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Reconstruction of paleostorms and paleoenvironment using geochemical proxies archived in the sediments of two coastal lakes in northwest Florida

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A B S T R A C T

Late Holocene paleoclimate records from coastal regions are important for understanding long-term variability of hurricane activity. Here we present a nearly 4000-year record of severe storm landfalls and environmental changes based on organic geochemical proxies (OGPs) preserved in sediment cores from two coastal lakes in northwest Florida. Our analysis shows that there are significant variations in $\delta^{13}C$, $\delta^{15}N$, C%, N% and C/N with depth, reflecting changes in lake environment, which in turn affected the processes delivering water and sediment to the lake as well as biological productivity within the lake. Isotopic signatures of modern organic materials in the lakes and their surrounding areas show that the major sources of sedimentary organic matters in the lakes are aquatic and terrestrial C_3 vegetation. C_4 grasses do not contribute significantly to the sedimentary organic matters in the lake, although they can be found in the mostly forested watershed. Thus, the positive C and N isotopic shifts, concurrent with negative shifts in C/N ratios, most likely indicate shifts to a marine-like environment in coastal lakes following the influx of marine water and nutrients and marine biota associated with major storm events. Some of these isotopic shifts observed in the sediment cores correspond to visible sand layers presumably representing overwash deposits associated with severe storm events. Radiocarbon dating of bulk sediment organic matters, wood fragments and shells indicates that the sediment in these cores was deposited over the last 3000–4000 years. Based on our age model and OGP interpretation, Eastern Lake data suggest that the recurrence interval of severe storms (i.e., large enough to cause seawater flooding of the lakes) is approximately 84 years over the last 2900 years, whereas Western lake data suggest an average recurrence interval of 86 years in the past 3900 years.

1. Introduction

In recent years, several tropical systems struck the U.S. Gulf Coast, causing considerable damage to property and loss of human life in densely populated cities along the Gulf Coast. A general trend towards increasing hurricane activity and intensity has been observed since 1923 (Goldenberg et al., 2001; Grinsted et al., 2012). A number of scientists have recently suggested the observed trend toward increasing tropical system intensity can be correlated with increases in ocean surface temperature (Elsner et al., 1999, 2008; Enfield et al., 2001; Emanuel, 2005; Trendberth, 2005; Webster et al., 2005; Landsea et al., 2006; Wang and Lee, 2010; Grinsted et al., 2012). These scientists argue that as oceans warm, more kinetic energy is available to be converted to tropical cyclone wind and thus may provide more favorable conditions for developing stronger storms (Elsner et al., 2008).

Historical records reveal that hurricane activities vary widely on interannual and multidicadal time scales, which may be directly or indirectly correlated to several climatic phenomena such as Atlantic Multi-decadal Oscillation (AMO), temporal shifts in the position of the Bermuda high and El Nino-Southern Oscillation (ENSO) (Elsner and Kara, 1999). For example, the more southwesterly position of the Bermuda High would result in more hurricane landfalls on the Gulf of Mexico Coast instead of the Atlantic Coast (Liu and Fearn, 2000). Although, the frequencies of major hurricanes are not directly correlated with AMO, it has been suggested that the number

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of minor hurricanes of category 1 and 2 is increased during the warm phase of AMO than in the cool phase (Chylek and Lesins, 2008). However, significant variations in both hurricane activity and ocean surface temperature on long time scales make it difficult to establish any long-term trend with certainty (Elsner et al., 1999, 2000; Trendberth, 2005; Webster et al., 2005; Landsea et al., 2006; Vecchi and Soden, 2007; Emanuel et al., 2008; Wang and Lee, 2010). Unfortunately, the 100 to 150-year long instrumental record is too brief to detect long-term trends or to clearly distinguish between natural and human induced effects on tropical cyclones (Elsner et al., 1999; Trendberth, 2005; Landsea et al., 2006). Therefore, reconstructing a reliable paleostorm history has been recognized as important to understanding the possible link between future climate change and increasing storm intensity and frequency (Emanuel, 2006; Landsea et al., 2006; Emanuel et al., 2008).

