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Regional Acidification Trends in Florida Shellfish Estuaries: a 20+ Year Look at pH, Oxygen, Temperature, and Salinity

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Regional Acidification Trends in Florida Shellfish Estuaries: a 20+ Year Look at pH, Oxygen, Temperature, and Salinity

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Abstract
Increasing global CO_2 and local land use changes coupled with increased nutrient pollution are threatening estuaries worldwide. Local changes of estuarine chemistry have been documented, but regional associations and trends comparing multiple estuaries latitudinally have not been evaluated. Rapid climate change has impacted the annual and decadal chemical trends in estuaries, with local ecosystem processes enhancing or mitigating the responses. Here, we compare pH, dissolved oxygen, temperature, and salinity data from 10 Florida shellfish estuaries and hundreds of shellfish bed stations. Over 80,000 measurements, spanning from 1980 to 2008, taken on Atlantic Ocean and West Florida coast showed significant regional trends of consistent pH decreases in 8 out of the 10 estuaries, with an average rate of decrease on the Gulf of Mexico side estuaries of Florida of 7.3×10^{-4} pH units year^{-1}, and average decrease on the Atlantic Coast estuaries of 5.0×10^{-4} pH units year^{-1}. The rates are approximately 2–3.4 times slower than observed in pH decreases associated with ocean acidification in the Atlantic and Pacific. Other significant trends observed include decreasing dissolved oxygen in 9 out of the 10 estuaries, increasing salinity in 6 out of the 10, and temperature increases in 3 out of the 10 estuaries. The data provide a synoptic regional view of Florida estuary trends which reflect the complexity of changing climate and coastal ocean acidification superimposed on local conditions. These data provide context for understanding, and interpreting the past and predicting future of regional water quality health of shellfish and other organisms of commercial and ecological significance along Florida's coasts.

Keywords Shellfish estuary · pH · Sea surface temperature · Salinity · Dissolved oxygen · Ocean acidification

Introduction

The biogeochemical processes within estuaries are major drivers for the maintenance and resilience of plant and animal species within these types of ecosystems. Human-driven geochemical changes are being superimposed on this natural condition, forcing water quality variables (e.g., pH, temperature, salinity, dissolved oxygen) to new levels that are negatively impacting the natural biogeochemical balance within ecosystems worldwide (Doney et al. 2009; Cai et al. 2011). This growing human alteration of coastal zones presents increasing stressors in coastal waters such as excess nutrients, organic pollutants, and trace metals (Doney 2010; Sui et al. 2017). Estuaries throughout the USA and globally are undergoing rapid environmental change, including acidification (Feely et al. 2010; Waldbusser et al. 2013; Robins et al. 2016). Ocean acidification (OA), a direct consequence of rising atmospheric CO_2, has resulted in an average decrease of 0.08 pH units in ocean surface water between the 1700s and 1994 (Sabine et al. 2004; Jacobson 2005). Regional changes in ocean surface water pH vary about this average decrease value for a variety of reasons, including variations in biological uptake of CO_2, as well as differences in the buffering capacity that affect the overall response of the CO_2 system, to changes in total dissolved inorganic carbon and alkalinity (Egleston et al. 2010). Along coastal regions, additional factors can affect pH change, such as geochemical changes associated with

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land use changes, freshwater stream and coastal aquifer discharges, acidic precipitation, biological productivity and respiration rates, and hydrodynamic circulations (e.g., tides, coastal upwelling). Atmospheric inputs, such as rainout of NO_x sulfur oxide, pollutants, and dust, can also affect pH of coastal waters (Turk 1983; Prospero et al. 1987; Nickerson and Madsen 2005; Doney et al. 2007; Hagens et al. 2014). An additional negative impact of rising CO_2 in open ocean and coastal water is the concomitant decreases in dissolved oxygen concentrations (Gruber 2011; Zang et al. 2011). In coastal ecosystems, reduction of oxygen has been linked to agricultural run-off and increased nutrient loads, which in turn promote algal growth and subsequent oxidation of organic matter and release of CO_2 (Feely et al. 2010; Cai et al. 2011). Collectively, these stressors have put increasing negative pressures on the development and productivity of organisms and ecosystem services within impacted estuaries.

Responses of estuarine organisms to water chemistry changes due to OA and water quality have been the focus of many studies (Gazeau et al. 2007; Salisbury et al. 2008; Green et al. 2009; Miller et al. 2009; Talmage and Gobler 2009; Waldbusser et al. 2010; Feely et al. 2010; Cai et al. 2011). Shellfish, in particular, have received considerable attention because this group of marine organisms is a major component of commercial fishery industries and alterations in the geochemistry of the overlying water have been shown to negatively impact the quantity and quality of the shellfish (Dupont et al. 2014; Waldbusser et al. 2015a, b).

A recent analysis of pH, salinity, and temperature in the Chesapeake Bay from 1985 to 2008, for example, shows that surface water pH has increased in some areas and decreased in others, and these shifts were substantial enough to affect biocalcification and recruitment of the oyster *Crassostrea virginica* (Waldbusser et al. 2011). Although many marine organisms can adapt to changing water quality conditions, this response may be inadequate when exposed to multiparametric changes, such as simultaneous increases in temperatures and salinities or losses of habitats. For example, subtle alterations to salinity ranges in combination with temperature stress, and or increases and decreases in pH or DO can be lethal to oyster larvae (Arnold and Berrigan 2002; Hu et al. 2015; Velez et al. 2016).

Florida Shellfish Estuaries—Characteristics

Florida's coastline measures 2173 km along the Gulf and Atlantic coastal plains (Ning et al. 2003) and within that coastline, coastal geomorphic characteristics include barrier islands, wetlands, and embayments that provide a diversity of estuarine environments that support shellfish aquaculture. Common to all estuaries in Florida is the limestone platform they sit on; however, the sediment cover varies by latitude, with siliciclastics dominating on the west Florida shelf to the

north of the Tampa Bay estuary and carbonate sediment veneer to the south, with the demarcation of carbonate sediments dominating on the Atlantic coast being located at approximately Miami Beach, where longshore sediment of siliciclastic transport declines significantly (Dean and O'Brien 1987).

The higher energy narrow shelf on the east coast contrasts with the broad, shallow shelf and extensive coastal wetlands on the Gulf of Mexico. On both coasts, barrier islands and large embayments tend to have higher populations and development pressure which provide higher inputs of nutrients which promote eutrophication and impact the shellfish industry. Declining water quality and increasing salinities and surface water withdrawals in localized areas have occurred since the 1950s which have coincided with rapid urban, agriculture, industry, and recreational development (Marella 2004, 2008).

