March 2015. Values are given as mean \pm SEM ($n = 4$) and are presented relative to the distance from the sewage outfall (0 km, gray bar) in upstream (station 5) and downstream (stations 3–1, towards the entrance of Todos os Santos Bay) direction of the high tide currents. Abbreviations: DO = dissolved oxygen; BOD = biological oxygen demand.

0.1, $\delta^{15}\text{N}$: $8.1 \pm 0.4\%$) (both $p < 0.001$). At all downstream stations (3–1), $\delta^{13}\text{C}_{\text{org}}$ and $\delta^{15}\text{N}$ were significantly depleted compared to the upstream reference station 5 (Fig. 2B) (all $p < 0.05$). At the monitoring site (station 1), $\delta^{15}\text{N}$ was significantly depleted during high tide ($8.5 \pm 0.1\%$) compared to low tide ($6.9 \pm 0.13\%$; $p < 0.05$) throughout the study period,

while $\delta^{13}\text{C}_{\text{org}}$ remained constant (high tide: $-21.3 \pm 0.1\%$; low tide: $-21.2 \pm 0.2\%$). Further, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}_{\text{org}}$ values at station 1 were positively correlated to the day-specific tidal range ($\delta^{15}\text{N}$: $r^2 = 0.73$, $p = 0.04$; $\delta^{13}\text{C}_{\text{org}}$: $r^2 = 0.70$, $p = 0.05$).

4. Discussion

4.1. Impacts of untreated sewage discharge at the Rio Vermelho outfall site

Pollution impacts by untreated sewage discharge are clearly evident at the Rio Vermelho submarine sewage outfall off the coast of Salvador. Salinity differences measured between the sewage plume and surrounding seawater ($\Delta S = 0.3$) are similar to values reported for other sewage outfall sites and provide evidence for the substantial freshwater input by this discharge system (Petrenko et al., 1997; Ramos et al., 2007). More severely, our findings of $\delta^{15}\text{N}$ -depleted suspended particulate organic matter at the outfall site ($3.4 \pm 0.2\%$) are distinctive of particulate organic matter in untreated sewage, which is commonly depleted in $\delta^{15}\text{N}$ (0.0–3.5‰) relative to oceanic background levels (~5–8‰) (Rogers, 2003; Savage, 2005; Michener and Lajtha, 2008). In contrast, sewage-derived suspended particulate organic matter from wastewater treatment plants is usually $\delta^{15}\text{N}$ -enriched and has been shown to range from ~9‰ for secondary treatment (Jones et al., 2001) to ~16‰ for tertiary treatment (Piola et al., 2006), as a result of microbial isotope fractionation processes and postdepositional diagenesis (Heaton, 1986; Owens, 1987; Jordan et al., 1997). Likewise, our results for $\delta^{13}\text{C}_{\text{org}}$ -depleted suspended particulate organic matter at the outfall site ($-22.8 \pm 0.3\%$) are consistent with established literature values for untreated sewage sludge and wastewater effluents (-21.8 – 23.5%) (DeBruyn and Rasmussen, 2002; Nara et al., 2010; Law et al., 2013), thus providing further evidence for on-site pollution by untreated sewage discharge.

The Rio Vermelho sewage outfall site is further characterized by significantly depleted seawater DO concentrations ($201.80 \mu\text{mol O}_2 \text{ L}^{-1}$) and highly elevated BOD ($4.76 \pm 0.23 \mu\text{mol O}_2 \text{ L}^{-1} \text{ h}^{-1}$) compared to background levels at station 5 ($225.77 \mu\text{mol O}_2 \text{ L}^{-1}$ and $1.19 \pm 0.11 \mu\text{mol O}_2 \text{ L}^{-1} \text{ h}^{-1}$, respectively). This indicates high microbial activity and rapid decomposition of organic matter in local surface seawater (Rand et al., 1976; Hiraishi et al., 1989). Even considering dilution and aeration (i.e. by mixing with surrounding seawater) of discharged sewage while flowing upwards from the outfall (27 m water depth) towards the surface, our findings from 5 m water depth are still in the range of BOD reported for untreated and undiluted sewage effluents (3.9 – $26 \mu\text{mol O}_2 \text{ L}^{-1} \text{ h}^{-1}$) (Terrell and Perfetti, 1991; Petrenko et al., 1998; Hunt et al., 2010). Water with such high BOD rates ($> 4.16 \mu\text{mol O}_2 \text{ L}^{-1} \text{ h}^{-1}$) is generally categorized as being of very poor quality and highly polluted, and thus is regarded unacceptable for the