Proxy-based paleostorm reconstruction has a number of advantages over the instrumental record as it allows reconstruction of storm activities on millennial or longer time scales and thus can provide a longer record of storm activities (Chappell et al., 1983; Liu and Fearn, 1993; Elsner et al., 2000; Donnelly et al., 2001a, b; Liu et al., 2001; Donnelly et al., 2004; Donnelly and Webb, 2004; Lambert et al., 2008). Long-term records of storm activities can help constrain models in predicting recurrence interval of landfalling large storms in coastal areas in the near future (Liu and Fearn, 1993, Elsner et al., 2000, Frappier et al., 2007).

In this study, three sediment cores from two coastal lakes in northwest Florida were analyzed for their organic geochemical signatures including their carbon (C) and nitrogen (N) isotopic compositions, C%, N% and C/N ratios. Radiocarbon ages of selected sediment, wood and shell samples were also determined in order to establish a chronological framework for the interpretation of the geochemical data. In addition, the C and N isotopic compositions of plants and soils from the lake areas as well as dissolved organic matter and particulate organic matter in the lakes were analyzed. The data were used to reconstruct a record of changes in lake environment and paleostorm activity over the last ~3900 years in the area.

2. Background and paleostorm records for the Gulf of Mexico Coast

The most common method for reconstructing the frequency of landfalling paleohurricanes is to identify and count overwash sand deposits in coastal lakes or ponds (Liu and Fearn, 1993; Liu et al., 2008). According to Liu and Fearn (2000), each sand layer deposited by overwash process represents a storm event. They postulate that the size and thickness of an overwash sand layer is proportional to the storm intensity. Thus, larger storms (i.e., category 4–5) produce larger overwash deposits, resulting in thicker sand layers in the sedimentary record (Liu and Fearn, 2000).

This method has been applied to two coastal lakes along the northern Gulf Coast: Lake Shelby in Alabama and Western Lake in northwest Florida, with the interpretations of these studies suggesting a recurrence interval of 600 years and 280 years for major storms for these two lake areas, respectively (Liu and Fearn, 1993; 2000). The underlying assumptions associated with this method are: (1) sand layers are derived exclusively from the Gulf of Mexico during storms; (2) sand layer distance from the lake shoreline is proportional to the storm intensity, and (3) radiocarbon dates bracketing the sand layers (and uncorrected for reservoir effects) yield a reliable chronology. The validity of these assumptions is, however, difficult to evaluate without a good understanding of the lake setting and dynamics and in particular the nature and spatial distribution of the lake sediments (Otvos, 1999; 2002). A recent study found substantial ^{14}C deficiencies in Lake Shelby, highlighting the problem of reservoir effects on ^{14}C dating of coastal lake sediments (Aharon and Lambert, 2009). Thus, recurrence intervals based on uncorrected ^{14}C dates on coastal lake sediments are not accurate if the reservoir effect is a significant factor as observed in Lake Shelby.

Recent studies have demonstrated that geochemical proxies, including $\delta^{13}C$ and $\delta^{15}N$ in lacustrine sediments, $\delta^{18}O$ in tree rings, or $\delta^{18}O$ in speleothems, provide a valuable tool for reconstructing high-resolution paleostorm records (Lambert et al., 2003; Lambert et al., 2008; Frappier, 2009; Miller et al., 2006). Lambert et al. (2008) used organic geochemical proxies (OGPs) in sediment cores from Lake Shelby, to reconstruct a millennia-long record of landfalling severe storms that inundated the coastal lake with marine water. The Lambert et al. (2008) study suggested a recurrence interval of ~62 years over the past millennia, which is an order of magnitude greater than the ~600 year recurrence interval proposed by the overwash sand layer record by Liu and Fearn (1993). To increase sensitivity and confidence in paleostorm identification, Lambert et al. (2008) considered positive shifts in $\delta^{13}C$ and $\delta^{15}N$ in lake sediments as indicative of seawater inundation during catastrophic storm surge, because organic matters of marine origin are generally more enriched in the heavy isotopes ^{13}C and ^{15}N than those of freshwater origin or those derived from terrestrial plants (Lambert et al., 2008). Proxy records based on overwash deposits likely only record very large hurricanes of category 4 or larger while the OGPs appear to have recorded severe storms that were large enough to cause seawater flooding of the coastal lake (Lambert et al., 2008). Although the OGPs are likely a more sensitive indicator of storm events than overwash deposits and can detect storms where overwashed sand is not visible, the method has not been further tested or applied to similar coastal lakes.