Long-term data on changes in water quality variables are critical in understanding the context associated with the responses of marine organisms. However, these types of data sets are not common for most estuaries, but when available, can provide critical insight into the chemical changes over time. These long-term data sets are valuable although may be imperfect because of the manner in which they were collected may not be acceptable by recent standards; there may be missing data; lack of georeferencing; undocumented changes in station location and/or name designations; changes in instrumentation used to collect the data. Here, we present the synthesis of approximately 80,000 records of pH, surface water temperature, salinity, and dissolved oxygen collected by Florida Department of Agriculture and Consumer Services (FDACS) from 10 estuaries in Florida associated with commercial shellfish harvesting for more than 20 years. These estuaries span wide latitudinal (~ 30 to 25.7°N) and water mass differences (Atlantic Ocean vs Gulf of Mexico) and include several geomorphic types (enclosed, semienclosed, behind a barrier island, open to shallow shelf, and semi-enclosed embayment).

Methods

Study Sites

The study region incorporates FDACS shellfish monitoring site data in 10 estuaries located on the Gulf of Mexico and Atlantic Ocean coasts of Florida (Fig. 1; Table 1). Gulf of Mexico sites include the estuaries St. Joseph, Wakulla, Cedar Key, Citrus County, Tampa Bay, and Ten Thousand Islands. The Atlantic Ocean estuaries include St. Johns North, South Banana River, Indian River, and Volusia. These estuaries represent a range of climatological, geographical, and environmental conditions (Table 1). Collectively, data were collected from 433 sample sites, including between 23
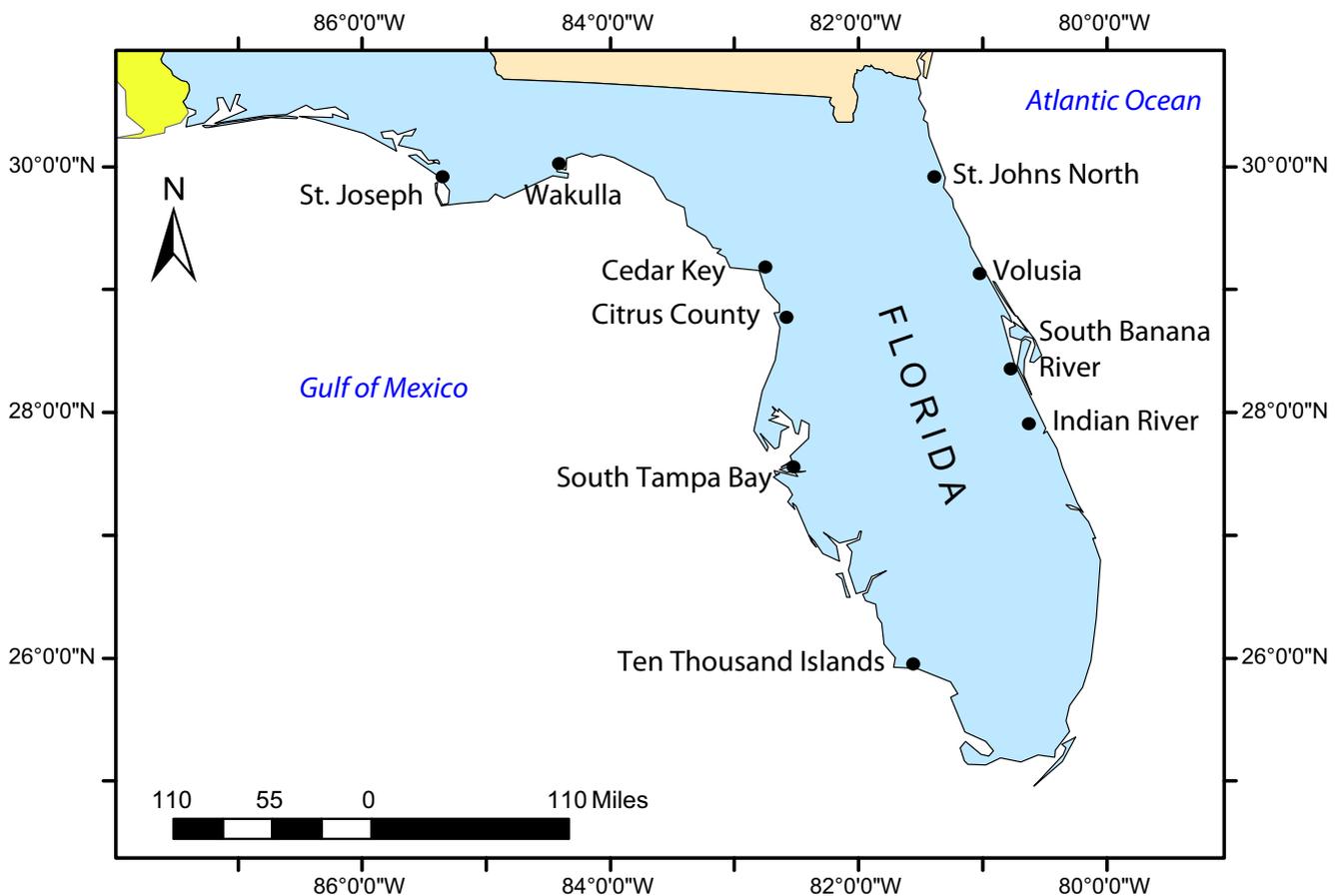
Fig. 1 Estuary locations in Florida from which water quality data were analyzed

and 75 sample sites per estuary and a total of 80,693 data points (Table 1). A brief description of each estuary follows:

St. Joseph estuary is a shallow, semi-enclosed lagoon bounded to the south and west by a barrier island system (Fig. 2a). The embayment supports extensive and diverse seagrass beds, shellfish beds (scallops), and fishing. Urban development is expanding although silviculture has been the prominent land use.

Wakulla estuary, a relatively open embayment, is located near spring-fed rivers and submerged groundwater seeps at the edge of Apalachicola Bay (Fig. 2b). Surrounding land is dominated by extensive urban and rural development with accompanying water quality issues such as increases in total phosphorus and nitrate and water supply issues (Hoyer et al. 2002). This estuary has historically been a commercial leader for oyster harvesting but is currently considered in decline due to alterations in freshwater discharge rates and saltwater balance (Arnold and Berrigan 2002).

