Atmospheric nitrogen deposition from a remote source enriches macroalgae in coral reef ecosystems near Green Turtle Cay, Abacos, Bahamas

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Abstract

Over the past several decades, the fixation of "new" nitrogen to the biosphere has doubled. For the early 21st century, the most significant rate increases in atmospheric nitrogen deposition are predicted for developing nations. Wet nitrogen deposition was assessed on the remote island of Green Turtle Cay, Bahamas in a dry and wet season from January to July 2000. Episodic deposition of nitrate ($\sim$1–137 μM) and ammonia ($\sim$2–122 μM) represented a mean deposition rate of $\sim$0.2 mg DIN m^{-2} yr^{-1}. Wet deposition of nitrogen to the climatologically-linked east coast of Florida is $\sim$4 times greater than the estimated annual wet nitrogen deposition value at Green Turtle Cay, suggesting the continental US as a principal airshed for this loading source. Short-term bioassays of macroalgal productivity with a 5% rainfall solution caused depressed net productivity and increased dark respiration, well known "transient metabolic" responses by nutrient-limited tropical macroalgae. Wet deposition of inorganic nitrogen from episodic rainfall events may provide up to 20% of the "new" nitrogen necessary to meet growth demands of macroalgae on coral reefs near Green Turtle Cay.
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1. Introduction

Within the past two decades, the fixation of "new" nitrogen to the biosphere has doubled as a result of human activities (Vitousek et al., 1997a). This biogeochemical modification has altered patterns of primary productivity throughout the global environment, because most ecosystems on land and in the sea tend to be nitrogen-limited (Vitousek and Howarth, 1991). Coastal marine ecosystems, in particular, are typically limited by nitrogen (Ryther and Dunstan, 1971; Howarth, 1988; Nixon, 1995) and may be one of the more impacted environments on earth as a result of increased nitrogen loadings from coastal watersheds and airsheds (NRC, 2000).

Like the oligotrophic seas they have evolved in, hermatypic coral reef ecosystems are adapted to nutrient limited conditions where natural nitrogen fixation is the dominant source of "new" nitrogen (Wiebe et al., 1975). Alternatively, reefs with high water column nitrogen and phosphorus concentrations are more favorable for fleshy opportunistic algae (Lapointe, 1997) and are less favorable for calcifying macroalgae (Delgado and Lapointe, 1995; Lapointe and Thacker, 2002) and hermatypic corals (McConnaughey et al., 2000). Saturating concentrations of DIN for growth of tropical marine macroalgal are $\sim$0.5–1 μM (see Lapointe, 1999). Because of this adaptation to low nutrient conditions, coral reefs are particularly susceptible to eutrophication via anthropogenic nutrient loading (Lapointe, 1997,

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1999). Macroalgal blooms are becoming more conspicuous on hermatypic coral reefs throughout their circumtropical distribution (Done, 1992; Hodgson, 1999). As a result, macroalgal dominated reefs (see Lighty, 1982; Hughes, 1994; Lapointe, 1997; NRC, 2000) may represent an alternative "stable-state" in the era of increased anthropogenic domination of ecosystems (Vitousek et al., 1997b). Oceanic islands, such as Jamaica, with high topography and carbonate geology are particularly prone to mobilization of sewage, fertilizers and other sources of nitrogen to adjacent reef systems (Marsh, 1977; Rawlins et al., 1998; Lapointe and Thacker, 2002).

Atmospheric deposition of nitrate over the past century has mirrored increases in anthropogenic fossil fuel emissions of NO_x in both North America and Europe (Brimblecombe and Stedman, 1982). Since the 1950s, for example, atmospheric deposition to the northern hemisphere has increased five-fold (Galloway et al., 1994). By the year 2020, atmospheric nitrogen deposition is predicted to increase by 25% in urbanized North America, but more significantly, may double in developing regions such as SE Asia and Latin America, and will increase by more than 50% in oceans of the Northern Hemisphere (Galloway et al., 1994). Recognition of the scope of anthropogenic atmospheric nitrogen deposition (AAND) to the global environment has expanded with reports of continental atmospheric pollution plumes reaching remote oceanic locations (e.g. Abram et al., 2003). Climatological forcing delivers anthropogenic nitrogen downwind from eastern continental regions into western oceanic environments (Miller and Harris, 1985; Levy and Moxim, 1987; Moody and Galloway, 1988). For example, the eastward flux of anthropogenic nitrogen from North America (Galloway et al., 1984a) has been monitored and reported for several decades in Bermuda (see Menzel and Spaeth, 1962; Jickells et al., 1982). On northern Pacific oceanic islands, anthropogenic continental sources from Asia are responsible for 40–70% of nitrate aerosol concentrations that represent values three times higher than background concentrations in the southern Pacific (Prospero and Savoie, 1989).