In this study, the OGPs approach (Lambert et al., 2008) was applied to sediment cores from Eastern Lake and Western Lake on the Gulf Coast of northwest Florida in an effort to reconstruct the paleostorm history for the area. The results are compared with those from the traditional method used previously for Western Lake (Liu and Fearn, 2000).

3. Study sites

3.1. Eastern Lake

Eastern Lake (30°18′41.857″ N, 86°5′35.647″ W) is located within 200 m of the NW Gulf Coast of Florida (Fig. 1). The lake occupies 20.8 ha. The average water depth of the lake is approximately 3 m (Florida Lakewatch, 2008). The lake is stratified throughout the year. From 2001 to 2008, the average salinity of the lake water was 12.9 ppt at the bottom and 10.6 ppt at the surface, and the average pH was 7.5 (Florida Lakewatch, 2008). For brief periods of time the lake has been connected to the Gulf of Mexico through a small outlet following large storms. The Eastern Lake shoreline is moderately developed. The lake area has relatively flat topography. A small highway bridge separates the lake into north and south lake segments. The most abundant plant species in the lake shore area is black needlerush. Other plants found in or around the lake are widgeon grass, cordgrass, saltbush, water-pennywort, big rose mallow, green algae, salt marsh fringerush and giant bulrush (Florida Lakewatch, 2008).

3.2. Western Lake

Western Lake (30°19′37″ N, 86°9′4″ W) is located in Grayton Beach State Park, along the northwest Gulf Coast of Florida (Fig. 1). The lake covers ~37.6 ha and is separated from the Gulf of Mexico by 150–200 m wide, well-developed sand dunes, as much as 9.3 m

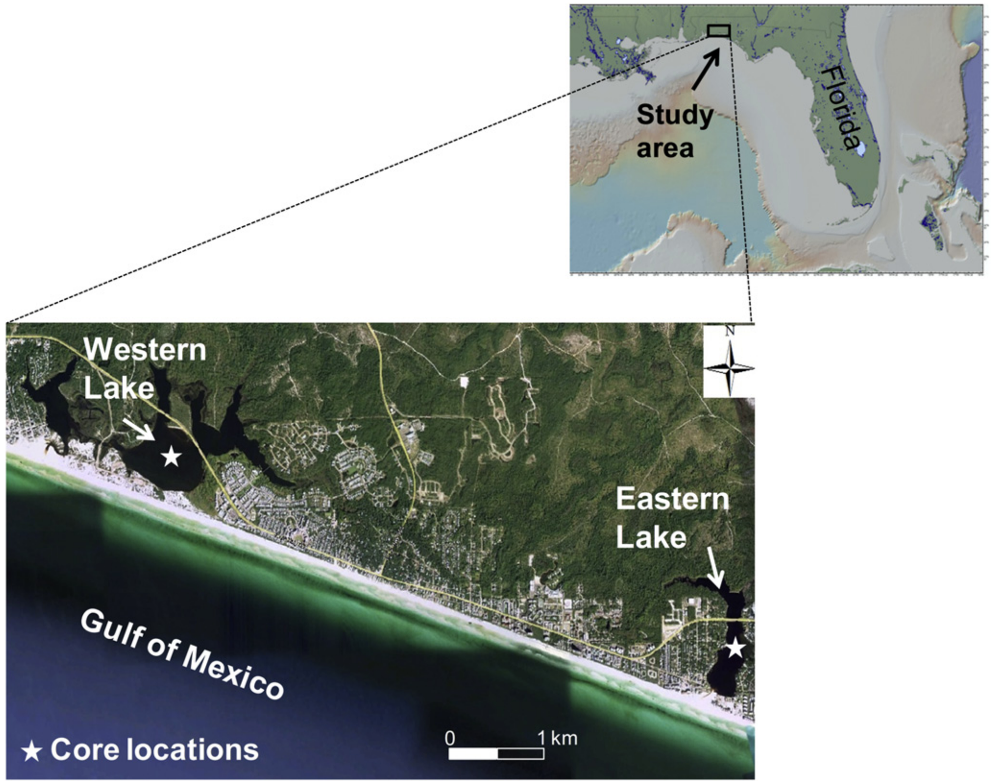

Fig 1. The location of Eastern Lake and Western Lake in Florida. The coring sites are marked by white stars. Base images from http://www.geomapapp.org (Ryan et al., 2009) (upper) and Google Earth (lower).