St. Johns North estuary, located behind barrier islands, is a remnant of an ancient intercoastal lagoon system influenced by the St. Johns River that runs northward twice a day at high tide (Fig. 2c). The water quality is influenced by land use, which is primarily urban and silviculture, and engineered structures intended to divert and re-direct surface water within

the estuary. This estuary has been subjected to storm water and agricultural runoff and industrial wastewater discharge providing increased nutrient stress of phosphorus (Cooksey and Hyland 2007), leading to FDACS designating areas within this estuary as 'conditionally restricted' for shellfish harvesting.

Cedar Key is an open and shallow estuary that is impacted by the freshwater input from the Suwannee River, a black water river (Fig. 2d). The estuary is only occasionally subject to hypoxia (Timmerman and Chapman 2004). This estuary has the greatest clam harvest rates in the state of Florida. This estuary's watershed is primarily rural, dominated by agriculture and silviculture with a local dependence on shellfish harvesting and commercial fishing industries.

Citrus County estuary is an open estuary at the confluence of Chassahowitzka, Homosassa, Hall's and Withlacoochee rivers, freshwater springs, and groundwater seeps (Fig. 2e). This estuary has the lowest salinity values of all the estuaries in this study and supports seagrass beds, coastal marshes, manatee populations, shellfish harvesting, and commercial fishing. Recent and rapid increases in the local populations have led to increased nitrogen and phosphorus negatively affected estuarine and benthic habitats which support shellfish productivity (Hauxwell et al. 2007).

Table 1 Estuary characteristics

Estuary	Coast	Estuary classification	Geomorphological classification	Relative latitudinal classification	Period of record	Total data points	Sample stations per estuary	Total possible data points (as months)	Data points used in the final analysis (as months)
St. Joseph	Gulf of Mexico	Barrier island	Semi-closed	North	1985–2008	6620	23	300	136
Wakulla	Gulf of Mexico	Shallow shelf	Open	North	1982–2008	10,092	50	372	170
St. Johns North	Atlantic Ocean	Barrier island	Semi-closed	North	1982–2004	8690	46	360	191
Cedar Key	Gulf of Mexico	Shallow shelf	Open	North-Central	1985–2008	10,603	45	348	208
Citrus County	Gulf of Mexico	Shallow shelf	Open	North-Central	1984–2008	6767	36	348	187
Volusia	Atlantic Ocean	Barrier island	Semi-closed	North-Central	1981–2004	12,609	46	804	375
Tampa Bay	Gulf of Mexico	Embayment	Semi-closed	Central	1983–2008	6738	35	336	168
South Banana River	Atlantic Ocean	Barrier island	Semi-closed	Central	1992–2004	10,152	75	288	119
Indian River	Atlantic Ocean	Barrier island	Semi-closed	Central	1978–2004	4792	44	360	141
Ten Thousand Islands	Gulf of Mexico	Shallow shelf	Open	South	1983–2008	3630	33	556	138

Volusia estuary is a relatively closed estuary behind barrier islands which has a limited exchange with the Atlantic Ocean via a dredged canal (Fig. 2f). As part of the St. Johns River system, it is located on the northern end of the Indian River Lagoon. As with the other estuarine systems in Florida, land use changes from growing population and urbanization have physically altered the estuary and have caused accumulation of fine-grained, organic-rich clays and silt to be deposited into the estuary (Trefry et al. 2007) along with excess nitrogen and phosphorous (Trefry and Trocine 2002).

Tampa Bay estuary is the largest embayment on the Gulf of Mexico (Fig. 2g). The watershed is metropolitan with urban, industrial, and agricultural uses dominating the land use categories. This estuary has a history of poor water quality because of the excess nitrogen loads and inadequate water supply issues (Greening and Janicki 2006). Historically, the estuary supported extensive areas of seagrass meadows, and productive oyster and scallop beds which supported commercial harvesting (Arnold and Berrigan 2002). During the mid- to late-1900s, approximately 80% of the sea grass beds were lost due to declining water quality. Since the early 2000s, seagrass beds have been re-established and expanding as a result of stringent water quality regulations being promulgated and an aggressive seagrass re-planting and restoration program being implemented (Morrison and Greening 2011; Greening et al. 2011; Sherwood and Greening 2014).

The Indian River estuary is a shallow barrier island complex comprised of three lagoons—Mosquito Lagoon, Banana River, and Indian River Lagoon—with high rates of tidal exchange with the Atlantic Ocean (Phlips et al. 2010) (Fig. 2i). It spans two geographical (temperate and subtropical) provinces. The estuary is biologically diverse and includes shellfish (De Freese 1995; Boudreaux et al. 2006), but has been negatively impacted by increasing urbanization within the watershed, the placement of engineered structures and poor water quality, which led to decreases in shellfish harvests (De Freese 1995; Marelli and Arnold 1996).

South Banana River estuary is part of the Indian River Lagoon estuary system and is a relatively closed, shallow estuary behind barrier islands (Fig. 2h). The estuary receives substantial freshwater input from the Indian River, however, it has limited circulation with the Atlantic Ocean due to the restriction of a small inlet, which results in relatively long residence times (Phlips et al. 2010).

