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Megacity development and the demise of coastal coral communities: Evidence from coral skeleton $\delta^{15}N$ records in the Pearl River estuary

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Abstract

Historical coral skeleton (CS) $\delta^{18}O$ and $\delta^{15}N$ records were produced from samples recovered from sedimentary deposits, held in natural history museum collections, and cored into modern coral heads. These records were used to assess the influence of global warming and regional eutrophication, respectively, on the decline of coastal coral communities following the development of the Pearl River Delta (PRD) megacity, China. We find that, until 2007, ocean warming was not a major threat to coral communities in the Pearl River estuary; instead, nitrogen (N) inputs dominated impacts. The high but stable CS-$\delta^{15}N$ values (9‰–12‰ vs. air) observed from the mid-Holocene until 1980 indicate that soil and stream denitrification reduced and modulated the hydrologic inputs of N, blunting the rise in coastal N sources during the early phase of the Pearl River estuary urbanization. However, an unprecedented CS-$\delta^{15}N$ peak was observed from 1987 to 1993 (>13‰ vs. air), concomitant to an increase of NH_4^+ concentration, consistent with the rapid Pearl River estuary urbanization as the main cause for this eutrophication event. We suggest that widespread discharge of domestic sewage entered directly into the estuary, preventing removal by natural denitrification hotspots. We argue that this event caused the dramatic decline of the Pearl River estuary coral communities reported from 1980 to 2000. Subsequently, the coral record shows that the implementation of improved wastewater management policies succeeded in bringing down both CS-$\delta^{15}N$ and NH_4^+ concentrations in the early 2000s. This study points to the potential importance of eutrophication over ocean warming in coral decline along urbanized coastlines and in particular in the vicinity of megacities.

KEYWORDS

corals, eutrophication, megacities, stable nitrogen isotopes, urbanization

1 | INTRODUCTION

In the second half of the 20th century, humanity entered a phase of unprecedented urbanization. Today, more than 50% of the global population lives in a city (Grimm et al., 2008), and urban population is expected to rise to 80% by 2050 (Bettencourt & West, 2010). The world's urbanization does not spread homogeneously over the globe, but is concentrated mainly within coastal areas (Tibbetts, 2002). Coastlines host 45% of the global population, with an average population density over a 100 ind.km^{-2}, which is >2.5 times the mean global density. The world's highest population densities are found in immense urbanized areas hosting >20 million people, which result from the clustering of adjacent cities. These urban complexes are often referred to as "megacities" (Tibbetts, 2002). The world currently hosts 12 megacities, 10 of them located in coastal areas (Figure 1a). As such, the environmental footprint of humankind is disproportionately high within coastal areas, leading to concerns over the fate of coastal marine habitats.

Urbanization, and the development of megacities in particular, strongly alters the terrestrial to marine *continuum* at local, regional, and global scales (Fang et al., 2018; Folke, Jansson, Larsson, & Costanza, 1997; Lenzen, Moran, Suh, & West, 2013; Warren-Rhodes & Koenig, 2001). Such transformations impact freshwater and marine ecosystems worldwide, mainly by the release of pollutants and nutrients (Booth, Roy, Smith, & Capps, 2016; Deegan et al., 2012; Valiela & Martinetto, 2007; Wear & Thurber, 2015). Excess nutrients, nitrogen (N) in particular, have been identified as a mounting global threat to freshwater bodies (Booth et al., 2016; Walsh et al., 2005) and coastal areas (Boesch, 2002; Howarth & Marino, 2006). The ocean is a common sink for anthropogenic N sources (sewage, artificial fertilizers, NOx), with the transfer occurring either indirectly (through atmospheric deposition, groundwater, or river discharge) or directly (by sewage outfall). Excess N is detrimental to the marine ecosystems and it has been linked to a myriad of cascading effects: anoxic zones formation (Rabalais, Turner, & Wiseman, 2002), harmful algal blooms (NRC, 2000), and reduction in biodiversity (Fabricius, De'ath, McCook, Turak, & Williams, 2005).

The world's coastlines host sensitive and diverse ecosystems, such as salt marshes, mangroves, forests, and coral reefs, that provide

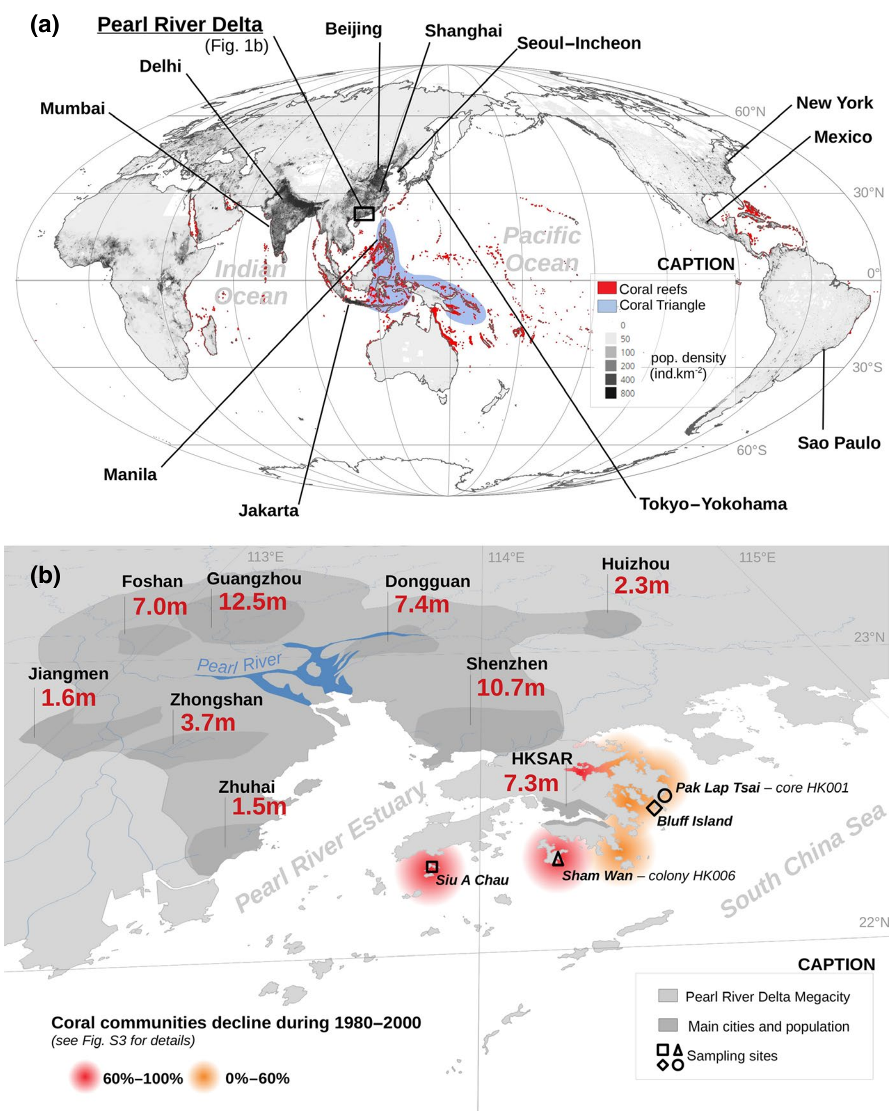

FIGURE 1 The urban development on the world's coastlines is a rising threat for coral reefs. (a) The map shows the population density (source: CIESIN, 2017), the world's megacities (source: UN, 2018), coral reefs (source: UNEP-WCMC, WorldFish Centre, WRI, TNC, 2010), and the world's hotspot of coral biodiversity, the Coral Triangle (Veron et al., 2011). (b) Detail map of the Pearl River Delta region showing the eight major cities composing the megacity and the Hong Kong Special Administrative Region (HKSAR) and their populations. The four sites sampled for this study are indicated with outlined symbols. The coral communities having experienced a major decline are highlighted in color. Maps were made with the geographic information software QGIS (www.qgis.com)

critical services. Coral reefs in particular, provide a wealth of environmental services that sustain millions of people throughout the world: food (Hughes et al., 2012), income (Spalding et al., 2017), coastal protection (Ferrario et al., 2014), biodiversity (Knowlton et al., 2010), and medicinal potential (Bruckner, 2002). Coral reefs are vulnerable to rising temperatures (Hughes et al., 2018), overfishing (Maire et al., 2016; Newman, Paredes, Sala, & Jackson, 2006), and eutrophication (Duprey, Yasuhara, & Baker, 2016). In particular, coastal urbanization is now exerting a strong pressure on coral reefs near two megacities located at the edge (Jakarta, the Pearl River Delta [PRD]) and within (Manila) the Coral Triangle, the world's coral biodiversity hotspot (Figure 1a). Information is desperately needed on the role of coastal urbanization on the N budget of coral reef ecosystems.