in height (Liu and Fearn, 2000). The average water depth of the lake is 3.3 m. The lake is stratified throughout the year. From 2001 to 2008, the average salinity of the lake water was 10.9 ppt at the bottom and 6.7 ppt at the surface, and the average pH was 7.3 (Florida Lakewatch, 2008). The lake has an outlet slough which temporarily connects it to the Gulf during high water events. Black needlerush and duck-potato are the most common plant species on the lake shoreline area. Other plants include sawgrass, cattail, big rose mallow, water-pennywort, water lily and tape grass (Florida Lakewatch, 2008).

4. Materials and methods

4.1. Collection and sampling of lake sediments for geochemical analysis

Two sediment cores (052209-02, 0.6 m and 052209-03, 1.03 m) from Eastern Lake and one sediment core (052109-03, 1.61 m) from Western Lake were collected from the center of each lake (Fig. 1). Piston push cores were used to collect the samples in order to prevent disturbance of lake sediments. Coring tubes were held vertical to keep sediment integrity while coring the sample from the lake and afterward were sealed at both ends of the tubes. These cores were sub-sampled using clean plastic coffee straws at 3–4 mm intervals for analyses of their organic carbon (C) and nitrogen (N) concentrations as well as δ^{13}C and δ^{15}N signatures (Lavoie et al., 1996). A total of 556 sediment samples were obtained from the Eastern Lake cores and 600 samples from the Western Lake core for geochemical analyses. Samples were freeze-dried, ground and homogenized into fine powder, and checked for the presence of carbonate with dilute (5%) HCl. Samples from Eastern Lake did not contain detectable amounts of carbonate and were directly wrapped into tin cups (5–100 mg) for isotope analysis. Samples from Western Lake were subjected to HCl fumes to remove carbonate from the samples following the method described in Harris et al. (2001). About 5–100 mg of each sample was weighed into a silver cup and wet with de-ionized water, and then placed, along with a beaker containing concentrated HCl (12 N), in a glass desiccator for 24 h. The carbonate-free samples were dried overnight at 70 °C in an oven. Dried samples were wrapped in ultra-clean tin cups for isotope analysis.

4.2. Processing DOM, POM and plants for isotope analysis

Dissolved organic matter (DOM) and particulate organic matter (POM) samples were collected from Eastern and Western Lake for isotope analysis. Water samples for DOM analysis were filtered through 0.45 μm pre-combusted glass filters in the field into clean 1 liter bottles using a small pump. The filtered water samples were freeze-dried to reduce the volume to less than 100 ml and then acidified with ~50–100 μl 6 N HCl. After acidification, the DOM samples were completely freeze-dried. The freeze-dried samples were then wrapped into tin cups for isotopic analysis. POM samples were collected on pre-combusted glass filter papers in the field. The glass filter papers bearing the POM were dried in an oven at 60–70 °C. Dried filter papers were cut into small pieces and wrapped into tin cups for elemental and isotopic analysis.

Plants were collected from Western Lake and its watershed as well as along the edges of Eastern Lake. Plant samples were cleaned with de-ionized water and dried in an oven at 60–70 °C. Approximately 1–5 mg of each dried plants were ground into powder for analysis of their isotopic signatures and bulk organic C and N contents.