The Ten Thousand Islands estuary has a diverse ecosystem dominated by mangrove islands and numerous small tidal and inland creeks (Fig. 2j). Now this estuary supports commercial crab and shellfish harvesting activities. Although relatively low urban development rates have occurred in this watershed, the water quality in the estuary has been affected by large-scale agricultural run-off and the redirection of freshwater flow in the upstream reaches to the headwaters of the Everglades to augment surface water flows (Lindell 1973; Haman and Svendsen 2006).
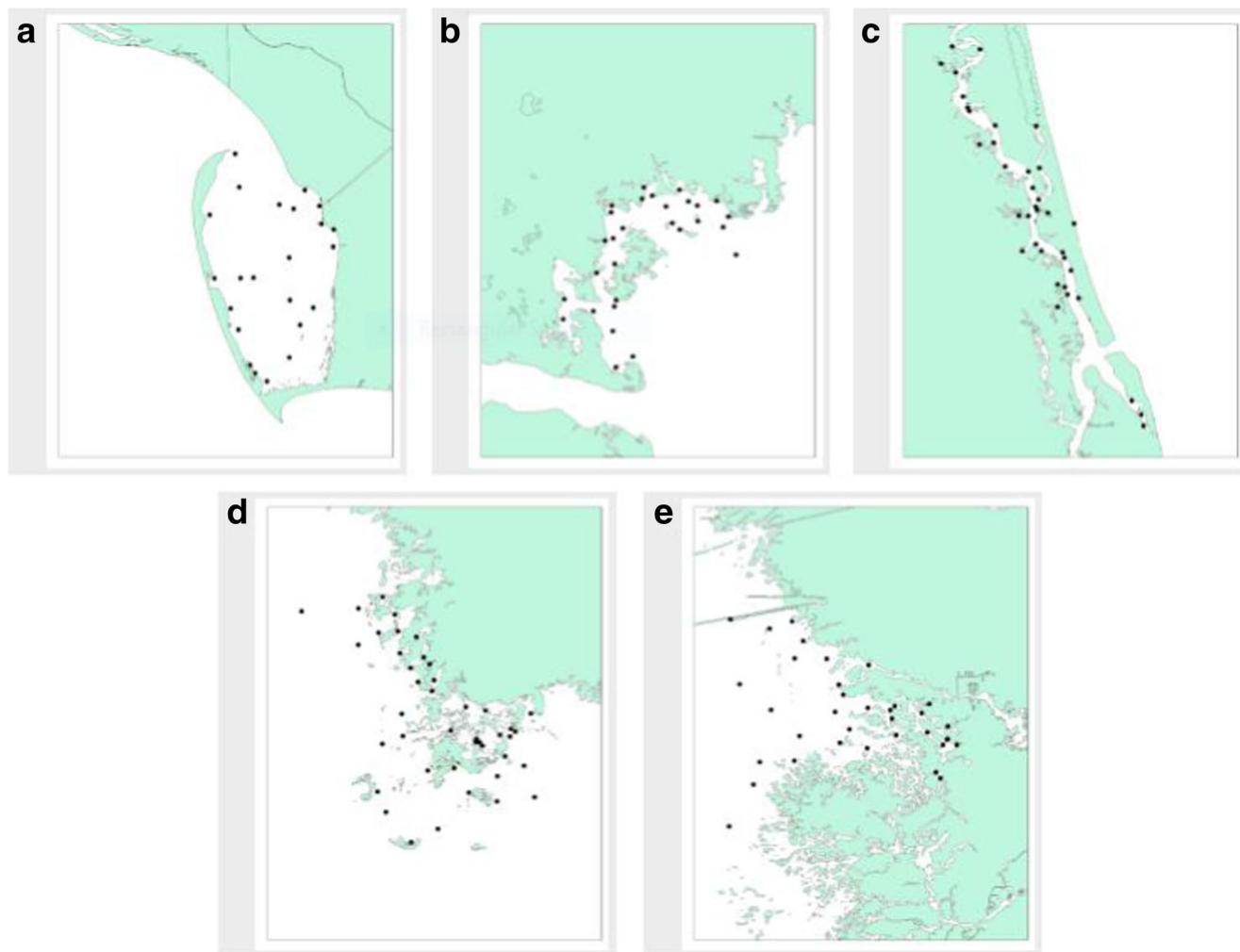
Fig. 2 Estuaries and respective sample sites in the study area. **a** St. Joseph estuary. **b** Wakulla estuary. **c** St. Johns North estuary. **d** Cedar Key estuary. **e** Citrus County estuary. **f** Volusia estuary. **g** Tampa Bay estuary. **h** South Banana River estuary. **i** Indian River estuary. **j** Ten Thousand Islands estuary

Data Sources

Surface water quality data [i.e., temperature (°C); salinity; dissolved oxygen (DO; mg L^{-1}); pH$_{NBS}$ (standard units)] from each of the estuaries were provided by the FDACS (http://www.freshfromflorida.com/Divisions-Offices/Aquaculture). Water samples were usually surface, collected from a median depth of 1.2 m below water surface over the time interval of this study. All data were collected by FDACS employees in accordance with standard methods and protocols, including calibrations used by the state, in FDACS and commercial laboratories (personal communication C. Brooks and J. Fleiger both at FDACS).

Data spanning over 30 years (1978–2008) were available for the estuaries, except for the South Banana River estuary which was monitored for only 14 years (Table 1). The sample collection frequency per estuary was generally monthly, though all sample sites within each estuary were not always sampled. Additionally, some sample sites were removed or

added during the period of record. Though the sample site identification numbers for each estuary were provided by FDACS, the latitude and longitude coordinates were not available for all sites within each estuary. All sites are seen in Fig. 2 as provided by FDACS.

Data Processing and Statistical Analysis

All water quality variable data sets were first assessed for outlier values. Based on typical value ranges for Florida, outlier data points were identified for seawater temperatures < 5 °C, pH values < 5.0 and > 9.5, DO values < 1.00 and > 10.00 mg L^{-1}, and salinity values < 3.00 and > 45.00 (Hand 2004). The resulting data set was then analyzed for the detection of statistical outliers using the Grubbs outlier test (α = 0.05). After the removal of the outlier values, the final data set had a total of 80,693 individual water quality data points from a total of 433 sample sites located in all estuaries (Fig. 1; Table 1). The data sets were then processed to provide a single
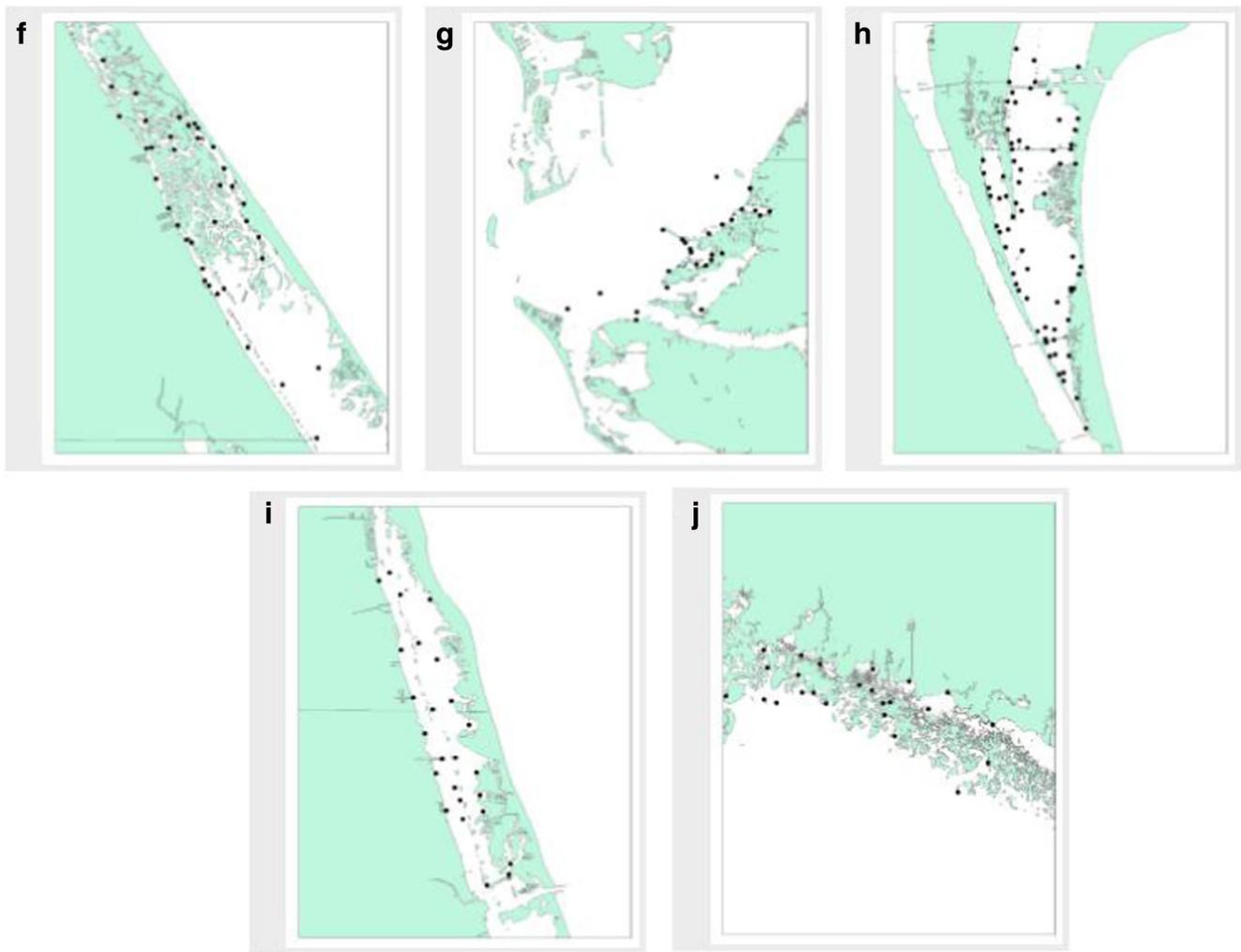
Fig. 2 (continued)