Evaluating the respective impacts of global change (i.e., ocean warming and/or acidification) and regional processes (i.e., eutrophication) on coral reef ecosystems can be challenging. Satellites and underwater loggers are continuously monitoring the world ocean's temperature while coral geochemical records (e.g., stable oxygen isotopes $\delta^{18}O$) extend these records over the last 300 years (Hennekam et al., 2018; Zinke, Loveday, Reason, Dullo, & Kroon, 2014). Yet, the historical and current pressure of eutrophication on coral reefs is much more difficult to assess. Indeed, the lack of reliable environmental baselines and the challenge of identifying the anthropogenic N sources responsible for coastal eutrophication hamper our efforts to reduce anthropogenic impacts on marine ecosystems by designing and implementing effective N mitigation policies. Coastal areas with a long history of urbanization provide key case studies on the history of anthropogenic N footprint on the marine environment. The PRD (Guangdong province, China) has been experiencing intense urbanization for at least the last 200 years, with a population of 1.5–2 million reported in the city of Guangzhou at the turn of the 19th century (Benedict, 1988; Gauducheau, 1911). In the 1980s, the start of the PRD Economic Zone resulted in the clustering of eight major cities in the delta (Guangzhou, Foshan, Dongguan, Jiangmen, Zhongshan, Zhuhai, Shenzhen, and Huizhou) into one of the most populated megacities, gathering ~120 million inhabitants (Figure 1b). Today, the Pearl River estuary is notorious for its severely eutrophic waters with DIN concentrations comprised between 2 µM and above 50 µM toward the rivermouth (Ye, Ni, Xie, Wei, & Jia, 2015), whereas typical DIN concentrations on coral reefs are generally less than 1 µM (Bell, Elmetri, & Lapointe, 2014). This eutrophy is responsible for coral biodiversity loss (Duprey et al., 2016) and harmful algal blooms that have severely affected the fish aquaculture economy in the area (Yin, Harrison, Chen, Huang, & Qian, 1999). Paradoxically, the coastlines surrounding the Pearl River estuary are still home to coral communities hosting about 90 scleractinian coral species (Duprey et al., 2016; Veron et al., 2011), including the long-lived genus *Porites* (Goodkin et al., 2011), offering the opportunity to investigate the link between the evolution of the N footprint of the PRD megacity and the decline of coral communities over time.

The nitrogen isotopic composition of hard coral's skeletal organic matrix (CS-$\delta^{15}N$, where $\delta^{15}N = ([(^{15}N/^{14}N)_{sample}/(^{15}N/^{14}N)_{air}] - 1) \times 1{,}000$) has been shown to reflect the isotopic composition of the nitrogen supply to coral reef (Wang et al., 2016). Long-lived hard coral, such as the genus *Porites*, can thus be used to track changes in natural and anthropogenic N sources over time (Duprey, Wang, et al., 2017; Ren, Chen, et al., 2017; Wang et al., 2018) . Here, we report CS-$\delta^{15}N$ records spanning the last 5,000 years from coral fragments recovered from sedimentary deposits, museum-held coral specimens, and coral cores. The objectives of this study are (a) to produce an environmental baseline by identifying the natural sources/processes regulating the N cycle in the Pearl River estuary before the PRD megacity started to develop, (b) to document the evolution of the N footprint of the Pearl River over the last 5,000 years, and (c) to compare this evolution to the decline of coral communities located in the Pearl River estuary.

2 | MATERIALS AND METHODS

2.1 | Hard coral material

All the material used in this study was collected on the eastern edge of the Pearl River estuary, within the Hong Kong Special Administrative Region—HKSAR (Figure 1b). Local coral communities are typically found at depths less than 10 m (Thompson & Cope, 1980). For the present study, only scleractinian (i.e., photosymbiotic) genera were selected, to ensure that the skeletal geochemistry would record the upper part of the water column (i.e., <10 m) and that isotopic data from different corals were comparable (Wang et al., 2014, 2016). Two specimens of the massive coral *Porites* (*references HK001 and HK006—The University of Hong Kong coral cores repository*) were selected for this study. Both samples were collected along undeveloped coastlines (*Pak Lap Tsai* for HK001—circle symbol on Figure 1b, and *Sham Wan* for HK006—triangle symbol), away from direct land-based pollution sources, ensuring that any geochemical variability would reflect regional environmental changes. HK006 spans continuously the decade 2000–2010. HK001 spans the last 200 years, but it is comprised of three sections split by two growth stops of unknown duration. A more complete description of these samples is available in Goodkin et al. (2011), Wang et al. (2011), and Yang and Goodkin (2014). These coral records were also complemented by coral samples from various genera held in museum collections of three institutions: *The Swire Institute of Marine Science* (SWIMS, The University of Hong Kong, Shek O, HKSAR), *The Smithsonian National Museum of Natural History* (Washington, DC, USA), and *The Yale Peabody Museum of Natural History* (New Haven, CO, USA). Museum-held specimens present the advantage of having a record of the sampling location and of the collection date; however, the oldest specimens available were collected in the 1850s, limiting the timeframe of the proposed study. To overcome this issue, fossil coral fragments were obtained from a tombolo excavation pit in *Siu A Chau* (Soko Islands; square symbol on Figure 1b) and sediment cores extracted in two locations of the HKSAR hosting, or

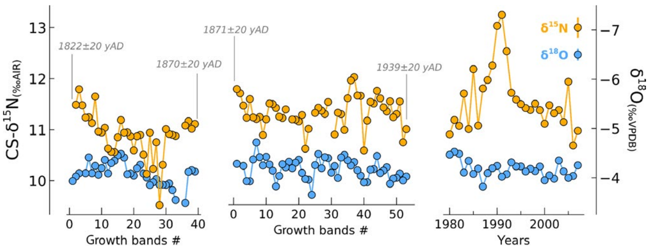

FIGURE 2 $\delta^{18}O$ and CS-$\delta^{15}N$ records from *Porites* core HK001. The analytical error is 0.12‰ for $\delta^{18}O$ and 0.17‰ for CS-$\delta^{15}N$ analyses

known to have hosted, coral communities in the past: *Sham Wan* (Lamma Island, triangle symbol) and *Bluff Island* (diamond symbol). Museum-held and sedimentary coral samples used in this study are listed in Tables S1–S4.

2.2 | Age model and dating

The age model of coral HK006 was based on growth band counting. The age model of core HK001 is complicated by the two growth stops (Figure 2). The age model was thus based on a reanalysis of the original material of Wang et al. (2011) and from additional slabs. X-rays of all slabs were obtained at the *Ocean Park Veterinary Hospital* (Aberdeen, HKSAR) to reveal the annual growth bands and build a new composite chronology used to constrain the calibration of the original radiocarbon dating. The original radiocarbon dating of the core HK001 was performed at the *WM Keck Carbon Cycle Accelerator Mass Spectrometry Laboratory* at *the University of California*; coral samples from sedimentary deposits were dated at the *Beta Analytics Mass Spectrometry Laboratory* in Miami, Florida, USA. All radiocarbon dates were calibrated with OxCal version 4.3.2 (Ramsey, 2009, 2017) using the updated Marine13 radiocarbon age calibration curve (Reimer et al., 2013). Additional information about the calibration is available in the Supporting Information section: *Calibration of the radiocarbon dates*.

2.3 | Coral skeleton stable nitrogen isotope ratio (CS-δ15N) analysis

HK001 and HK006 were sampled at an annual resolution by milling each growth band using a diamond burr mounted on a hand-held drill. Coral fragments (museum and sedimentary) were ground using a mortar and pestle. Coral samples were then sieved and the fraction with a grain size of 63–250 µm was kept for analysis in combusted (500°C—5 hr) borosilicate vials. CS-$\delta^{15}N$ analysis was by persulfate oxidation to nitrate followed by nitrate isotopic analysis with the denitrifier method (Sigman et al., 2001; Wang et al., 2014; Weigand, Foriel, Barnett, Oleynik, & Sigman, 2016). Samples were prepared at *The School of Biological Sciences*, *The University of Hong Kong*, HKSAR and analyzed at the Sigman Laboratory at *Princeton University*, NJ, USA and replicated at the Martínez-García Laboratory at the *Max Planck Institute for Chemistry*, Mainz, Germany. In both laboratories, international amino acid standards (USGS-40 and USGS-41) and nitrate standards (IAEA-NO_3 and USGS34) were used for calibration and blank correction, and an in-house coral standard was used to monitor reproducibility. The average analytical reproducibility was 0.17‰ (n = 22; Figure S1). More information about the analysis can be found in the Supporting Information section: *Coral skeleton stable nitrogen isotopes analysis*.