4.3. Mass spectrometry analysis

The samples were analyzed for their C and N isotopic ratios using a Carlo Erba Elemental Analyzer (EA) connected to a Finnigan

MAT Delta Plus XP stable isotope ratio mass spectrometer (IRMS) through a Conflo III interface at the Florida State University. Approximately, 25% replicated samples were also analyzed. The isotope results are reported in the standard delta (δ) notation in per mil as $\delta^{13}C$ values with reference to the international VPDB standard and $\delta^{15}N$ values with reference to AIR (Faure and Mensing, 2005). The precision of the C and N isotope analysis is ± 0.2‰ or better on the basis of repeated analysis of five different laboratory standards including YWOMST-1 (cane sugar), YWOMST-2 (phenylalanine), YWOMST-3 (L-phenylalanine), YWOMST-5 (urea) and Urea-2. The analytical precision is 3% and 1% for C% and N%, respectively, based on repeated analysis of the same laboratory standards.

Radiocarbon ages of selected samples were measured at the National Ocean Science Accelerator Mass Spectrometry (AMS) Facility in Woods Hole Oceanographic Institution in Massachusetts and the Keck Carbon Cycle AMS Facility at University of California in Irvine, California.

5. Results

5.1. Lithologic units

The Eastern Lake sediment cores consist of three specific lithologic units: dark colored clay, gray colored silt, and light grey colored sand units. The clay unit is organic-rich, lacks visible beddings structure and does not show obvious signs of bioturbation (sand burrows). The grey colored silt unit contains mainly silt but also sand and clay and is organic-poor (with a few wood fragments) and is observed at different depths of the cores. The unit showed signs of bioturbation at several depths in the form of sand burrows. Sand laminae were found at various depths of the cores, varying in thickness from 2 to 30 mm. They were light grey and organic-poor. Both cores from Eastern Lake lacked carbonate shell fragments.

The Western Lake core mainly consists of two clay and sand lithologic units. The clay unit is dark brownish grey in color, fine grained, organic-rich and largely composed of clay with a small amount of silt. The relative proportion of clay and silt varied throughout the cores. The units did not show visible bedding, but showed evidence of bioturbation in the form of scattered sand pockets. The unit often contained root and/or wood fragments, shell hash, and/or shell fragments. The clay unit was interbedded with sand lamina which varied in thickness. The sand unit is grey colored, medium to coarse grained and organic-poor. The unit was characterized by numerous inarticulated and articulated shells (bivalve) and shell fragments.

5.2. Radiocarbon chronology

Age models were developed for the sediment cores based on radiocarbon dates of selected bulk sediment organic matter, wood fragments and shells from the cores (Table 1, Fig. 2). Radiocarbon dates were converted to calendar years using the on-line calibration software of Calib 6.1.0 Calibration Curve (Stuiver and Reimer, 1993; Stuiver et al., 2005). For Eastern Lake core 052209-02, three radiocarbon dates of bulk sediment organic matter (OM) were obtained from the top, middle and bottom layers of the core as no wood or shell samples were found in the core (Table 1). The ^{14}C date of the topmost layer of the core was 985 years BP, which is considered as reservoir age to correct the radiocarbon dates of bulk sediment OM samples from core 052209-02 (Fig. 2A, Table 1). The age models for Eastern lake core 052209-03 was based on ^{14}C dates of four wood samples clustering between 85 and 90 cm depth as no wood sample was available at the top part of the core (Fig. 2B).

Table 1
Radiocarbon ages of bulk sediment organic matters, wood fragments and shells in sediment cores for Eastern Lake and Western Lake, Florida.

Core ID	Depth in core (cm)	^{14}C age (yr BP)	Analytical error (yr BP)	Corrected age (yr BP)[d]	Calendar age (yr BP)[e]	Model age (yr BP)[f]
052209-02	0					80
	0–0.3[a]	985	±35	0	−59	92
	20[a]	2440	±35	1455	1345 ± 41	858
	23.7					1000
	Sand layer (55.3–56.3)					2250
	57.4[a]	3010	±35	2025	1976 ± 50	2312
	60					2413
052209-03	0					−65
	36.6					1000
	85[b]	2360	±20		2354 ± 11	2407
	Sand layer (85–88)					2450
	88–90[b]	2445	±20		2482 ± 107	2494
	90–92[b]	2530	±20		2625 ± 68	2552
	92–94[b]	2520	±20		2594 ± 41	2610
	103					2930
052109-03	0					49
	8[b]	380	±15		470 ± 25	240
	39.8					1000
	49.5[b]	1110	±15		1010 ± 27	1231
	83.5[c]	2285	±20		2330 ± 19	2043
	94.3–94.5[c]	2340	±20		2348 ± 6	2306
	94.5–95[c]	2350	±20		2351 ± 7	2318
	121[c]	2635	±20		2755 ± 9	2939
	122.5[c]	2795	±20		2898 ± 26	2975
	140.3					3400
	160					3871