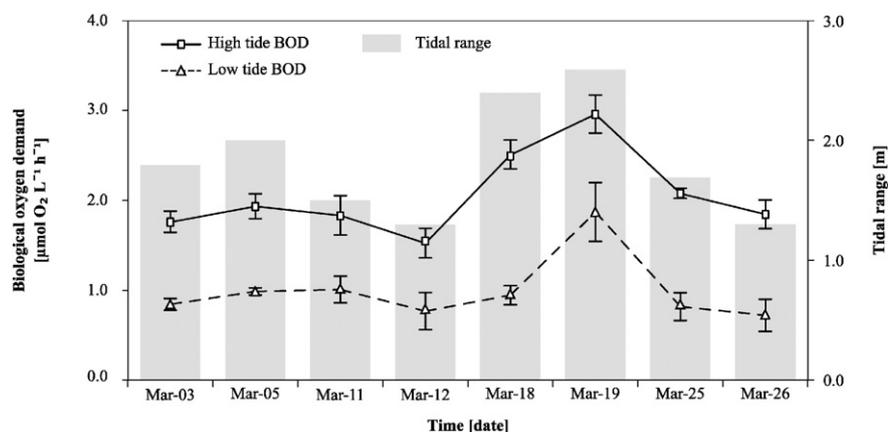


Fig. 3. Biological oxygen demand during high and low tides and the respective tidal range at the monitoring site (station 1). Values are given as mean \pm SEM ($n = 4$). Abbreviation: BOD = biological oxygen demand.

release into the environment (Chapman and World Health, 1996). This reveals the urgent necessity for appropriate wastewater treatment of municipal sewage collected in Salvador to establish controlled and sustainable quality standards for effluents before their release.

4.2. Advective transport of sewage into near-shore waters

Our findings provide first evidence based on in situ measurements that at high tides during summer untreated sewage discharged at the Rio Vermelho outfall is transported into near-shore waters of Salvador. In particular, our dual stable isotope approach proved as effective to identify and trace the distinct $\delta^{15}\text{N}$ and $\delta^{13}\text{C}_{\text{org}}$ isotopic signatures of suspended particulate organic matter in the sewage plume for more than 6 km downstream (i.e. northwestern direction) from the outfall site (Tucker et al., 1999; Gaston and Suthers, 2004; Rožič et al., 2014; Mancinelli and Vizzini, 2015). The sewage plume extends towards near-shore waters through directed advective transport that is forced by the local wind- and tide-driven shelf circulation during summer (Cirano and Lessa, 2007; Amorim et al., 2012). This process appears enhanced during high tide conditions, especially spring tides, with potential additive effects of seasonal shelf circulation and tidal currents (Cirano and Lessa, 2007; Amorim et al., 2012). This assumption is supported by our findings of a strong positive correlation of the day-specific tidal range with sewage plume stable isotope signatures, DO concentrations and BOD observed at station 1 throughout the study period. Moreover, 2-fold increased BOD rates during spring tide, when wind conditions are favoring shelf current acceleration, indicate that sewage transport is enhanced by increasing tidal range and rising NE wind speed. Depleted seawater DO concentrations and elevated BOD rates along the downstream section of the coastal transect provide further leads that effluents of the Rio Vermelho outfall effectively reach the inlet of Todos os Santos Bay and recreational city beaches of Salvador at high tides during summer.