While nutrient loading from more recognizable and historically significant point and non-point sources (e.g. municipal wastewater, urban stormwater) have been subjected to management programs for reductions (e.g. EPA's Total Maximum Daily Load (TMDL) program, NOAA-Estuarine Eutrophication Survey (see Bricker et al., 2000)), increasing loads from AAND may offset the benefits of these reductions (Paerl, 1995). The symptoms of coastal eutrophication (harmful algal blooms, hypoxia) appear to have intensified despite these initiatives for reductions of traditionally important sources (Smayda, 1990; Bricker et al., 2000), leading some to point to AAND as an emerging and potentially dominant source leading to eutrophication of coastal ecosystems (Paerl, 1995; Jaworski et al., 1997). Coastal waters downwind from industrialized centers such as the Chesapeake Bay may receive greater than 50% of nitrogen loading from atmospheric deposition (Correll and Ford, 1982). Jaworski et al. (1997) estimated that 64% of the nitrogen export from NE US watersheds during 1990–1993 was derived directly or indirectly from NO_x emissions. Increased AAND to land-locked oceans such as the Baltic (Rodhe et al., 1980) and the Mediterranean (Bethoux et al., 1998) have driven broad-scale biogeochemical transitions in surface waters. Likewise, in downwind western oceanic basins Fanning (1989) attributed a shift in stoichiometric ratios of major macronutrients from nitrogen to phosphorus limitation as a result of increased AAND.

Considerable attention has been given to the role of excess nitrogen, particularly AAND, as a principal driver in the alteration of productivity patterns in terrestrial ecosystems, especially for European forests. Controversy, however, surrounds the potential role of these nitrogen additions in driving additional C fixation on the ecosystem or biome scale, particularly as a sink for the "missing carbon" from global biogeochemical budgets (Townsend et al., 1996). Clearer though, is the importance of AAND as it contributes to regional enhancement of productivity in coastal ecosystems. Nitrogen-limited coastal seas downwind of urban areas such as the Gulf Stream off of NC (Willey et al., 1988) appear particularly susceptible to enhancement of primary productivity by AAND (Paerl et al., 1990; Willey and Cahoon, 1991). A more enigmatic question is the role of AAND as its relation to primary production in marine waters remote from source production.

Beyond the obvious cases of anthropogenic nutrient enrichment from local sources, overgrowth by macroalgae has been reported on coral reefs in remote locations where sources of nutrient enrichment are not obvious (see Szmant, 2001). The Abaco Barrier Reef, near Green Turtle Cay, Bahamas (Fig. 1) has been characterized as a high-latitude reef system dominated by macroalgae (Lighty, 1982). Here, we report the significance of AAND on an oceanic coral reef environment, Green Turtle Cay, downwind from a major continental atmospheric source; and the first evidence that AAND significantly alters the productivity of coral reef macroalgae.

2. Methods

2.1. Analysis of wet deposition

Rainwater was collected from 15 rainfall events in a dry and wet season between January 18 and July 29, 2000 in clean polyethylene basins placed in an open field, <0.5 km from the meterological station on Green Turtle Cay (GTC), Abacos, Bahamas. Rainwater was
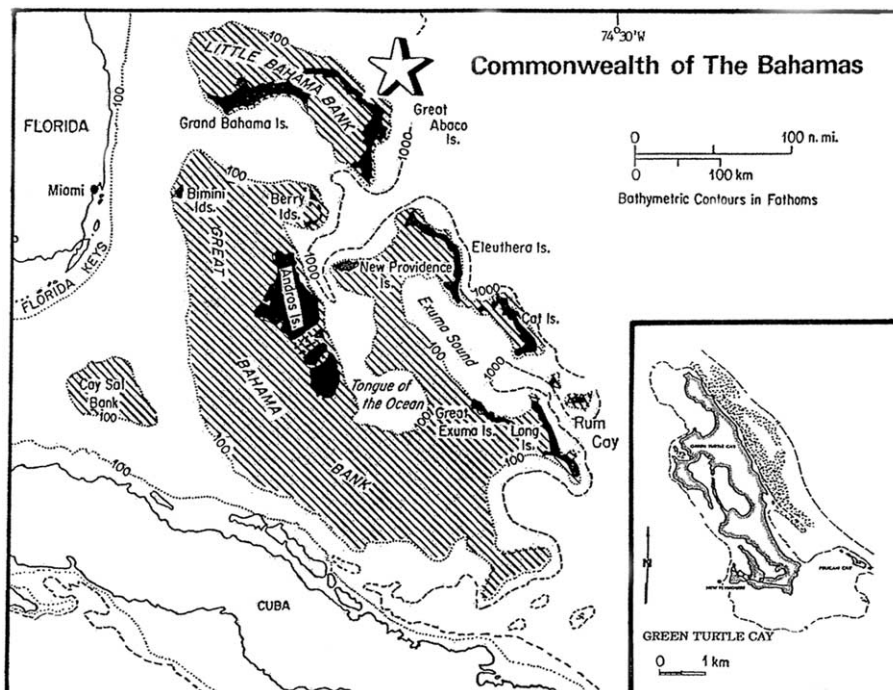
Fig. 1. Map of the Bahamas, including inset of Green Turtle Cay, and spatial reference to proximity of Florida on the mainland US.

immediately decanted into 500 mL polyethylene Nalgene™ bottles and frozen to stabilize the samples until analysis (see Dore et al., 1996). Samples were filtered to 0.25 μm using Gelman GF/F filters and analyzed with an Alpkem 500 series autoanalyzer for the following chemical species, including detection limits and method description, (Nitrate: 0.134 μM, Modified EPA method 353.2; Nitrite: 0.024 μM, Modified EPA method 353.2; Ammonia: 0.129 μM, Modified EPA method 350.1; SRP: 0.008 μM, Modified EPA method 365.1), at the HBOI Environmental Laboratory in Ft. Pierce, FL. Samples were also analyzed for pH using a Corning Model 240 pH meter with an Orion sure-flow electrode (EPA method 150.1).