2.4 | $\delta^{18}O$ analysis

Coral skeleton $\delta^{18}O$ is widely used as a proxy for sea surface temperature (SST) and/or sea surface salinity (SSS; Weber & Woodhead, 1972). To assess the impact of climate change on the coral communities of the Pearl River estuary, 100 µg aliquots of HK001 samples were analyzed for $\delta^{18}O$ in the inorganic stable isotopes Laboratory at *The Max Planck Institute for Chemistry*, Mainz, Germany. Two in-house carbonate standards, calibrated to international standards, were used to ensure the quality of the analysis. The analytical reproducibility was 0.12‰ (n = 192; 1*SD*).

2.5 | Environmental datasets

The Hong Kong Environmental Protection Department water quality monitoring program (www.epd.gov.hk) provided monthly SST (Figure 3a), SSS (Figure 3b), and dissolved inorganic nitrogen concentration (DIN; Figure 3f) records from 76 stations. The data from 13 stations located in the vicinity of core HK001 (MM2, MM3, MM4, MM5, MM6, MM7, MM8, MM13, MM14, MM15, MM16, MM17, and MM19) were selected to characterize the environmental fluctuation over the period 1986–2010 (Figure S4). Each of the three datasets included 2,960 monthly measurements. A historical rainfall time series measured at the Hong Kong Royal Observatory, spanning the period 1853–2004, was complemented by 15 rainfall records measured throughout the HKSAR during the period 1963–1992 (Figure S2). More information about the datasets used to construct this time series can be found in Table S5.

2.6 | PRD demographic data

Population time series over the period 1950–2010 were compiled for each of the eight major cities of the PRD megacity (Guangzhou, Foshan, Dongguan, Jiangmen, Zhongshan, Zhuhai, Shenzhen, Huizhou) and for the HKSAR using the United Nation Report: Revision of the World Urbanization Prospects (UN, 2018). Population data were available for years 1950, 1960, 1970, 1980, 1990, 2000, and 2010 and the population evolution was expressed as the average percentage of population change between each of these years, for each city (Figure 3g; Table S6).

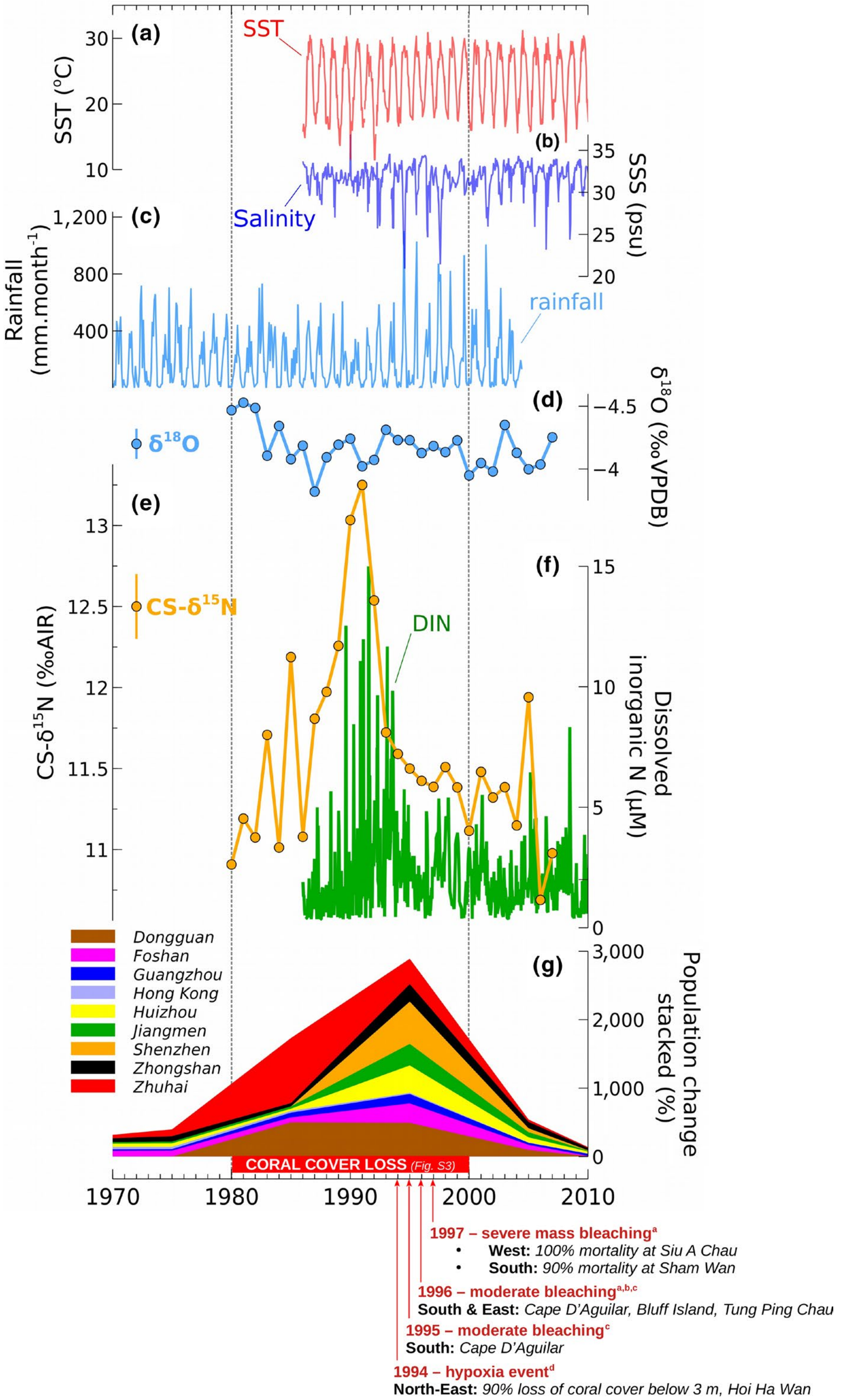

FIGURE 3 Time courses of sea surface temperature (SST), sea surface salinity (SSS), rainfall and dissolved inorganic nitrogen (DIN) over the period of the decline of the Pearl River estuary's coral communities in the decades 1980–2000. (a) SST measurements (data: HKEPD stations MM2-MM19), (b) SSS measurements (data: HKEPD stations MM2-MM19), (c) composite rainfall time series (data: Hong Kong Observatory), (d, e) $\delta^{18}O$ and CS-$\delta^{15}N$ records from *Porites* core HK001, (f) dissolved inorganic nitrogen (DIN) concentration in eastern Hong Kong (data: HKEPD stations MM2-MM19), (g) stacked record of percent population change in the PRD since 1970 (Table S5). The coral communities degradation timeline is based on the report of DeVantier and McCorry (2003; see Figure S3 for details) and on the following sources: [a]McCorry (2002); [b]McCorry (1996); [c]Clark (1998); [d]Collinson (1997)

3 | RESULTS

3.1 | Environmental datasets

The Pearl River estuary has a strong seasonality marked by an average summer (June–July–August) SST of 28.2 ± 1.4°C and average winter (December–January–February) values of 18.0 ± 2.2°C. Rainfall is higher (386 ± 198 mm/month) in summer and lower in winter (32 ± 38 mm/month). Precipitation and SSS variations are correlated (annually averaged data over 1986–2004; *R*: −.71; *p* < .001; *Pearson Correlation*).

Seasonal hydrological changes (rainfall, Pearl River outflow) results in a 2 psu drop in salinity (30.0 ± 2.5 psu) in summer relative to winter values (32.5 ± 0.9 psu). No trend is observed in the

151 years-long rainfall record ($p > .05$; *Pearson Correlation*). Similarly, SST and SSS records do not show a trend over the period 1986–2010 ($p > .05$; *Pearson Correlation*), indicating stable conditions over the Pearl River estuary area at decadal timescales. However, seasonal precipitation extrema (>800 mm/month) were recorded 20 times since the beginning of the rainfall monitoring (Figure S2). The period 1990–2000 stands out with seven rainfall extrema recorded during the decade 1990–2000.