monthly average for each water quality variable within each estuary by taking the raw data values from all sample sites within that estuary (Table 1). The monthly average values were organized chronologically and coded for four seasons (Spring: March, April, May; Summer: June, July, August; Fall: September, October, November; Winter: December, January, February) within each calendar year. A decomposition step was applied to each variable's data set by first smoothing using a centered moving average with length equal to that of a coded season (i.e., 3). Using an additive model, the moving averages were subtracted from the monthly average values (raw seasonal values). Subsequently, the medians of the raw seasonal values within each seasonal cycle were calculated and then adjusted so their average value was equal to zero. These adjusted median values were then subtracted from the data to provide the seasonally detrended data on which the linear regressions were performed (Makridakis et al. 1998). The slopes of the respective regression models were first determined if they were independently and significantly different from zero (i.e., changes in the

respective water quality variable were significantly increasing or decreasing over time). The rates of change for the respective water quality variables (i.e., the slopes of the regression models) were compared to determine if there were significant differences between the estuaries based on their location on the Gulf of Mexico or Atlantic Ocean coasts and latitudinally (Table 1). These comparisons used the general linear model, where the water quality variables were the responses, the estuary locations per Table 1 were the factors and the covariate was the months of the data collection. The interaction term was set as (estuary location * months) and the resulting comparisons were evaluated using the Tukey post hoc multiple comparison test ($\alpha = 0.05$). The monthly data used in the regression analyses were also used to determine if there were significant differences between the water quality variables among the respective estuaries using one-way ANOVA and the Tukey post hoc multiple comparison test ($\alpha = 0.05$). Additionally, summary statistics were generated and correlations were determined between

selected water quality variables (Table 2). All statistical tests were performed using Minitab (ver 17) (Minitab, Inc., State College, PA).

Results

One of the operational hypotheses of this study was to determine if there were significant differences between the estuaries when comparing the (1) average water quality parameter data and (2) rates of change in those parameters, when grouped by latitudinal or coastal location (Table 1). The average monthly values for the 10 estuaries were DO (6.85 ± 0.50 mg L^{-1}), pH (8.05 ± 0.08), salinity (26.37 ± 5.66), and temperature (23.54 ± 1.48 °C) for all estuaries were relatively consistent, showing minimal mean differences between the estuaries at a state-wide level (Table 2). The ANOVA found no significant differences between any of the estuaries when water quality variables were compared, regardless of being grouped by latitude or coastal location. However, within this robust dataset the range of values for each water quality variable were relatively large (DO 1.11–10.00 mg L^{-1}; pH 5.37–9.11; salinity 3.27–44.93; temperature 7.04–41.53 °C), indicating there were significant chemical and biogeochemical heterogeneity within all the estuaries (Table 2).

Similar to the ANOVA results, there were no latitudinal or coastal relationships between the rates of change (i.e., slopes of the regression models) for any of the water quality variables in the respective estuaries. However, there were statistically significant increasing or decreasing rates of change for one or more of the water quality variables within each of the estuaries during the period of this study.

The pH values were shown to significantly decrease over time in 8 of the 10 estuaries, at an average rate of 6.63 × 10^{-4} pH units year^{-1} (Table 3). The least to greatest rates of change occurred in Citrus County (2.52 × 10^{-5} pH units year^{-1}) and Ten Thousand Islands (2.16 × 10^{-3} pH units year^{-1}) estuaries, respectively (Table 3).

There was significant decreasing DO values in 8 of the 10 estuaries at an average rate of 2.44 × 10^{-3} mg L^{-1} year^{-1} and ranged from 1.30 × 10^{-3} mg L^{-1} year^{-1} (Citrus County) to 5.56 × 10^{-3} mg L^{-1} year^{-1} (South Banana River). One estuary, Ten Thousand Islands, had a significant increase in DO (1.40 × 10^{-3} mg L^{-1} year^{-1}), while Tampa Bay data showed no significant trend in either direction (Table 3).

The majority of the estuaries had significant changes in salinity during the study period and averaged an increase in 8.73 × 10^{-3} year^{-1} (Table 3). Of the six estuaries showing changes, South Banana River (1.58 × 10^{-2} year^{-1}) and Citrus County (1.12 × 10^{-2} year^{-1}) had the greatest rates of increase. These rates of increasing salinity were approximately 1.4-fold to 3.8-fold greater than the rates in the other four estuaries

(Table 3), with Tampa Bay having the lowest rate of increase at 4.19 × 10^{-3} year^{-1}.