2.2. Macroalgal productivity bioassays

A factorial productivity assay was designed to simulate field responses of dominant reef algae to rainfall nutrient enrichment. One gram (wet wt.) thalli of the conspicuous reef rhodophyte *Laurencia microcladia* were incubated for 1 h in 1 L Wheaton wide-mouth borosilicate bottles on 24 April 2000 in the following manner. A replicated 3×4 block design was utilized with the following treatments: (1) a *control* treatment with 1 μm filtered Sargasso seawater (FSS), (2) an *ammonium* treatment FSS with 2.5 μM NH_4^+ added, (3) an ammonium + SRP treatment with FSS with 2.5 μM NH_4^+ and 0.5 μM SRP, and (4) a *rainfall* treatment with FSS and 5% v/v (50 mL/L) rainwater. The rainwater used in this experiment was collected from

the 17 April 2000 rainfall event yielding 6.85 μM NO_3^-, 1.02 μM NH_4^+ and 0.02 μM SRP. Bottles were incubated in a clear 15 L polyethylene basin with flow-through seawater at 25 °C, and covered with shade cloth to maintain a light regime of 800–1200 μE m^{-2} s^{-1} to provide saturating light intensity while preventing photoinhibition during the assays. Thalli were similarly incubated in "dark" polyethylene 1L bottles using the identical nutrient treatment design to measure treatment effects on dark respiration. Oxygen production was measured to 0.01 mg/L with a YSI Dissolved Oxygen meter (Model 58) with a stirring probe, and converted to mg C g dry wt^{-1} h^{-1} using a photosynthetic quotient of 1.25. Blank bottles were incubated without algae to correct for "bottle effects" and production by unfiltered plankton and bacteria in the assay (Littler, 1979).

Treatment effects for net productivity and dark respiration were compared with one-way ANOVA after values were tested for homogeneity of variances around mean production values with an *F*-test. Multiple comparisons of groups within significant main treatment effects were measured with Fisher's Multiple Comparisons test.

2.3. Stable nitrogen isotope analysis of macroalgal tissue

Macroalgal tissue was collected from reef environments along a gradient from inshore waters of Abaco Island to an offshore reef site ∼3 km east on the barrier reef abutting the Sargasso Sea. At least 5 thalli of each species were collected from each site and combined as

a composite sample ($n = 2$) for analysis. Thalli were rinsed in deionized freshwater, dried at 60 °C for 48 h and homogenized with a mortar and pestle. Samples were analyzed for precision in replicate ($n = 2$) for $\delta^{15}N = [(R_{sample}/R_{atm}) - 1] * 10^3$ where $R = {}^{15}N/{}^{14}N$ and atm denotes atmospheric (N_2) gas as a standard. Atmospheric N_2 is used as a standard for $\delta^{15}N$ analyses as it provides a baseline value of 0‰ (Owens, 1987).

3. Results

3.1. Rainfall chemistry

Rainfall samples from 15 rain events were collected between 18 January and 29 July 2000 were analyzed for pH and nutrient chemistry. Concentrations of ammonia and nitrate ranged from 1.68 to 122 µM (median = 8.67 µM) and 0.61 to 137 µM (median = 15.9 µM), with means of 22.47 µM and 26.42 µM, respectively (Fig. 2). Concentrations for soluble reactive phosphorus (SRP) ranged from 0.24 to 20.20 µM (median = 0.36 µM), with a mean concentration of 2.31 µM (Fig. 3). Measurements of pH (Fig. 4) ranged from a low of 4.8 on 18 January, to a high of 7.2 on 17 April, with the lowest values in the winter months and the highest values between April and June. The average concentrations of ammonia (22.47 µM) and nitrate (26.42 µM) for this site, representing both dry and wet season, give a collective DIN wet deposition value of 22 µM DIN m^{-2} d^{-1}, or ~0.2 mg DIN m^{-2} yr^{-1} based on the 25 yr average rainfall of 1660 mm at Green Turtle Cay, Bahamas (Bahamas Meterological Department). Official wind direction observations (Bahamas Metero-

Fig. 3. Soluble reactive phosphorus values (in micromolar concentrations) in rainfall samples for 18 January–29 July 2000 at Green Turtle Cay, Bahamas.
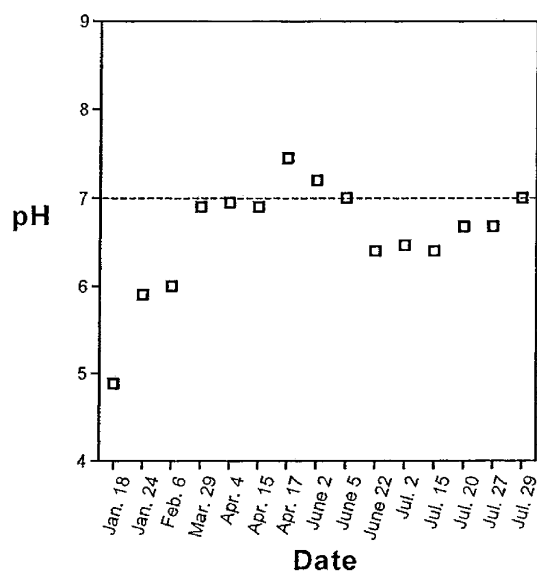
Fig. 4. pH values in rainfall samples for 18 January–29 July 2000 at Green Turtle Cay, Bahamas.

logical Department) accompanying these rainfall collections at Green Turtle Cay ranged from northwest to southwest with a dominant westerly vector.