3.2 | Population change in the PRD cities

The population of the major PRD cities was stable from 1950 to 1980 (Table S6). In 1950, aside from Guangzhou and Hong Kong, which had already populations >1 million, none of the seven other cities of the delta had populations above 100,000 inhabitants. The entire population of the delta remained close to 8 million until 1980. During the period 1980–2000, a dramatic population increase occurred, with percent population change being higher than 100% for the eight major cities of the PRD reaching 500% in Dongguan (1980–1990), 620% in Shenzhen (1990–2000), and as high as 950% in Zhuhai (1980–1990). Unlike neighboring cities, the population of the HKSAR increased slowly during this period, with population increases of 26% and 18% over the decades 1980–1990 and 1990–2000. By 2000, the cumulative population of the eight major PRD cities and the HKSAR had reached 32 million and by 2010, 48 million.

3.3 | Coral core HK001

The age model reanalysis of core HK001 provided the following dates for the three sections: 1822 ± 20 years to 1870 ± 20 years for the lower section, 1871 ± 20 years to 1939 ± 20 years for the middle section, and 1980–2007 for the top section (Figure 2). The δ^{18}O record yields a mean value of −4.2 ± 0.2‰ from the early 19th century to 2007, with decadal fluctuations of ~1‰ but no remarkable trends throughout the record (Figure 3d). The δ^{18}O record of the top section was correlated with neither SST ($p > .05$; *Pearson Correlation*) nor rainfall ($p > .05$; *Pearson Correlation*), suggesting a discrepancy between the environmental conditions measured by meteorological stations and the conditions at the coral communities depth. The CS-δ^{15}N time series displayed relatively large oscillations (between 10‰ and 12‰) and recorded a strong ^{15}N enrichment (>12‰) during the period 1987–1993 (Figure 2). The lower section of the core recorded a mean CS-δ^{15}N value of 10.9 ± 0.4‰ ($n = 36$) whereas the middle and the top section (years 1987–1993 excluded) recorded a similar value of 11.3 ± 0.3‰ ($n = 50$ and $n = 21$, respectively). The 0.4‰ difference between the lower section and the middle/top section was significant ($p < .05$; *Mann–Whitney pairwise comparison with Bonferroni correction*). The main feature of the CS-δ^{15}N record is the 1987–1993 peak, with a mean CS-δ^{15}N of 12.4 ± 0.6‰ and peak values reached in 1990 (13.0‰), 1991 (13.3‰), and 1992 (12.5‰; Figure 3e). The mean CS-δ^{15}N over the period 1987–1993 was higher than the lower section (by 1.5‰), the middle section (by 1.0‰), and the top section (by 1.0‰, 1987–1993 excluded, $p < .05$, *Mann–Whitney pairwise comparison with Bonferroni correction*).

Although both δ^{18}O and CS-δ^{15}N records did present decadal fluctuation, no correlation was found between δ^{18}O and CS-δ^{15}N for each of the core sections ($p > .05$; *Pearson correlation*), indicating that, in an estuarine context, seawater temperature and salinity variation were not coupled to changes in N cycling.

3.4 | CS-δ^{15}N baseline

A total of 34 coral samples from 12 genera was gathered from the museum collections, and 8 coral samples were recovered from the sediment core/ tombolo excavation (Tables S2–S4). The trophic level differences among coral genera are still not well described and may result in taxonomic offsets in CS-δ^{15}N. As such, the comparison focused mainly on the most abundant genus, *Acropora*. The samples were pooled into three distinct periods. The *pre-industrial* group included all but one of the samples recovered from the sedimentary deposits ($n = 7$). Samples of this group spanned the period ca. 5,000 yBP to the 15th century, when the anthropogenic N footprint is assumed to be negligible compared to the natural processes driving the N cycle. All samples from the 19th century were pooled into a second group. This group included all the samples collected during the *US North Pacific Exploration Expedition* in 1854 in “coral bay” (now *Chai Wan* typhoon shelter) and one sample from a sediment core collected in Bluff Island. Samples from this period ($n = 8$) are assumed to characterize the early anthropogenic N footprint of the PRD development. The last group included coral samples from the period 1972–1983 ($n = 27$) held in museum collections. This group is composed of samples collected before the 1987–1993 CS-δ^{15}N peak recorded in core HK001 (Figure 3e). All three periods were characterized by CS-δ^{15}N values ranging from 9‰ to 11‰, and no remarkable trend was identified (Figure 4). Mean CS-δ^{15}N values (genus *Acropora* only) were: *pre-industrial* group = 9.8 ± 0.8‰ ($n = 6$), *19th century* group = 10.5 ± 1.0‰ ($n = 5$), and *1972–1983* group = 9.7 ± 0.6‰ ($n = 14$). Mann–Whitney pairwise comparison failed to detect any difference among the three periods, suggesting that the processes driving the N cycle in the PRD estuary were relatively stable over the last 5,000 years until the 1980s.

3.5 | DIN concentration in eastern HK

Coral communities of the HKSAR have been shown to present a dramatic reduction of coral-specific richness and coral cover at DIN concentrations exceeding 2 μM. This value was proposed by Duprey et al. (2016) as a threshold for local coral communities.

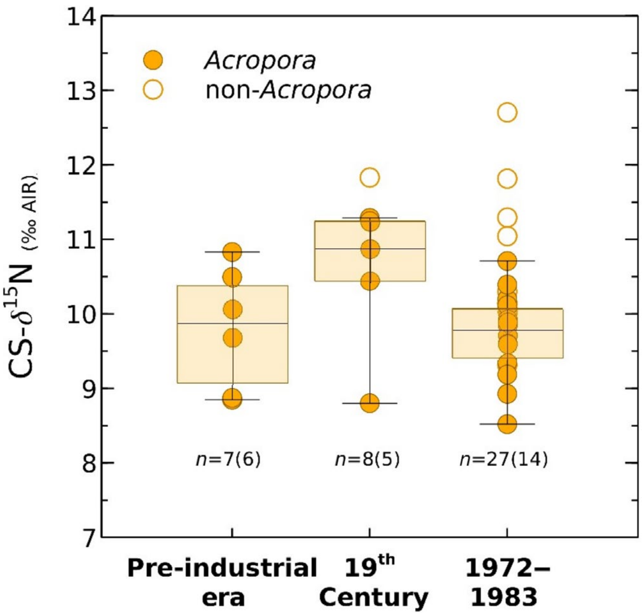

FIGURE 4 CS-δ^{15}N ranges from the pre-industrial period and during the early phase of the PRD megacity development. The boxplots are calculated from the *Acropora* data only. Numbers below the boxplot indicate the total number of samples for each period, and the numbers in brackets indicate the number of *Acropora* samples

Until 1989, annually averaged DIN concentration remained <2 μM (1986–1989: DIN = 1.4 ± 0.3 μM). The next 5 years were characterized by DIN concentrations consistently higher than 2 μM (1990–1995: DIN = 3.3 ± 1.3 μM) with maximum values measured in 1991 (5.4 μM) and in 1993 (4.2 μM). DIN concentrations decreased over the period 1996–2010 but remained >2 μM half of the time (1996–2010: DIN = 1.9 ± 0.5 μM). The DIN peak recorded in the early 1990s was due to an increase in NH_4^+ concentration (Figure 5a). Over the period 1987–1993, 80% of DIN was composed of NH_4^+, whereas NO_3^- concentration remained relatively stable throughout the period 1986–2010 (0.3 ± 0.16 μM). The CS-δ^{15}N record was correlated with NH_4^+ concentration over the period 1986–2007 (R = .57, p < .05, *Pearson correlation*, Figure 5b), suggesting that the 1987–1993 peaks in NH_4^+ concentration and CS-δ^{15}N record were causally connected, with the rise in NH_4^+ concentration leading to the rise in CS-δ^{15}N. Over 1994–2010, NH_4^+ concentration was negatively correlated with salinity (R = –.53, p < .05, *Pearson correlation*, Figure 5c), pointing to the Pearl River as the main source of NH_4^+. However, during the 1987–1993, the NH_4^+ concentration was not correlated with salinity (p > .05, *Pearson correlation*), which remained within a narrow range (31–32 psu) despite a fourfold increase in NH_4^+ concentration. The decoupling between salinity and NH_4^+ concentration indicates that the NH_4^+ spike was not related to a change in the Pearl River discharge, but instead suggests an increase in the NH_4^+ load in the estuary. This inference is supported by the lack of correlation between δ^{18}O and CS-δ^{15}N (p > .05, *Pearson correlation*).