[a] Radiocarbon ages were obtained from bulk organic matters in lake sediments.
[b] Radiocarbon ages were obtained from wood fragments present in the cores.
[c] Radiocarbon ages were obtained from carbonate shells present in the core.
[d] Corrected for "reservoir effect" (see text for explanation).
[e] Calibrated using the on-line calibration software Calib 6.1.0 Calibration Curve (Stuiver and Reimer, 1993; Stuiver et al., 2005).
[f] Model ages were determined from linear regression equations obtained by extrapolation of the regression lines representing the age models to the bottom of each core, assuming a constant sedimentation rate throughout the core.

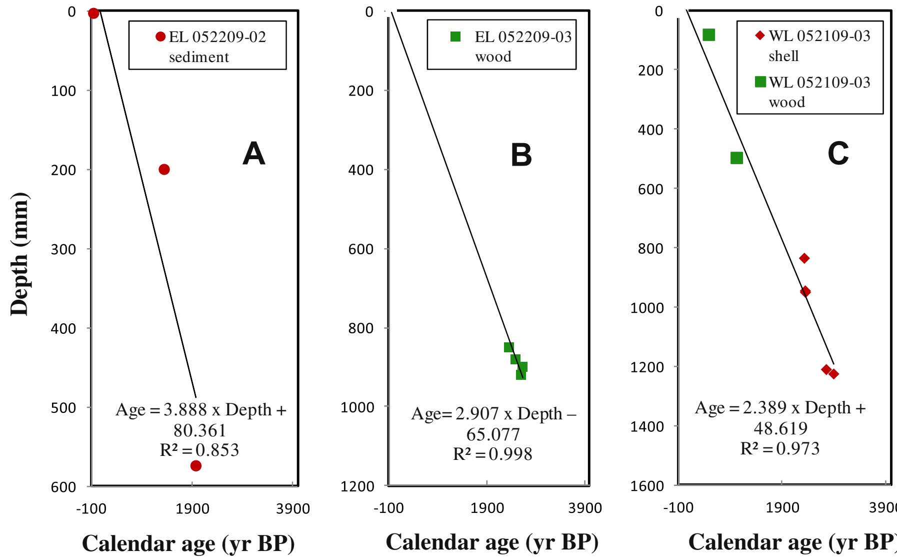

Fig 2. A. Radiocarbon-based age model for Eastern Lake core 052209-02. B. Radiocarbon-based age model for Eastern Lake core 052209-03. C. Radiocarbon-based age model for Western Lake core 052109-03.

Western Lake core 052109-03 was based on seven radiocarbon dates of wood fragments and shells (Fig. 2B and C). There are very strong linear correlations ($R^2 > 0.97$) between the radiocarbon dates of wood/shell fragments and depths observed in these two cores assuming the surface age is 0 year BP (Fig. 2B and C). The intercepts obtained from the radiocarbon age and depth regression lines of Eastern Lake cores 052209-02 and, 052209-03 and Western Lake core 052109-03 (Fig. 2) are 80, −65 and 49 years BP, respectively, indicating that the uncertainties of the age models are much larger than the analytical errors indicated in Table 1. The limited ^{14}C age data from Eastern Lake core 052209-02 suggest that there could be considerable uncertainty in the age model based on radiocarbon dates of bulk sediment OM (Fig. 2A). According to the age models, the sediments in Eastern Lake core 052209-02 and 052209-03 and Western Lake core 052109-03 were accumulated over the last 2413, 2930 and 3871 years, respectively, as determined by extrapolation of the regression lines representing the age models to the bottom of each core, assuming a constant sedimentation rate throughout the core (Fig. 2).