3.2. Macroalgal productivity bioassay

3.2.1. Net primary productivity

Net primary productivity (NPP) of *L. microcladia* in the nutrient addition treatments was not significantly different ($p = 0.793$) than the control treatment (Table 1). Mean values of net primary productivity (mg C g dry wt^{-1} h^{-1}) for *L. microcladia* (Fig. 5) in the
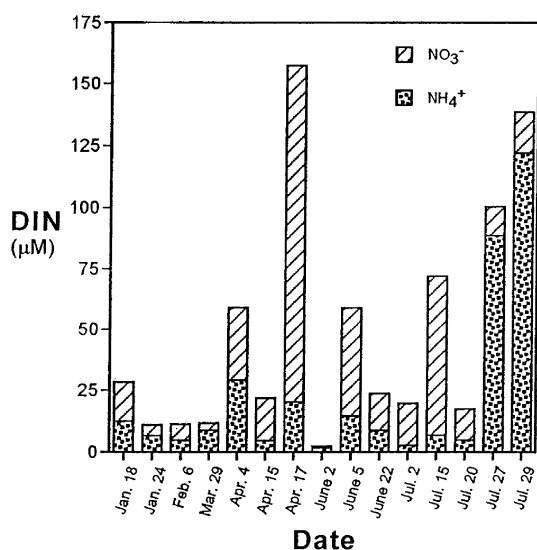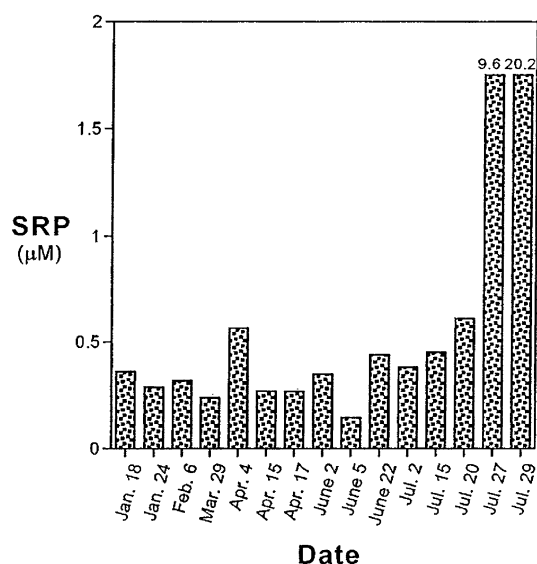

Fig. 2. Dissolved inorganic nitrogen (nitrate + ammonium) values (in micromolar concentrations) in rainfall samples for 18 January–29 July 2000 at Green Turtle Cay, Bahamas.

Table 1
Results of one-way ANOVA for net primary productivity (NPP) of *L. microcladia* from nutrient bioassay treatments

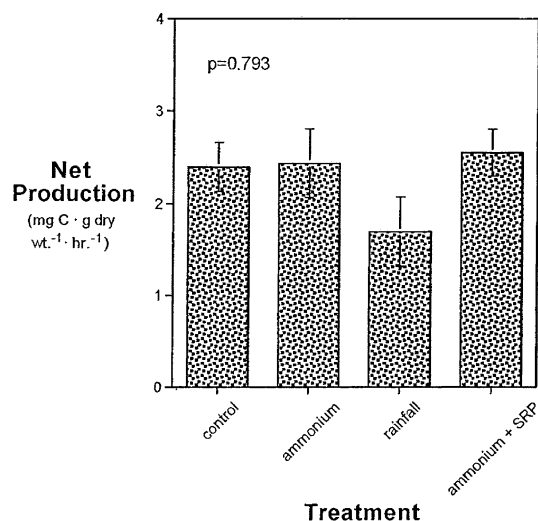| Source	df	MS	F	p
Laurencia treatments	3	0.013	0.0346	0.7934
Residual	8	0.039		

Fig. 5. Mean (±SE) net production values (mg C g dry $\mathrm{wt^{-1}\,h^{-1}}$) for *L. microcladia* in control, ammonium, rainfall, and ammonium + soluble reactive phosphorus treatment.

nutrient addition treatments were slightly elevated as compared to the *control* treatment with the exception of the *rainfall* treatment. The mean productivity values for the *ammonium* treatment (2.44 ± 0.37), the ammonium + SRP treatment (2.55 ± 0.25), were approximately equal to the untreated *control* group (2.40 ± 0.26), but the *rainfall* treatment (1.70 ± 0.37) had the lowest net productivity of any treatment group.