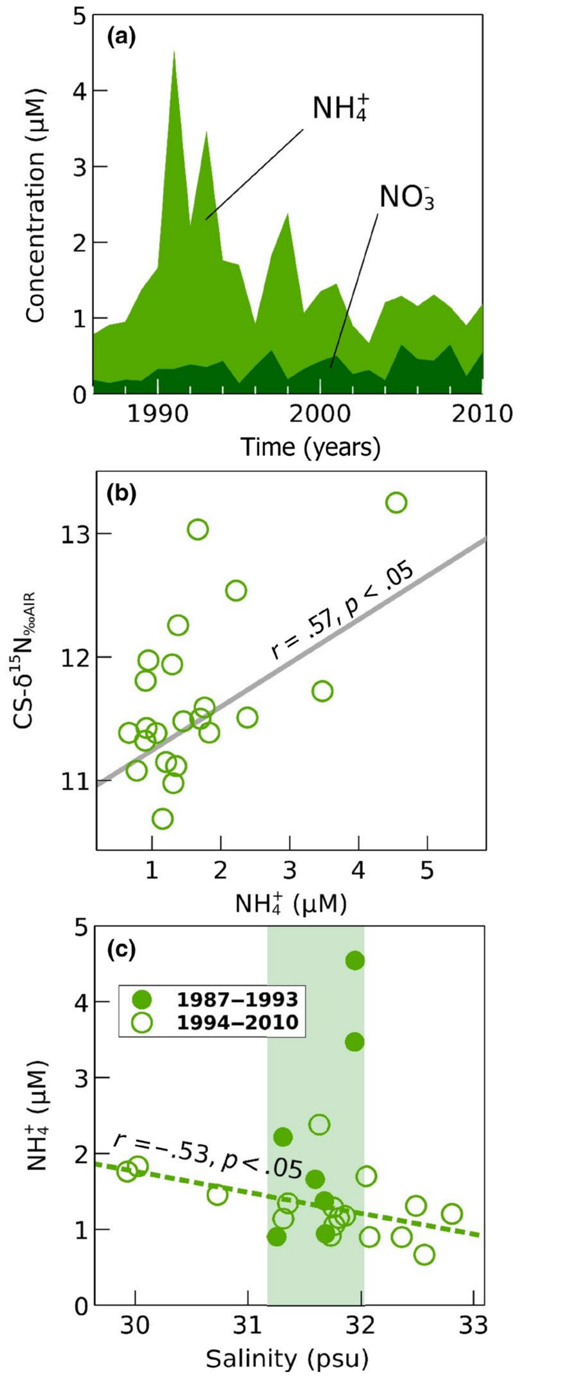

FIGURE 5 Dissolved inorganic nitrogen (DIN) concentration during the period 1986–2010 in comparison to CS-δ^{15}N and salinity changes. (a) Ammonium (NH_4^+) and nitrate (NO_3^-) concentrations in eastern Hong Kong (*source*: www.epd.gov.hk), (b) correlation between HK001's CS-δ^{15}N record and NH_4^+ concentration (*Pearson Correlation*), (c) correlation between NH_4^+ concentration and salinity during 1987–1993 and 1994–2010 (*Pearson Correlation*). Each water quality data point is the average of 25–156 measurements. The dataset used for this figure is available in Table S7

3.6 | East–West CS-δ^{15}N gradients

The *Porites* samples HK001 (*Pak Lap Tsai*, eastern HKSAR) and HK006 (*Sham Wan*, western HKSAR) provide information about

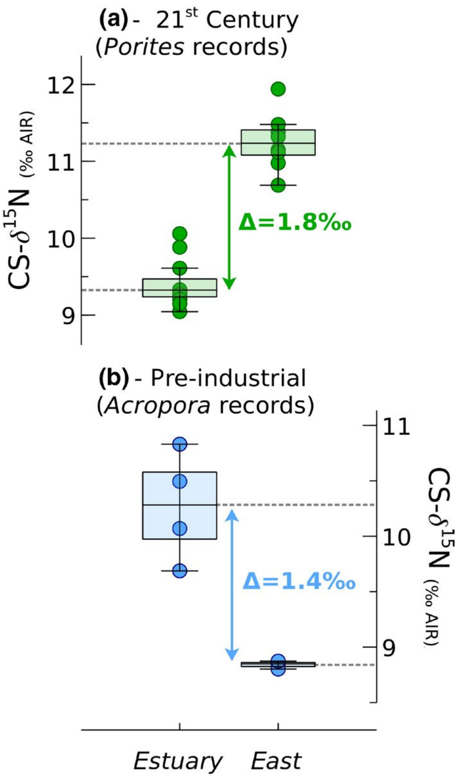

FIGURE 6 CS-δ^{15}N difference (Δ) between the Pearl River estuary and waters to the east during (a) the 21st century (*Porites* data; Figure S5) and (b) the pre-industrial period (*Acropora* data). Direct comparison of *Acropora* and *Porites* CS-δ^{15}N values was avoided due to potential inter-genera offsets caused by trophic level differences. Estuarine sites included Siu A Chau and Sham Wan, and eastern sites included Pak Lap Tsai and Bluff Island (Figure 1b)

west–east δ^{15}N gradients, offering insight into the N footprint of the Pearl River (Figure 1b). HK006 recorded a mean CS-δ^{15}N value of 9.4 ± 0.3‰ (n = 11) over the decade 2000–2010 and HK001 recorded a mean value of 11.3 ± 0.4‰ (n = 8) over the period 2000–2007 (Figure 6a). This 1.8‰ difference was significant (p < .05, *Mann–Whitney Pairwise comparison*) and was relatively constant during the decade 2000–2010 (Figure S5).

Samples from the *pre-industrial* group provided a way to characterize the isotopic west–east gradient of the Pearl River estuary before the PRD urbanization. Samples collected closer to the Pearl River (*Siu A Chau*, *Sham Wan*; Figure 1b) and samples collected further east (*Bluff Island*) recorded a different gradient in CS-δ^{15}N than after 2010: CS-δ^{15}N was higher by 1.4‰ on the west (10.3 ± 0.5‰, n = 4) than in the east (8.8 ± 0.04‰, n = 3; Figure 6b). Although this difference was not significant (p < .1, *Mann–Whitney Pairwise comparison*), it indicates that a change in the east–west dynamic of the N cycle has occurred. HK001 and HK006 are *Porites* specimens, whereas the *pre-industrial* group is composed of samples from the genus *Acropora*. Due to possible trophic level differences between these coral genera, we made no direct comparison of the CS-δ^{15}N between these two periods.

4 | DISCUSSION

4.1 | Are coral communities in the Pearl River estuary affected by global warming and its associated hydrological changes?

The PRD megacity provides a unique opportunity to assess the respective roles of global and regional anthropogenic activities in coral reef decline. Although the impact of urbanization in the PRD on marine ecosystems has been widely documented over the last 40 years (McCorry & Blackmore, 2000; Morton, 1975; Wong et al., 2018), the impact of global warming and its associated hydrological changes remains unclear. Coral δ^{18}O provides a good proxy for in situ salinity and temperature (Juillet-Leclerc & Schmidt, 2001) experienced by coral communities and thus allows assessment of the extent to which the environmental parameters are impacting coral communities. The stable coral δ^{18}O values observed indicate that both SST and SSS remained stable over the period 1820–2007 (Figure 2). This is corroborated by the rainfall record (proxy for salinity) covering the period 1853–2004 (Figure S2), indicating a stable rainfall regime over the period of interest. Although much shorter, the SST dataset from the Hong Kong EPD, extending back to 1986, also recorded stable conditions over the most recent part of the coral record (Figure 3a). The absence of summer coral bleaching events in the Pearl River estuary since 2000 (Ang et al., 2005; Wong et al., 2018) supports this assumption, although the exception of a weak bleaching event reported in 2014 indicates that the situation may be changing (Xie et al., 2017). That said, we argue that prior to 2007 (the scope of this study), global warming and its associated hydrological changes had not greatly impacted coral communities in the Pearl River estuary.