Within only four of the estuaries were the DO values significantly correlated with salinity values, three of those being negatively (Cedar Key, $r^2 = -0.221$; Citrus County, $r^2 = -0.340$; South Banana River, $r^2 = -0.297$) and one positively (Ten Thousand Islands, $r^2 = 0.337$) correlated (Table 4). The relationship between DO and pH was significantly and positively correlated in only two of the estuaries (Cedar Key, $r^2 = 0.326$; Citrus County, $r^2 = 0.277$) (Table 4).

Water temperatures significantly increased in three of the estuaries during the study period at an average rate of 2.80 × 10^{-3} °C year^{-1} (Table 3). The rates of increase were similar in Tampa Bay (3.18 × 10^{-3} °C year^{-1}) and Ten Thousand Islands (3.50 × 10^{-3} °C year^{-1}) which were approximately 2.0-fold greater than the rate of increase in South Banana River (1.68 × 10^{-3} °C year^{-1}). There were no significant decreases in water temperature in the estuaries of this study.

Discussion

Ocean acidification (OA) is the result of increasing atmospheric CO_2, which increases the rates of absorption into surface water. This increase in aqueous CO_2 decreases the pH of the water which subsequently alters the geochemistry, specifically carbonate chemistry (Doney 2010). Factors which directly influence the rates at which atmospheric CO_2 is absorbed, include temperature, salinity, and biological activities [i.e., CO_2 production (source) from respiration, CO_2 consumption (sink) from photosynthesis] (Doney et al. 2009; Miller et al. 2009; Koch et al. 2013; Dupont et al. 2014). However, how estuaries respond to increases in atmospheric CO_2 and other anthropogenic stressors compared to offshore waters is a reflection on their relatively shallower depths, higher rates of productivity in the water column and sediments (i.e., plants, animals, microbial), juxtaposition to terrestrial discharges of fresh water and coincident nutrient input, temperature fluctuations and being less buffered than marine waters (Waldbusser et al. 2011).

These OA-driven changes in an estuary's carbonate chemistry can exert significant negative pressures on the development and maturation of resident shellfish and other calcifying organisms (Miller et al. 2009; Pörtner 2010; Sui et al. 2017). The negative pressures can have significant economic consequences if the estuary supports commercial shellfish harvesting. All the estuaries in this study were active shellfish harvesting ecosystems during the time period of the data sets. Current sites contribute approximately 59% of the annual $257.7 million private and commercial seafood industry in Florida (http://www.freshfromflorida.com/Business-Services/Aquaculture).

Table 2 Estuary summary statistics for water quality variables based on monthly averages

Variable	Estuary	Mean ± SD	Minimum value	Maximum value
DO (mg L^{-1})	St. Joseph	7.47 ± 1.42	2.63	9.69
	Wakulla	7.63 ± 1.53	2.45	10.00
	St. Johns North	6.61 ± 1.40	2.46	9.95
	Cedar Key	7.22 ± 1.26	1.11	9.94
	Citrus County	7.21 ± 1.15	2.65	9.99
	Volusia	6.25 ± 1.78	1.68	9.93
	Tampa Bay	6.63 ± 0.87	3.31	8.99
	South Banana River	6.30 ± 1.29	2.77	9.22
	Indian River	6.73 ± 1.47	2.44	9.90
	Ten Thousand Islands	6.42 ± 1.04	3.72	8.28
pH (standard units)	St. Joseph	8.11 ± 0.35	6.35	8.72
	Wakulla	8.02 ± 0.34	6.38	8.90
	St. Johns North	7.89 ± 0.30	5.37	9.11
	Cedar Key	7.98 ± 0.16	7.45	8.39
	Citrus County	8.05 ± 0.18	7.58	8.63
	Volusia	8.04 ± 0.28	6.54	8.63
	Tampa Bay	8.09 ± 0.22	7.59	8.80
	South Banana River	8.17 ± 0.33	6.89	8.78
	Indian River	8.10 ± 0.29	6.44	8.76
	Ten Thousand Islands	8.03 ± 0.25	7.19	8.80
Salinity	St. Joseph	30.48 ± 4.31	12.56	37.13
	Wakulla	20.80 ± 4.63	5.13	31.67
	St. Johns North	31.38 ± 4.26	11.80	39.64
	Cedar Key	24.31 ± 4.00	10.26	32.78
	Citrus County	16.58 ± 4.24	3.27	27.80
	Volusia	31.62 ± 4.25	18.60	44.43
	Tampa Bay	30.05 ± 2.60	20.37	36.98
	South Banana River	19.25 ± 4.30	11.39	30.48
	Indian River	28.61 ± 5.43	12.67	43.97
	Ten Thousand Islands	30.65 ± 4.43	16.73	41.40
Temperature (°C)	St. Joseph	22.82 ± 6.15	9.99	31.75
	Wakulla	21.32 ± 5.82	9.64	31.82
	St. Johns North	21.83 ± 5.88	7.56	30.88
	Cedar Key	22.52 ± 6.08	8.53	32.47
	Citrus County	22.78 ± 5.54	7.12	32.38
	Volusia	23.86 ± 5.72	7.04	32.05
	Tampa Bay	24.93 ± 5.07	13.17	33.01
	South Banana River	25.04 ± 4.32	13.80	30.99
	Indian River	24.77 ± 4.65	8.94	41.53
	Ten Thousand Islands	25.55 ± 4.47	14.12	33.07

The complexity of the data for the respective variables within each of these estuaries demonstrates unique, local narratives, where intra-estuary influences exist juxtaposed to regional trends. Borges and Gypens (2010) showed through numerical modeling of nearshore coastal marine systems that eutrophication could counter the effect of rising CO_2 due to the input of nutrients of terrestrial origins. Additionally, the dissolution of carbonate sediments (Barnes and Cuff 2000; Alongi et al. 2006; Yates and Halley 2006; Cyronak et al. 2013; Green et al. 2013) and dissolving carbonate platform that underlies all of Florida, phytoplankton blooms, and the presence of healthy seagrass meadows (Garrard et al. 2014) have been shown to play an active role in ameliorating the effects of decreasing pH. This complexity, as exemplified by

Table 3 Yearly average rates of change (± SEM) for water quality variables based on the deseasonalized linear regression analyses of monthly averages multiplied by 12