3.2.2. Dark respiration

For *L. microcladia*, significant main effects ($p = 0.011$) were detected for dark respiration (mg C g dry $\mathrm{wt^{-1}\,h^{-1}}$) among the treatments (Table 2, Fig. 6). Fisher's multiple comparisons test revealed that the respiration mean for

Table 2
Results of one-way ANOVA for dark respiration of *L. microcladia* from nutrient bioassay treatments

Source	df	MS	F	p
Laurencia treatments	3	0.040	9.365	0.0111
Residual	8	0.004		
Fisher's multiple comparisons				
Control -v- ammonium				0.0127
Control -v- rainfall				0.0029
Control -v- ammonium + SRP				0.0098

Fisher's multiple comparisons test for treatment means where main effects are significant ($p < 0.05$) for ANOVA.
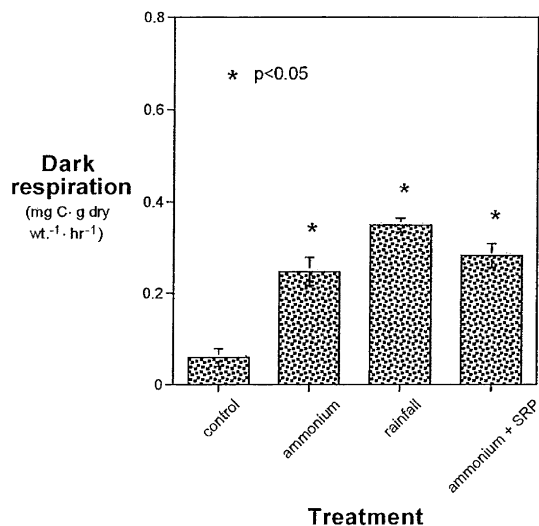

Fig. 6. Mean (±SE) dark respiration values (mg C g dry $\mathrm{wt^{-1}\,h^{-1}}$) for *L. microcladia* in control, ammonium, rainfall, and ammonium + soluble reactive phosphorus treatment.

the *rainfall* treatment (0.35 ± 0.02) was significantly higher ($p = 0.003$) than the mean for the *control* treatment (0.06 ± 0.02). Also, the means for the *ammonium* treatment (0.25 ± 0.03), and the ammonium + SRP treatment (0.28 ± 0.03), were significantly higher ($p = 0.013$; $p = 0.010$), respectively, than the *control* treatment. However, there was no significant difference in respiration between the three nutrient addition treatments.

3.3. Stable nitrogen isotopes from macroalgae

Stable nitrogen isotope (δ^{15}N) values for macroalgae along a spatial gradient from the mainland of Abaco Island to the offshore barrier reef are presented in Fig. 7. Macroalgal species in the sewage impacted Town Harbor of Green Turtle Cay (GTC) averaged ~+8‰. Macroalgae collected 10 m from the shore of Abaco Island averaged +3.5‰. However, at the seaward reef sites isotope values were considerably lower. At Bita Bay reef, within 0.25 km of GTC, macroalgae averaged +2.2‰, while at the offshore fringing reef, Golden Grotto (~3–4 km seaward of GTC), macroalgae averaged +1.8‰. The value for *L. microcladia*, measured from the sample group used in the productivity assay, averaged +2.1‰.

4. Discussion

4.1. Regional climatology and atmospheric nitrogen deposition

Atmospheric chemistry data are commonly used to discriminate atmospheric back trajectories by using isobaric gradients (Moody and Galloway, 1988). Climatological
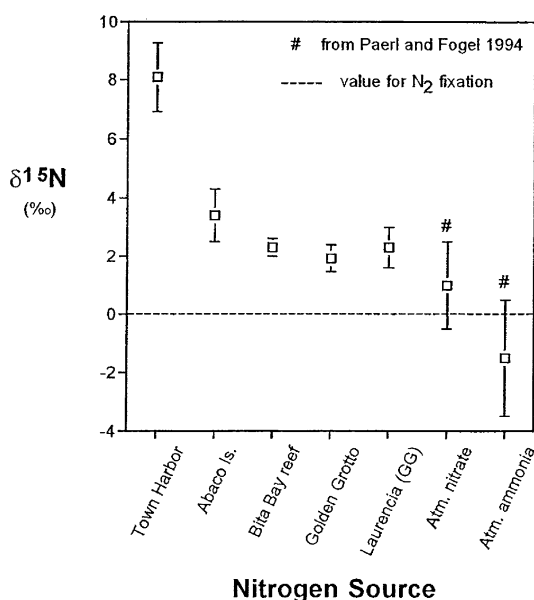
Fig. 7. Mean (±SE) stable nitrogen isotope values ($\delta^{15}N$) for macroalgae at Green Turtle Cay study sites, and previously reported atmospheric nitrogen sources in rainfall.

data (Miller and Harris, 1985) suggest a dominant eastward trajectory from Florida across the Bahama platform toward Bermuda in both cool (October–March) and warm (April–September) seasons. Several lines of evidence suggest a coupling between airsheds of the southeastern United States (particularly Florida) with the Bahamian archipelago platform. In both winter (dry) and summer (wet) seasons, local meteorological conditions associated with wet deposition events at Green Turtle Cay (GTC) indicate a predominate westerly wind flow pattern from the mainland US. There are two likely scenarios that support distinct seasonal mechanisms for the transport of atmospheric solutes from Florida to the northern Bahamas. First, meteorological events during winter months are dominated by continental high pressure fronts transporting cool air masses over peninsular Florida and subsequently, the Bahamas platform. Whelpdale et al. (1984) reported that the winter season climatological pattern at the surface (1000 mb contour) for the east coast of Florida produces a dominant latitudinal advection vector directed toward the northern Bahamas. Alternatively, during the summer, the Bermuda High dominates the climatology of the northern Bahamas and eastern Florida which elicits an antagonistic westerly land breeze that leads to seaward airmass transport during the evening and cooler morning hours of the summer months following daily convective heating on the Florida peninsula.