4.2 | Controls of the N cycle in the Pearl River estuary from the mid-Holocene until the 1980s

The stable nitrogen isotope values from coral samples found in sedimentary records and natural history museum collections provide insight into the changes that affected the N cycle of the Pearl River estuary over the last 5,000 years. *Acropora* CS-δ^{15}N values did not change markedly from the mid-Holocene to the 1980s, with values ranging from 9‰ to 11‰ (Figure 4); similarly, *Porites* CS-δ^{15}N values remained stable from the 1820s until the 1980s (10‰–12‰, Figure 2). A closer look at the pre-industrial period dataset shows that CS-δ^{15}N values tended to be ~1.4‰ higher toward the Pearl River estuary than at sites located further east (Figure 7a). The Pearl River estuary was thus the main source of ^{15}N-enriched N, whereas lower δ^{15}N values in the east were probably the result of mixing with South China Sea seawater, which has a shallow thermocline nitrate δ^{15}N value of ~5‰ (Ren, Chen, et al., 2017). N_2 fixation is an additional possible contributor to the eastward CS-δ^{15}N decline. Denitrification and related redox processes in the PRD, its tributaries, and the Pearl River estuary may lead to an excess of phosphorus

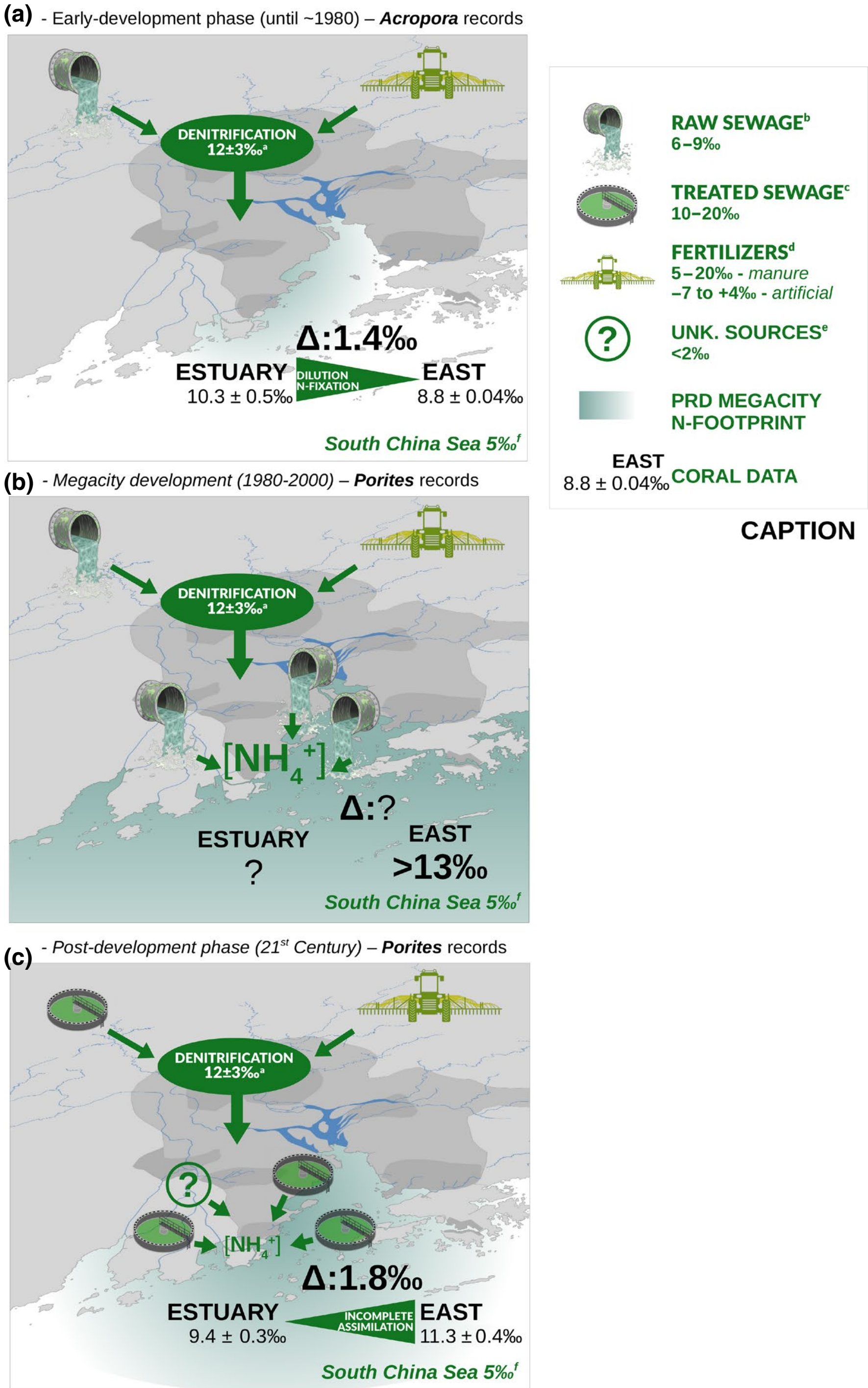

FIGURE 7 Cartoon showing the main anthropogenic DIN sources and processes controlling the isotopic composition of the DIN pool in the Pearl River estuary in summer (i.e., the period imprinted on the coral CS-δ^{15}N records). (a) In the early development phase of the PRD, the δ^{15}N of anthropogenic N sources is homogenized through upstream denitrification, causing the estuary to be isotopically enriched compared to the east, which is under the influence of low δ^{15}N DIN from the South China Sea. (b) During the development of the PRD into a megacity, the massive release of ammonium, originating from raw sewage, directly in the estuary, bypassed the upstream denitrification and caused an increase in the concentration and δ^{15}N of DIN in coastal waters. (c) The post-development of the PRD megacity is accompanied by an overall improvement of the sewage treatment facilities, curbing the DIN concentration in the east. Yet, the Pearl River N footprint has not returned to its pre-development configuration, leaving the east isotopically enriched through incomplete nutrient assimilation by planktonic communities. Sources: [a]Chen, Jia, and Chen (2009); [b]Kendall et al. (2007); [c]Archana, Li, Shuh-Ji, Thibodeau, and Baker (2016); McClelland, Valiela, and Michener (1997); [d]Fry (2006), Kendall et al. (2007); [e]Ye et al. (2015); [f]Ren, Sigman, et al. (2017)

relative to N, which then encourages N_2 fixation as the Pearl River estuary waters mix with low-nutrient seawater. The substantial anthropogenic inputs do not appear to cause a large N_2 fixation response in the Pearl River estuary today. However, this may be due to elevated DIN concentration throughout the Pearl River estuary, with N_2 fixation occurring further offshore.

In the Pearl River estuary, coral growth is challenged by the low winter temperature (down to 13°C) causing frequent "low temperature bleaching," in particular in massive *Porites* (Ang, 2002; McCorry, 2002); as a result, in the Pearl River estuary, corals achieve most of their annual growth in summer (i.e., April–October). The N turnover in the tissues and symbionts of the genus *Porites* is close to 3 months (Rangel et al., 2019); as such, the N sources present in the summer are likely overrepresented in the CS-δ^{15}N records. Given that 80% of the Pearl River discharge occurs in summer (Lee, Harrison, Kuang, & Yin, 2006), when the Southwest monsoon pushes the river plume eastward (i.e., toward the sampling sites; Figure 1b), we suggest that the coral CS-δ^{15}N records are biased toward PRD-driven high δ^{15}N relative to annual average conditions (Wong, Duprey, & Baker, 2017).

Stable CS-$\delta^{15}N$ values ranging from 9‰ to 12 ‰ until the 1980s were unexpected considering that the population in the PRD has experienced dramatic changes over this period, with a population increase from 1–2 million in mid-19th century to over 8 million in 1980 (Benedict, 1988; Gauducheau, 1911; Table S5), and the development of livestock agriculture (Morton, 1989) and aquaculture (Morton, 1975), together with the use of synthetic fertilizers (www.earth-policy.org/data_center). These changes should have strongly altered the pre-industrial isotopic baseline, with sewage, manure, livestock, and aquaculture being enriched in ^{15}N, whereas synthetic fertilizers tend to be depleted in ^{15}N (Kendall, Elliott, & Wankel, 2007). The invariant, elevated coral $\delta^{15}N$ values until the 1980s suggest the existence of strong regulating mechanisms, capable of maintaining a stable isotopic composition despite the increase in anthropogenic N inputs. We propose that summer soil and stream denitrification in the Pearl River watershed controlled the concentration and isotopic composition of the DIN pool.