Increases in BOD are generally associated with high microbial activity and abundance (Chapman and World Health, 1996). Thus, it appears likely that the Rio Vermelho sewage plume is also transporting fecal bacteria, as previously shown for untreated sewage plumes discharged at other global sites (Shuval, 2003; Stewart et al., 2008; Moynihan et al., 2012; Betancourt et al., 2014). Hydrological model visualizations of the tidal transport of coliform bacteria from the Rio Vermelho outfall site towards the shore-line project fecal coliform and total coliform cell counts to reach $>2.5 \times 10^4 \text{ L}^{-1}$ at the city beaches (de Souza, 2011). These projections exceed coliform cell counts ($>2.0 \times 10^4 \text{ L}^{-1}$) classified by the U.S. Public Health Service as seriously health-affecting (Cabelli, 1989), thus calling the health situation in near-shore waters and at Salvador's city beaches into question. Weekly microbiological monitoring by the Instituto do Meio Ambiente e Recursos Hídricos (INEMA) at the popular city beach Praia do Farol (here: station 2) indicates high abundances of *Escherichia coli* (cell counts $>2.0 \times 10^4 \text{ L}^{-1}$) for most sampling occasions (INEMA, 2015). Although *E. coli* is generally not regarded as harmful, its origin is exclusively human and/or animal feces, which suggests the potential co-presence of pathogenic bacteria, protozoans and viruses associated with human and animal digestive systems (EPA, 2012; INEMA, 2015). In support of this pathogenic co-presence, increased *E. coli* counts in recreational waters are often positively correlated to swimmers experiencing diarrhea, vomiting and skin rashes (Alexander et al., 1992; Wade et al., 2006; Colford Jr et al., 2007). Repeated findings of high *E. coli* abundances at Praia do Farol indicate that the Rio Vermelho sewage plume reaches near-shore waters, potentially increasing the risk for human bacterial infections. To date, reports of diseased beach visitors related to seawater pollution are still lacking and the number of potential past victims is unknown. In fact, the Salvador City council advises against bathing at most city beaches. However, updated information on local water quality conditions is currently only provided online (INEMA, 2015), and the actual usage of this service is unknown. Instead, warning signs informing visitors about the

local conditions and potential health risks from bathing are lacking at Salvador's recreational beaches, and thus general public awareness is low.

5. Conclusions

The here identified pollution impacts associated with the discharge of untreated sewage into coastal waters of Salvador may pose serious health threats to local residents and visitors bathing at its recreational beaches. As demonstrated, wind- and tide-driven southwestern currents promote the transport of sewage towards the shore at high tides during summer, when Salvador's beaches are most frequented. Whether seasonality in wind and tidal current directions, inducing northeastern flow during winter, may weaken pollution impacts in near-shore waters remains to be investigated (Lessa et al., 2001; Amorim et al., 2012). Establishing a continuous water quality monitoring during all seasons will thus provide essential information to evaluate the overall outfall impact. Parallel ecological assessments of local marine ecosystems may reveal further sewage pollution related impacts, as reported from other tropical regions (e.g. Lapointe et al., 2011; Moynihan et al., 2012).

In view of an estimated 40% increase in sewage-served population by 2027 (GEOHIDRO, 2015), the currently common practice of untreated sewage discharge will require critical re-evaluation and management by local authorities to identify feasible and sustainable approaches for effective improvement. In the first instance, wastewater treatment (i.e. primary and secondary) before release could critically improve the composition of Salvador's municipal sewage, reducing fecal bacteria by 85–99% (Dumontet et al., 1999; Paluszak et al., 2003), while lowering BOD by a factor of 10 (Terrell and Perfetti, 1991). These effects on water quality highlight only a few of the substantial benefits of installing appropriate sewage treatment facilities (reviewed in Balkema et al., 2002; Molinos-Senante et al., 2010) that may eventually establish and maintain acceptable and ecologically sustainable water quality standards in the coastal zone of Salvador.

Acknowledgments

We are grateful to the Institute of Geosciences at the Federal University of Bahia for logistical support. D. Dasbach, C. Staschock and D. Peterke (ZMT) are acknowledged for analytical support. RPKK is fellow (PQ-1D) of the National Council for the Scientific and Technological Development (CNPq). This work was funded by the European Commission 7th Framework Program Marie Curie Actions—People Grant PIRSES-GA-2011-295191 through the project SymbioCoRe (Synergies Through Merging Biological and Biogeochemical Expertise in Coral Research).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.marpolbul.2016.03.048>.

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