4.2. Significance of wet deposition to Green Turtle Cay

Of the 22×10^{12} g N yr^{-1} emitted from combustion sources to the global atmosphere, 94% is emitted in

the northern hemisphere (Hameed and Dignon, 1988), and over 80% is from land, hence, the impact on seas downwind from continents would be greatest (Duce et al., 1991). This study constitutes the first reported dataset of chemical analysis for rainfall from the Bahamian island archipelago. As a reference, however, Paerl et al. (1990) reported a ship-based rainfall collection near San Salvador Island, Bahamas of ∼3 µM nitrate (no reported ammonia), ∼2.8 µM orthophosphate, with a pH of ∼6.0. Dissolved inorganic nitrogen concentrations associated with rainfall collected at Green Turtle Cay (GTC) were episodically variable and did not show a seasonal trend. The ranges of 1.68–122 µM for ammonia and 0.61–137 µM for nitrate are similar to ranges for wet inorganic nitrogen species in rainfall events in other western Atlantic sites. The wet concentration values collected from Green Turtle Cay are similar to those collected at Beaufort, NC (Paerl and Fogel, 1994) for nitrate (0.7–144 µM) and ammonia (0.5–164 µM) from August 1990 to 1992. Owens et al. (1992) reported wet deposition of inorganic nitrogen at Bermuda to range from 0.7 to 1470 µM N m^{-2} per event. The average dissolved inorganic nitrogen (DIN = ammonia + nitrate) loading for Green Turtle Cay of 22 µM N m^{-2} d^{-1} is at the low end of the 16–50 µM N m^{-2} d^{-1} mean for Bermuda.

Presented in Table 3 are data on inorganic nitrogen deposition for several potential upwind sites from the Bahamas, including Miami, FL, Cape Canaveral, FL and Clinton, NC. Also included is an average for Bermuda, which is known to receive loadings from continental sources in the northeastern US. The closest "upwind" site to the northern Bahamas is the National Atmospheric Deposition Program (NADP, 2000) site on Cape Canaveral, FL (FL99, http://nadp.sws.uiuc.edu/sites/siteinfo.asp). As a result of the upwind spatial position, this location represents a reasonable site to provide a quantitative perspective of the southeastern US as a source of reactive nitrogen. The following data support the precept that the Cape Canaveral NADP site represents the "upwind" Florida peninsula as a probable airshed source. The average annual pH has been on a

Table 3
Reported values for nitrogen deposition at western Atlantic locations

Site	Atmospheric N input (µM N m^{-2} d^{-1})	Reference
Sargasso Sea	16–50[a]	Knap et al. (1986)
Bermuda	30	Galloway et al. (1989)
Miami, FL	52	NADP (2000)
Cape Canaveral, FL	86	NADP (2000)
Coastal NC	101[a]	Willey et al. (1988)
Clinton, NC	114	NADP (2000)
Bahamas	49	Prospero et al. (1996)
GTC, Bahamas	22[a]	This study

[a] Wet deposition only.

significantly decreasing trend for almost 2 decades from ~pH 5.0 to about 4.6. This decrease ironically has coincided with federal Clean Air Act mandates for stringent controls on sulfur dioxide emissions from industrial sources. Alternatively, unchecked increases in the emissions of nitrogen oxides are now playing a more prominent role in the acidification of lakes in the northeastern American continent (Brezonik et al., 1980; Galloway et al., 1984b; Vitousek et al., 1997a). In north-central Florida, sulfate and nitrate concentrations in rainfall increased by a factor of 1.6 and 4.5, respectively, from 1965 to 1980 (Brezonik et al., 1980).

At the Cape Canaveral NADP site, average annual deposition rates for ammonia and nitrate have increased significantly over the last two decades. For ammonia, deposition has increased 3-fold from an average value of ~0.5 kg/ha in the early 1980s to over 1.5 kg/ha in 1999. For nitrate, deposition has at least doubled, from a value of ~5 kg/ha in 1984 to over 10 kg/ha by 1999. The total deposition of inorganic nitrogen for 1999 for the Cape Canaveral NADP site is ~12 kg/ha or 86 μM N m^{-2} d^{-1} for biologically reactive inorganic N species. This value represents a loading estimate ~4× that of our estimate for Green Turtle Cay. With elevated atmospheric DIN loadings at an upwind position, the airmass from eastern Florida represents a likely source to the northern Bahamas. The loading estimate of 22 μM N m^{-2} d^{-1} for Green Turtle Cay also agrees well with Prospero et al.'s (1996) model value for the deposition of nitrogen to the North Atlantic basin. For the 10° grid overlying the Bahamas, the model predicts a deposition value of ~50 μM N m^{-2} d^{-1} for inorganic nitrogen species.

Increased acidity (i.e. lowered pH) is a reliable signature of rainfall contaminated with anthropogenic emissions. Galloway et al. (1984b) suggest that sulfate ion is the most conservative species contributing to acidification of rainfall. Indeed, in Bermuda concentrations of non-sea salt sulfate and nitrate are about a factor of three times higher than those in remote marine areas of the world, and appear to be responsible for acidity in Bermuda's precipitation. Church et al. (1982) reported that air-masses with continental origin lead to increased acidity in rainfall on Bermuda. Other analyses of marine rain support this contention. Willey et al. (1988) reported higher hydronium ion, non-sea salt sulfate ion, and nitrate concentrations associated with continental-based storm-fronts as compared to storms of coastal or oceanic origin in Wilmington, NC. The lowest pH value (4.82) was recorded in rainfall associated with a passing cold front on 18 January 2000. Higher values to near and above neutral pH (6.9–7.5) were recorded from March 29 through the summer sampling period. The lower value of pH 4.82 associated with the continental front agrees precisely with the mean value of 4.82 for winter 1999 at the Cape Canaveral NADP site.