Accelerated N loss in response to increased anthropogenic N loadings would have tended to raise the $\delta^{15}N$ of the residual DIN (Amundson et al., 2003; Kendall et al., 2007). As DIN is transferred through the terrestrial-estuarine *continuum*, it experiences various transformations, the most important being the microbially mediated removal of DIN through the oxidation of NH_4^+ to NO_3^- (nitrification) and the subsequent reduction of NO_3^- to N_2O and N_2 (denitrification) (Peterson et al., 2001; Seitzinger et al., 2006). This N removal can have a strong influence on the $^{15}N/^{14}N$ ratio, preferentially removing ^{14}N and leaving the DIN pool enriched in ^{15}N (Granger et al., 2011; Martinelli et al., 1999; Seitzinger et al., 2006). Soils and streams play a key role in N removal by providing suboxic and organic matter-rich substrates suitable for coupled nitrification/denitrification. In the case of the Pearl River watershed, the higher degree of consumption would have maintained that DIN at adequately low concentrations that prevented its $\delta^{15}N$ from overwhelming that of the other N sources to the Pearl River estuary. Moreover, because of the high degree of consumption in the sites of occurrence, the isotopic impact of denitrification in soils and marine sediments is often minimal (Brandes & Devol, 1997; Houlton, Sigman, & Hedin, 2006). Studies of heavily eutrophicated watersheds have revealed that the nitrification/denitrification process is particularly resilient to high anthropogenic loads and responds to increasing load by increasing rates of denitrification (Battaglin, Kendall, Chang, Silva, & Campbell, 2001; Panno, Hackley, Kelly, & Hwang, 2006; Peterson et al., 2001). These effects together would explain why the DIN input from the PRD was never so great as to completely overwhelm the low-$\delta^{15}N$ signal of South China Sea–sourced nitrate in the outer Pearl River estuary (Figure 7a).

A study led by Chen (2015) on the Beijiang River Watershed (the second largest tributary of the Pearl River) provides an example. The NO_3^- $\delta^{15}N$ values measured from the upper reach to the lower reach, at seven sites and five tributaries, cover a broad range, from 1.9‰ to 17.6‰, with a wide range of anthropogenic N sources (i.e., synthetic fertilizer, sewage) close to the sampling sites. Despite this extensive variation among the tributaries and the expectation for an increasing anthropogenic N footprint downstream, no trend was observed from the upper to the lower reach of the river. Instead, a strong seasonal signal was found in the NO_3^- $\delta^{15}N$ values throughout the watershed. This indicates that temperature and its control of soil/stream denitrification prevailed over N source variation in setting the river $\delta^{15}N$ DIN. The mean summer NO_3^- $\delta^{15}N$ was 11.8 ± 2.7‰, which is consistent with the CS-$\delta^{15}N$ range of 9‰–12‰ recorded in the Pearl River estuary corals until the 1980s.

Efficient N removal upstream of the PRD would have minimized DIN supply to the Pearl River estuary. Moreover, unlike most eutrophied estuaries, water in the Pearl River estuary has a short residence time, 1–3 weeks (Dong, Su, Li, Xia, & Guan, 2006), and a strong upwelling regime develops during summer, diluting the Pearl River plume offshore (Lee et al., 2006). These factors would tend to prevent the development of sub/anoxic water layers required for direct denitrification in the water column (Naqvi, 1994) or the high flux of organic matter to the seabed required for coupled partial nitrification and denitrification in the sediments. As these processes are most prone to drive strong isotope fractionation during DIN loss (Granger et al., 2011; Sigman et al., 2003), this would explain the lack of a clear isotopic signal from N processing within the Pearl River estuary up to the 1980s. Canonical sedimentary denitrification within the Pearl River estuary has likely always been active. However, as mentioned above, this process typically expresses minimal isotopic fractionation (Brandes & Devol, 1997; Lehmann, Sigman, & Berelson, 2004). In summary, we argue that DIN removal by upstream denitrification and the relatively active circulation in the Pearl River estuary were responsible for the elevated, yet stable, CS-$\delta^{15}N$ values found in the Pearl River estuary before 1980, despite the progressively increasing anthropogenic N discharge to the Pearl River hydrologic system (Figure 7a).

4.3 | Pearl River delta megacity development: 1980–2000

The second phase of the PRD urbanization is characterized by explosive population growth during the decades 1980–2000, bringing the delta population from 8 million before 1980 to over 32 million in 2000 (Figure 3g). Whereas the first urbanization phase affected mostly the upstream delta area (i.e., Guangzhou), the second phase was characterized by the development of coastlines, with the steady growth of the HKSAR and the booming of Zhuhai and Shenzhen (Figure 1b; Table S6). This phase coincided with a spike (>13‰) in the *Porites* CS-$\delta^{15}N$ record, reached in 1993 (Figure 3e). These elevated CS-$\delta^{15}N$ values were unprecedented in the 200 year-long core and in the 5,000 year-old fossil records (Figure 4), indicating dramatic changes in the N cycle of the Pearl River estuary after 1980.

Two trends are observed in the evolution of the CS-$\delta^{15}N$ and the DIN concentration during the 1980s. From 1987 to 1993, the N species concentration data show that the CS-$\delta^{15}N$ spike occurred concomitantly with a fivefold increase in DIN concentration caused by an increase in the concentration of NH_4^+ (Figure 4a,b). The lack of correlation between NH_4^+ concentration and salinity during 1987–1993 (Figure 5c) suggests that NH_4^+ did not enter via the Pearl River itself but instead that the NH_4^+ discharge occurred nearby or directly into the estuary. In this scenario, the anthropogenic N load partially bypassed the soil and stream denitrification in the Pearl River and its upstream hydrologic system, vastly raising the DIN load to the estuary (Figure 7b). At the same time, this anthropogenic N did undergo some processing and loss, leading to a high $\delta^{15}N$ for the DIN input, as recorded in the CS-$\delta^{15}N$ record. From 1993 to 2000, NH_4^+ concentration and CS-$\delta^{15}N$ decreased steadily to the early 1980 levels (Figure 3e,f). During this period, NH_4^+ concentration in the estuary was negatively correlated with salinity (Figure 5c), pointing to the Pearl River as the main source of NH_4^+ and suggesting a decline in or cessation of the direct discharge of N into the estuary.

These observations corroborate the study of Qin, Su, Khu, and Tang (2014) who highlighted the major lag between the population growth in Shenzhen and the development of the wastewater treatment infrastructure before 1995. During this period, the domestic wastewater discharge increased faster than the capacity of the sewage treatment facilities, until a point was reached in 1995–1996 when sewage removal capacities increased faster than the sewage discharge. We argue that the trends observed in CS-$\delta^{15}N$ and NH_4^+ concentration reflect this dynamic, with increasing values until 1993 corresponding to the net increasing untreated sewage discharge and the decline in CS-$\delta^{15}N$ and NH_4^+ concentration after 1993 reflecting the net reduction in the untreated sewage load. Shenzhen's example reflects the changes in sewage treatment infrastructure occurring at the level of the megacity, pointing to the urbanization of the PRD area, and in particular of coastal cities (cf., Shenzhen, Zhuhai) as the main cause for this major eutrophication event. The slight offset between the CS-$\delta^{15}N$ peak of 1993 and the transition in Shenzhen in 1995 could be explained by differences in the timing of sewage treatment infrastructure improvement between the cities of the PRD. In addition, the development of the HKSAR New Territories (*Shatin*, *Man On Shan*, *Tai Po*) in 1970–1990 (McCorry & Blackmore, 2000) and the resulting N discharge is likely to have influenced the CS-$\delta^{15}N$ record as well.