Estuary	Dissolved oxygen		pH		Salinity		Temperature	
	Months[a]	$(mg\ L^{-1})\ year^{-1}$	Months	Standard units $year^{-1}$	Months	$year^{-1}$	Months	$°C\ year^{-1}$
St. Joseph	136	$-2.11 \times 10^{-3} \pm 5.76 \times 10^{-4}$ **	158	$-5.52 \times 10^{-4} \pm 1.44 \times 10^{-6}$ **	175	$8.29 \times 10^{-3} \pm 1.42 \times 10^{-3}$ **	181	$1.25 \times 10^{-3} \pm 7.02 \times 10^{-4}$
Wakulla	170	$-1.55 \times 10^{-3} \pm 5.88 \times 10^{-4}$ **	198	$8.04 \times 10^{-5} \pm 1.20 \times 10^{-6}$	254	$5.65 \times 10^{-3} \pm 1.05 \times 10^{-3}$ **	258	$8.40 \times 10^{-4} \pm 4.82 \times 10^{-4}$
St Johns North	191	$-1.76 \times 10^{-3} \pm 4.68 \times 10^{-4}$ **	233	$-8.88 \times 10^{-5} \pm 9.16 \times 10^{-5}$	220	$-9.84 \times 10^{-4} \pm 1.34 \times 10^{-3}$	245	$1.74 \times 10^{-3} \pm 6.95 \times 10^{-3}$ *
Cedar Key	208	$-2.28 \times 10^{-3} \pm 4.68 \times 10^{-4}$ **	221	$-3.36 \times 10^{-4} \pm 5.06 \times 10^{-5}$ **	259	$-4.32 \times 10^{-4} \pm 1.03 \times 10^{-3}$	258	$-2.88 \times 10^{-4} \pm 6.02 \times 10^{-4}$
Citrus County	187	$-1.30 \times 10^{-3} \pm 4.56 \times 10^{-4}$ **	176	$-2.52 \times 10^{-4} \pm 6.54 \times 10^{-5}$ **	226	$1.12 \times 10^{-2} \pm 8.76 \times 10^{-4}$ **	228	$1.20 \times 10^{-3} \pm 6.29 \times 10^{-4}$
Volusia	375	$-2.95 \times 10^{-3} \pm 5.04 \times 10^{-4}$ **	496	$-3.96 \times 10^{-4} \pm 6.58 \times 10^{-5}$ **	478	$-8.40 \times 10^{-4} \pm 1.05 \times 10^{-3}$	485	$1.58 \times 10^{-3} \pm 1.40 \times 10^{-3}$
Tampa Bay	168	$5.64 \times 10^{-4} \pm 3.96 \times 10^{-4}$	182	$-3.60 \times 10^{-4} \pm 8.84 \times 10^{-5}$ **	205	$4.19 \times 10^{-3} \pm 2.64 \times 10^{-4}$ **	210	$3.18 \times 10^{-3} \pm 8.47 \times 10^{-4}$ **
South Banana River	119	$-5.56 \times 10^{-3} \pm 8.40 \times 10^{-4}$ **	122	$-7.44 \times 10^{-4} \pm 2.34 \times 10^{-4}$ **	123	$1.58 \times 10^{-2} \pm 1.45 \times 10^{-3}$	123	$1.68 \times 10^{-3} \pm 1.26 \times 10^{-3}$
Indian River	141	$-2.10 \times 10^{-3} \pm 5.88 \times 10^{-4}$ **	164	$-3.10 \times 10^{-4} \pm 1.02 \times 10^{-4}$ *	178	$-1.20 \times 10^{-3} \pm 1.51 \times 10^{-3}$	173	$4.80 \times 10^{-4} \pm 9.50 \times 10^{-4}$
Ten Thousand Islands	138	$1.40 \times 10^{-3} \pm 5.4 \times 10^{-4}$ **	162	$-2.16 \times 10^{-3} \pm 1.10 \times 10^{-4}$ **	172	$7.24 \times 10^{-3} \pm 12.6 \times 10^{-3}$ **	185	$3.50 \times 10^{-3} \pm 1.45 \times 10^{-3}$ *

[a] Months = total number of monthly averages included in the regression analyses during the period of record as listed in Table 1

$*p < 0.5; **p < 0.01$

Table 4 Correlation coefficients between monthly dissolved oxygen and salinity or pH

Estuary	Dissolved oxygen vs.	
	salinity	pH
St. Joseph	-0.043	0.063
Wakulla	-0.037	0.056
St. Johns North	0.046	0.043
Cedar Key	-0.221**	0.326**
Citrus County	-0.340**	0.277**
Volusia	0.016	0.112
Tampa Bay	-0.071	-0.057
Banana River	-0.297**	0.140
Indian River	-0.094	0.089
Ten Thousand Islands	0.337**	0.051

$*p < 0.05; **p < 0.01$

the wide range of values associated with average water quality variable value (Table 2) and lack of a consistent ecological response within all the estuaries, may be a contributing factor as to why no statistically significant differences were found between the estuaries, regardless of the water quality variable being compared and the location of the estuary. However, statistically significant rates of change for pH, salinity, temperature, and DO in select estuaries were detected over the time period of this study. Collectively, this suggests the physical and geochemical processes which are driving these significant rates of change are operating on a regional-to-continental spatial scale in which Florida and all its estuaries are equally affected.

Of the four variables, pH demonstrates the most consistent and significant changes, and significant decreases observed in 8 of the 10 estuaries. The trend in decreasing pH is greatest on the Gulf Coast estuaries ($n = 5$) with an average decrease of 7.3×10^{-4} pH units $year^{-1}$, while the average decrease in the Atlantic Coast estuaries ($n = 3$) is 5.0×10^{-4} pH units $year^{-1}$. These average rates of change in pH though significant, range from approximately 2–3.4 fold slower than that recorded in offshore waters [i.e., Atlantic 1.5×10^{-3} pH units $year^{-1}$ (Ríos et al. 2015) and Pacific 1.7×10^{-3} pH units $year^{-1}$ (Byrne et al. 2010)].

Assuming that the 10 Florida shellfish estuaries were subjected to similar atmospheric CO_2 conditions and therefore was consistent regionally, increases in CO_2 alone can account for the regional decreases in pH. However, since the decreases are less than open ocean, clearly local estuarine chemical and biogeochemical processes have controlled the rate of decline. Major processes that may have mitigated the effects of increasing CO_2 include regional dissolution of the Florida carbonate platform that underlays all of the estuaries as well as eutrophication in which phytoplankton blooms are enhanced (Aufdenkampe et al. 2011). Decreasing pH in the estuaries has

implications for the dissolution rate of the coastal carbonate Floridan Platform, with a limestone loss rate of 1.3–17.8 cm year^{-1} from north to south (Willett 2005). Further declines in pH will increase dissolution and accelerate carbonate platform removal and aid in the buffering of the estuarine waters. The significant positive correlation between pH with DO in only two of the estuaries not only suggests that increasing eutrophication has exacerbated these trends (Green et al. 2009; Zang et al. 2011) but also suggests that these parameters are decoupled in most of the estuaries. At present, there are a total of 425 waterbodies, including lakes, rivers, and water resources in Florida classified as nutrient impaired; approximately 27% of those are estuaries and coastal waters and 33% are surface water streams (Badruzzaman et al. 2012).