The emerging importance of atmospheric nitrogen to the Bahamas should not go unrecognized. The increase in population, industrialization and subsequent rises in nitrogen deposition in the southeastern United States (particularly, Florida) have obvious relevance to deposition patterns in the Bahamian archipelago. Galloway et al. (1994) predict an 80–100% increase in deposition of nitrogen to the Bahamas between 1980 and 2020.

4.3. Atmospheric nitrogen alters carbon fixation of reef macroalgae

This report constitutes one of the first documented scenarios of the consequences of atmospheric nitrogen transport to changes in metabolic uptake and productivity processes in a remote oligotrophic ecosystem. Here, we provide evidence for a source-sink transport and subsequent metabolic consequence of nitrogen addition to a dominant macroalgal bloom species from the Abaco barrier reef. Macroalgal nutrient enrichment bioassays utilizing rainfall produced predictable responses for nutrient limited macroalgae in oligotrophic waters. Rainfall stimulated dark respiration in *L. microcladia*, while net photosynthesis was not enhanced by either the rainfall or synthetic nutrient additions. For *L. microcladia*, respiration rate of the rainfall treatment, although significantly higher than controls, was similar in magnitude to the synthetic ammonium, and ammonium + reactive phosphorus addition treatments. These results suggest that ammonium played a critical role in the increased dark respiration. The enhancement of dark respiration by nitrogen-limited algae during ammonium assimilation has been known for quite some time in the phytoplankton literature (see Syrett, 1953; Morris et al., 1971), and has also been demonstrated for symbiotic dinoflagellates in corals (Cook et al., 1994). Likewise, macroalgae are known to increase dark respiration in response to ammonium enrichment, with examples from tropical macroalgae (Littler et al., 1988), including the frondose rhodophyte *Gracilaria tikvahiae* (Lapointe et al., 1984). Healey (1979) suggests that increased dark respiration is a more common response than increased net photosynthesis in the first few hours following nutrient addition.

Alternatively, net photosynthesis for *L. microcladia* in the rainfall treatment was apparently, but not significantly, suppressed compared to controls. This is likely a response to the high nitrate content from the rainfall used in this assay, whereas ammonium and ammonium + reactive phosphorus treatments were not suppressed. Differential uptake of nitrogen species, in light versus dark conditions (see D'Elia and DeBoer, 1978), are based upon lower energetic demands for ammonium uptake and assimilation. In contrast, uptake of nitrate is a light dependant reaction (MacIssac and Dugdale, 1972; Falkowski and Stone, 1975) where active

transport is necessary to assimilate and reduce nitrate, whereas ammonium does not have to be reduced for uptake during photosynthesis (Turpin, 1991). Nitrate uptake during light reactions competes with carbon fixation for ATP, and this has also been reported previously for the macroalgae *Codium fragile* (Hanisak and Harlin, 1978) and *Ulva fasciata* (Lapointe and Tenore, 1981). It is likely that *L. microcladia* would respond with higher net production when rainfall treatments are dominated with ammonium as compared to the nitrate, the dominant DIN species found in rainfall samples used in this study.

Short-term bioassays, like those utilized here, can provide a quick snapshot of the integration of limiting nutrients into algal biomass (e.g. Lapointe et al., 1987), and subsequent stimulation of productivity. The photosynthetic responses of the reef macroalga *L. microcladia* utilized in these assays are consistent with the metabolic responses of other algal taxa in similar conditions. In fact, there is substantial evidence from the physiological literature that suggests the metabolic response of a net or gross photosynthetic "lag period" following such enrichments. As described by Healey (1979), nutrient-deficient algae typically have a disposition for high nutrient uptake rates, which can potentially compete with light reactions of photosynthesis for ATP and reductant. In general, nutrient deficient algae are usually high in carbohydrate (see Lapointe and Tenore, 1981), or lipid, so there is no need for an increased rate of carbon fixation soon after addition of a deficient nutrient. To the contrary, metabolism should first be directed toward nutrient uptake, and subsequently toward carbon fixation (utilizing the new N and P subsidy) to support increased growth.

4.4. Ecosystem responses

Tropical benthic marine ecologists are currently debating the mechanisms underpinning the seemingly circumtropical emergence of macroalgal communities as a new dominant "stable state" in coral reef ecosystems. The symbiotic relationships underpinning the development of hermatypic coral reefs evolved under oligotrophic conditions to maximize the cycling of nutrients between the zooxanthallae and host coral polyp (Muscatine and Porter, 1977), and on an ecosystem scale between habitats within coral reefs (see D'Elia and Wiebe, 1990; Barile, 2004). These symbioses are ecologically advantageous under low and often undetectable concentrations of dissolved nutrients, which limit the production of potentially competitive endolithic turfs (Odum and Odum, 1955) and frondose macroalgae which now dominate many of the world's reef ecosystems (Done, 1992; Hodgson, 1999).