The development of eutrophic conditions in the estuary during 1980–2000 has had dramatic consequences for the marine ecosystem and for coral communities in particular. The severe eutrophication of Tolo Harbour, following the fast development of the cities of Sha Tin and Tai Po in 1970–1990, has been identified as the main cause of the coral demise in this area (McCorry & Blackmore, 2000; Wong et al., 2018). Yet, earlier studies failed to provide convincing explanations for the more widespread coral community decline observed across the Pearl River estuary during 1980–2000. The coral communities in the estuary started to show signs of stress in the early 1980s, with minor to moderate coral mortality (Clark, 1998; Collinson, 1997; Cope, 1984; McCorry, 2002). The following decade was characterized by more frequent and intense coral degradation events (Figure 3). In 1994, a localized hypoxia event took place in *Hoi Ha Wan* (NE), resulting in the loss of 90% of coral cover below 3 meters depth; in 1995, moderate bleaching affected corals at *Cape D'Aguilar* (South); in 1996, the bleaching extended to the south and the east of the HKSAR (*Cape D'Aguilar*, *Bluff Island* and *Tung Ping Chau*). The most dramatic event of the last 40 years occurred in 1997 when a severe mass-bleaching event affected the entire HKSAR area, ranging from the partial mortality of several communities in the eastern side to the complete collapse of *Sham Wan* (90% mortality) and *Siu A Chau* (100% mortality) communities in the western area (Figure 3). During this widespread event, 30% of the 90 hard coral species inventoried in the Pearl River estuary area exhibited a bleaching response (McCorry, 2002), highlighting the severity of the event.

The high rainfall observed in the decade 1990–2000 (>800 mm/month; Figure S2) and the subsequent salinity drop were proposed as the cause of the widespread coral mortality (Clark, 1998; Collinson, 1997; McCorry, 2002). Interestingly, coral-threatening SSS values, that is, <22 psu (Berkelmans, Jones, & Schaffelke, 2012; Coles & Jokiel, 1992), were only measured in summer 1994 (20.9 psu, for a corresponding rainfall of 1,101 mm/month) and in summer 1997 (21.4 psu, for 798 and 873 mm/month in August and June, respectively). 1994's summer presents the second most extreme event of the record, and 1997's summer includes the 11th (June) and the 20th (August) strongest events. Yet, SSS does not seem to respond to extreme seasonal rainfall events in the Pearl River estuary, probably due to the short residence time of the water in the estuary (Dong et al., 2006). Although these salinity values were on the lower end associated with coral reefs and probably induced a significant stress to the coral communities, we argue that the salinity drop alone was not sufficient to trigger the collapse of entire coral communities as observed in *Siu A Chau* and *Sham Wan* and the severe mortality observed along the HKSAR coastlines (Figure S3). Indeed, similarly high rainfall events occurred 13 times during the period 1853–1997, including the more recent years 1952, 1957, 1959, and 1966 (Figure S2). No coral die-offs were reported prior to the decade 1990–2000 (McCorry, 2002), arguing that the high rainfall events alone cannot explain the coral demise in the Pearl River estuary.

The progressive increase in the frequency and the magnitude of the coral communities' degradation events throughout the period 1980–2000 was synchronous with the increase in DIN concentration in coastal waters (Figure 3). This suggests that the coral decline in the Pearl River estuary was primarily driven by eutrophication caused by the development of the PRD megacity. The greater severity of mortality toward the Pearl River estuary (Figure S3), where DIN concentrations are higher (Duprey et al., 2016), also supports this interpretation. Duprey et al. (2016) have suggested an annual average concentration of 2 µM DIN as a threshold for the coral communities of the Pearl River estuary: beyond this threshold, coral communities experience a severe decrease in coral cover and coral-specific richness. During the period 1990–1995, the DIN concentration in eastern HK was consistently above this threshold (Figure 5a), suggesting that the coral communities' decline was caused by an increase in the DIN concentration. It is beyond the scope of this study to propose a mechanism for how excess DIN concentration leads to the decline in coral communities in the Pearl River estuary; however, elevated DIN concentration in coral reefs is known to be detrimental to corals at both ecosystem and physiological levels (Fabricius, 2005). Eutrophication of coastal waters is associated with reduced living coral cover and biodiversity as well as increased bioerosion (Cortes & Risk, 1985; Edinger et al., 2000; Fabricius, 2005). N-enrichment has also been linked to increased coral disease prevalence (Vega Thurber et al., 2014) and depressed coral reproductive success (Loya, Lubinevsky, Rosenfeld, & Kramarsky-Winter, 2004). The period 1980–2000 is also characterized by elevated DIN:DIP ratio (>30:1; Figure S6), compared to the typical range of 4.3:1 to 7.2:1 observed in healthy coral reefs (Crossland, Hatcher, Atkinson, & Smith, 1984; Furnas, Mitchell, & Skuza, 1995; Smith, Kimmerer, Laws, Brock, & Walsh, 1981). Elevated DIN concentration and high DIN:DIP ratio are known to lower coral thermal threshold by disrupting of the coral–algal symbiosis and causing coral bleaching (Baker, Freeman, Wong, Fogel, & Knowlton, 2018; Lapointe, Brewton, Herren, Porter, & Hu, 2019; Wiedenmann et al., 2012). Although eutrophication is the likely cause of the Pearl River estuary coral communities' demise in 1980–2000, it is important to keep in mind that coral communities in the Pearl River estuary region have been under strong anthropogenic pressure over the last 200 years, in particular from high population density (Benedict, 1988; Gauducheau, 1911) and overfishing (Cheung, 2017; Vasile, Manning, & Lemaitre, 2005). This long-term exposure to anthropogenic stress is likely to have weakened the resilience of the coral communities, making them more vulnerable to the severe eutrophication of 1980–2000.

4.4 | Anthropogenic N footprint in the Pearl River estuary in the 21st century

The implementation of waste management policies and the expansion and upgrade of the existing sewage and wastewater treatment infrastructure over the period 1990–2000 in both China (Qin et al., 2014) and the HKSAR (*Tolo Harbour Action Plan* in 1987; *Tolo Harbour Effluent Export Scheme* in 1998; *Harbour Area Treatment Scheme* in 2001) was followed by a drastic reduction in DIN concentration (Figure 3d) and a decline in CS-δ^{15}N down to 1980 values (Figure 3f). Noticeable improvement in water quality over the last 20 years has also been reported by Duprey, McIlroy, et al. (2017) and Wong et al. (2018), indicating that adequate N mitigation policies can have a positive impact on the water quality without impacting economic growth. Yet, the level of anthropogenic N discharge, in particular from domestic waste, remains high and the development of N removal infrastructure is still behind the current waste production rate (Qin et al., 2014).

The inversion of the west–east CS-δ^{15}N gradient along Hong Kong coastlines reveals that the N cycling in the Pearl River estuary remains strongly altered 20 years after the eutrophication event (Figure 7). During the pre-industrial period, the CS-δ^{15}N gradient in the Pearl River estuary, with CS-δ^{15}N decreasing eastward, was set by the high-δ^{15}N signal of watershed denitrification to the west and mixing with lower δ^{15}N NO_3^- from the South China Sea to the east (Figure 7a). The east–west isotopic gradient observed today is reversed, with CS-δ^{15}N being ~2‰ lower toward the Pearl River estuary compared to eastern values (Figure 7c). The presence of anomalously low-δ^{15}N DIN sources to the Pearl River estuary has been observed in NO_3^- δ^{15}N (Archana et al., 2018; Ye et al., 2015) and in *Porites* tissue δ^{15}N (Wong et al., 2017). This indicates that low-δ^{15}N N is being discharged directly into the estuary, bypassing the upstream isotopic homogenization described earlier (Figure 7c). The origin of the low-δ^{15}N DIN sources may be related to increased synthetic fertilizers use in coastal areas, but it may also originate from industrial waste discharge, the isotopic composition of which is poorly constrained (Huang et al., 2018; Ye et al., 2015).

Low-δ^{15}N N sources' discharge into the estuary cannot alone explain the reversed isotopic gradient observed today. A possible mechanism is an expansion eastward of the Pearl River's nutrient footprint due to the increased nutrient discharge in the estuary. Because high phytoplankton biomass is commonly found at the edge of the coastal plume (Xu et al., 2008; Yin, 2002), the expansion of the Pearl River plume has probably increased the supply of DIN with a high δ^{15}N due to isotopic fractionation during assimilation by phytoplankton. Indeed, Xu et al. (2008) found that, today, primary productivity is light limited due to the high turbidity found in the western part of the Pearl River estuary, whereas the summer peak chlorophyll-a concentration is found further east, in southern Hong Kong waters. As such, plankton assimilation is more likely to increase the δ^{15}N of the N pool in the eastern part of the Pearl River estuary than in the western part, potentially explaining the CS-δ^{15}N gradient (Figure 7c). This highlights that eutrophication is still a major issue in the Pearl River estuary, such that N mitigation should remain an important target of the PRC's and HKSAR's governments for the 21st century.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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