A note of caution is necessary for interpreting the pH data in this study due to the authors not knowing which type of pH electrode was used to collect these data. The magnitude and variability of carbonate chemistry such as pH in the coastal ocean are poorly constrained and often demonstrate extremes not observed in open ocean settings (Feely et al. 2008; Challener et al. 2016). There is a consensus opinion that pH measurements in estuarine waters with commonly used glass potentiometric electrodes can be problematic due to improper calibration with buffers optimized to the ionic strength of the estuarine water (Millero 1986; Department of Energy 1994; Waldbusser et al. 2011; Duarte et al. 2013). While pH electrodes calibrated with commonly supplied National Institute of Standards and Technology (NIST) pH buffers, which are then used on high ionic strength waters, they can potentially lead to drift and errors in measurements. As a result of such errors, recommendations to accurately measure pH in marine systems encourage use of total seawater scale and color indicators as standard operating procedure (Clayton and Byrne 1993). Unfortunately, estuarine systems, with mixtures of fresh to marine waters can be problematic in pH measurements. Therefore, systematic bias over the time scale of these older data sets cannot be ruled out.

While 9 of 10 estuaries show increases in surface water temperatures, only 3 of the 10 estuaries are experiencing significant increases. Maul and Sims (2007) proposed there was no geographic relationship with coastal surface water temperatures around Florida. Our analyses of temperature of the shellfish estuaries corroborate with this finding; no clear latitudinal trends were observed. However, Maul and Sims (2007) recognized that the temperature of Florida's coastal waters have increased at a rate of 0.2 to 0.4 °C century^{-1} when they included air temperatures in their model. Similarly, when the average water temperature increase rate from the three estuaries in this study is expressed on the same time scale, the rates corroborate (0.28 °C century^{-1}).

Amos et al. (2013) have also shown the surface water temperatures along coastal zones are increasing at rates an order of magnitude greater than the average global trend of 0.13 °C decade^{-1}, which is derived from open ocean measurements (IPCC 2007). They suggest these higher rates of change are due to coastal developments, resulting in increased net temperatures in rivers and lakes which discharge into coastal waters, and the radiative heating of the relatively shallower coastal waters. The three estuaries which showed significant increases in their surface water temperatures are impacted by discharges from major rivers whose watersheds are extensively urbanized (St. Johns North and Tampa Bay) (Hendrickson and Konwinski 1998; Greening et al. 2014) or have extensive engineered barriers and canal systems which hold and divert water into the coastal zone (Ten Thousand Islands) (Krauss et al. 2011).

Net estuarine salinity is most influenced at short (e.g., precipitation events, tidal flushing) and long temporal periods by the hydrology in the associated watershed. Long-term decreases in salinity can be driven by increased precipitation rates due to changing global weather patterns which increase the discharge rates of streams and lakes into estuaries (Stocker et al. 2013). An additional freshwater input, which is significant in Florida, is submarine groundwater discharge (SGD). Studies on these freshwater groundwater systems have shown discharge rates ranging from 0.23 to 5.3 m^3 m^{-2} year^{-1}, which is similar to smaller river and first magnitude spring discharge rates (Cable et al. 1997; Santos et al. 2008).

Six of the estuaries demonstrated significant increases in salinity over the time interval of this study, at an average rate of 8.7 × 10^{-3} year^{-1}, while none of the remaining estuaries showed significant decreases or freshening. Increases in estuary salinity are most often driven by reductions in the quantity of freshwater discharge, which can result from increased demand, reduction in the number and/or intensity of precipitation events, and evapotranspiration (Flemer and Champ 2006). An additional forcing of increasing salinities in estuaries is sea level rise (SLR). Satellite data from 1993 to 2003 shows an average SLR rate of 3.1 mm year^{-1} (Cazenave and Nerem 2004), which is approximately double the average rate (1.7 mm year^{-1}) during the early to mid-twentieth century (Bates et al. 2008). The pre-2006 SLR rates for Florida have been shown to average approximately 3.0 mm year^{-1}, but increasing to approximately 9.0 mm year^{-1} from 2006 to present (Wdowinski et al. 2016). The increase in sea level forces sea water with higher salinities into the estuaries for longer periods of time. Additionally, the higher sea level exerts increased hydraulic pressures on SGD processes and river and lake discharges, collectively reducing the respective discharge rates of fresh water into the estuaries (Passeri et al. 2015).

Dissolved oxygen concentrations are negatively affected by increasing temperatures and increasing salinities shown in the respective data sets in this study. Nine of the estuaries exhibit significant reductions in DO over the time period of this study at an average rate of 2.4 × 10^{-3} mg L^{-1} year^{-1}. In addition to the physical constraints on the saturation capacity

of DO in estuarine water (i.e., temperature and salinity), biogeochemical processes likely play substantial roles. The demand for DO increases with the addition of nutrients and simple and complex carbon compounds (e.g., sewage, agricultural runoff, urban runoff, etc.) (Rabalais et al. 2002; Smith 2003; Turner and Rabalais 2003). All of the estuary watersheds in this study are combinations of agricultural and urban development and carry elevated concentrations of most nutrients and carbon sources shown to promote eutrophication (McPherson et al. 2000; Badruzzaman et al. 2012).

Analyses of long-term datasets provide a way to detect changes in water quality variables which may be too small to detect at shorter temporal scales, thereby opening a "window to the past" and assisting in predicting environmental trends. As valuable as these datasets are in revealing long-term trends, their qualities are often imperfect. Nevertheless, the datasets presented here provide an important 26-year, synoptic survey of how Floridan estuaries on the Gulf of Mexico and Atlantic Ocean coasts are responding to climate change and anthropogenic stressors. Declining pH and oxygen trends associated with OA in estuary water quality are concerning because of their potential impacts on fisheries and aquatic resources which are economically important in Florida. However, robust data sets such as these suggest that local-to-regional processes are partially ameliorating the pH decreases associated with ocean acidification.

Compliance with Ethical Standards

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