Capone et al. (1977) estimated that 1.8 g N m^{-2} yr^{-1} (or 5 mg N m^{-2} d^{-1}) is derived from nitrogen-fixing epi-

phytes associated with macroalgae on western Bahamian reefs. Episodic rainfall events may produce an equally important loading of $\sim$5 mg N m^{-2} d^{-1}, making the contribution of nitrogen deposition from rainfall roughly 50% of the "new" nitrogen source. The daily nitrogen demand for macroalgae at the Abaco barrier reef site, based upon macroalgal biomass and production estimates from Barile (2001), is $\sim$24 mg N m^{-2} d^{-1}. Episodic rainfall events can potentially meet about 20% of this overall nitrogen demand. These results suggest the overall importance of nitrogen deposition to productivity "down-wind" from the North American continent, and suggest a revised assessment of the importance of AAND to the nearby Sargasso Sea, that Knap et al. (1986) previously deemed as "insignificant."

An emerging role of atmospheric nitrogen deposition is the "saturation" (Aber et al., 1989) of these upstream nitrogen-limited terrestrial ecosystems. Coastal ecosystems reaching the threshold of maximum nitrogen storage lose the capacity to retain additional burdens of nitrogen (sensu "nutrient retention hypothesis" Vitousek and Reiners, 1975) and subsequently "leak" excess nitrogen into tributaries, estuaries and coastal oceans. Land-based mobilization of nitrogen is problematic in tropical island ecosystems dominated by calcium carbonate or limestone geology with high hydraulic conductivity, leading to rapid movement to coastal waters via discharging groundwater from freshwater aquifers (see Marsh, 1977). In the Chesapeake Bay region, watershed export of inorganic nitrogen is tightly coupled in a linear fashion with increasing NO$_x$ deposition, with a mean of $\sim$65% of exported N derived from atmospheric deposition (Jaworski et al., 1997). That report suggests the "deposition saturation flux" is consistent with nitrogen saturation of upland watersheds. With regard to the northeastern Bahamas, we suspect that chronic watershed saturation of the mainland Abaco Island (see Fig. 1), in addition to direct deposition, are important mechanisms for the mobilization of atmospheric nitrogen to the reefs seaward of Green Turtle Cay. Direct deposition to shallow ($\sim$1–10 m) reefs is likely as a result of the small dilution factor of surface rainfall. For shallow estuaries such as the Albemarle-Pamlico Sound, NC atmospheric deposition has been demonstrated to stimulate phytoplankton communities (Paerl et al., 1990).

It is likely, however, that a more chronic and significant source is the result of deposition to the Abaco Island mainland, and subsequent "leakage" of accumulated nitrogen into aquifers, surface waters, and advection to the reef by tidal flux. Lighty (1982) attributes the demise of the Abaco barrier reef and the establishment of the fleshy-algal community to "off-bank transport of turbid and episodically cooled water masses from storm events." Alternatively, the off-bank transport reported by Lighty (1982) has probably become

saturated in the past several decades with dissolved inorganic nitrogen, from local and far-field sources. Nitrogen-laden tidal flows are known to drive bioerosional processes on adjacent portions of reef tracts such as the Florida Keys (Lidz and Hallock, 2000). Further, nitrogen enrichment phenomena have been linked to the instability and degradation of coral reef ecosystems across broad geological scales (Hallock, 1986, 1988).

Nitrogen fixation by both pelagic and benthic cyanobacteria has been recognized as an important source of "new" nitrogen to hermatypic coral reefs and associated reef flats (Odum and Odum, 1955; Wiebe et al., 1975; Capone, 1983). However, high water column and intercellular concentrations of both ammonium (Patriquin and Knowles, 1975; Evans and Barber, 1977) and nitrate (Dicker and Smith, 1980) are reported to inhibit nitrogen fixation at high DIN concentrations ($\sim$10 μM, Elmetri, 2003), as available extracellular inorganic nitrogen species compete for reducing power by the assimilatory nitrate reductase pathway. Hence, increasing nitrogen from rainfall (and other land-based sources) could inhibit nitrogen fixation or augment nitrogen availability in shallow reef ecosystems, and further accentuate the importance of rainfall nitrogen to remote coral reef ecosystem nitrogen budgets.

Stable nitrogen isotope values from macroalgae on the shallow barrier reef near Green Turtle Cay ($\sim$2.0‰, Fig. 7) are elevated as compared to values expected from reef algae receiving exclusively fixed nitrogen as a nitrogen source (see France et al., 1998). As the mean δ^{15}N value for the bloom forming rhodophyte *L. microcladia* is elevated above a N-fixation value at +2.1‰, it is likely that macroalgae and associated N-fixing cyanobacteria on the Abaco barrier reef are: (1) receiving anthropogenic nitrogen from either direct deposition or nitrogen "leaked" from landward watersheds, as evidenced by increasing δ^{15}N for macroalgae located in a gradient closer to the mainland, and (2) directly assimilating nitrogen from the water column, rather than depending upon energetically expensive N-fixation. The results of this study provide evidence for an additional biogeochemical pathway that may alleviate primary nutrient limitation of macroalgal production on a barrier reef ecosystem that borders the otherwise oligotrophic Sargasso Sea. The mobilization of nitrogen to remote ecosystems via atmospheric pathways provides further evidence of the magnitude of the consequences of human modification of the global biogeochemical cycle of nitrogen.

Acknowledgements

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