5.3. *Carbon and nitrogen isotopic characteristics of plants in and around the lake*

Thirty-eight plants were collected from the lake and its watershed (including forestland, sparsely vegetated dune areas and the lake itself) and analyzed for $\delta^{13}C$, $\delta^{15}N$, C% and N%. The study area is dominated by C_3 plants; however four C_4 species were also observed (Supplementary Table A.1). All of the C_4 plants are upland plants, except *Spartina sp.*, which is an emergent (or wetland) plant growing along the edge of the lake. The $\delta^{13}C$ and $\delta^{15}N$ values of upland plants range from −13.5 to −30.8‰ with a mean of −27.1 ± 5.1‰ ($n = 25$) and from 1.6 to −9.1‰ with a mean of −2.3 ± 2.5‰ ($n = 25$), respectively (Fig. 3). The $\delta^{13}C$ and $\delta^{15}N$ values of submerged plants are −23.2 to −25.6‰ (averaging −24.5 ± 1.2‰, $n = 3$) and 8.7 to 4.4‰ (averaging 6.7 ± 2.1‰, $n = 3$), respectively; whereas emergent plants have $\delta^{13}C$ values ranging from −14.3 to −28.2‰, with a mean of −26.0 ± 4.1‰ ($n = 10$), and $\delta^{15}N$ values of 8.9 to 2.5, with a mean of 4.8 ± 2.0‰ ($n = 10$) (Fig. 3). These data show that wetland plants (including emergent plants and submerged plants) generally have higher $\delta^{13}C$ and $\delta^{15}N$ values than the upland plants (Fig. 3). Also, submerged/floating plants are relatively more enriched in heavy carbon and nitrogen isotopes than emergent plants (Fig. 3). The C/N ratios of upland plants vary from 19 to 79.8, with a mean of 39 ± 17.1 (Fig. 4). The average C/N ratio of aquatic plants is 23.7 ± 4.7, lower than that of upland plants (Fig. 4).

5.4. *Carbon and nitrogen isotopic characteristics of organic matter in soil and dune*

Dune and soil samples collected from the lake area were analyzed for $\delta^{13}C$, $\delta^{15}N$, C% and N%. The $\delta^{13}C$ and $\delta^{15}N$ values of bulk OM in sand dunes range from −24 to −26.8‰, with a mean of −25.7 ± 1.8‰ ($n = 4$), and from −0.7 to −1.8‰, with a mean of −1.2 ± 0.5‰ ($n = 4$), respectively (Fig. 3). The C/N ratios of the dune samples varied from 7.1 to 3.7, averaging 5.8 ± 1.8 (Fig. 4). The $\delta^{13}C$ and $\delta^{15}N$ signatures of forest soil from the Western Lake area are −26.9‰ and −1.3‰, respectively (Fig. 3). The isotopic signatures of soil and dune samples from the lake surrounding areas suggest that C_3 plants are the predominant source of organic matter (OM) in soil and dune and that C_4 grasses, although present in local ecosystems, contributed negligible amounts to the OM pools in the study area.

5.5. *Carbon and nitrogen isotopic characteristics of POM and DOM in the lakes*

Particulate organic matter (POM) collected from Western Lake at different times exhibited more or less invariable carbon isotopic signatures with $\delta^{13}C$ values ranging from −27.7 to −27.8‰, but significant variations in the nitrogen isotopic ratios with $\delta^{15}N$ values varying from 0.8 to 5.1‰ during the study period of March–July of 2010 (Fig. 3). Similarly, dissolved organic matter (DOM) collected from the same lake at different times showed very small variations in $\delta^{13}C$ values (ranging from −27.1 to −27.6‰), but larger variations in $\delta^{15}N$ values (from −2.0 to 2.5‰) (Fig. 3). For Western Lake, the C/N ratios varied from 13.3 to 8.9 for POM and from 28.0 to 5.4 for DOM (Fig. 4). A sample of POM collected from Eastern Lake yielded more depleted $\delta^{13}C$ values (−33.0‰) than those from Western Lake, whereas its $\delta^{15}N$ value (3.6‰) is within the $\delta^{15}N$ range of the POM from Western Lake (Fig. 3). The $\delta^{13}C$ and $\delta^{15}N$

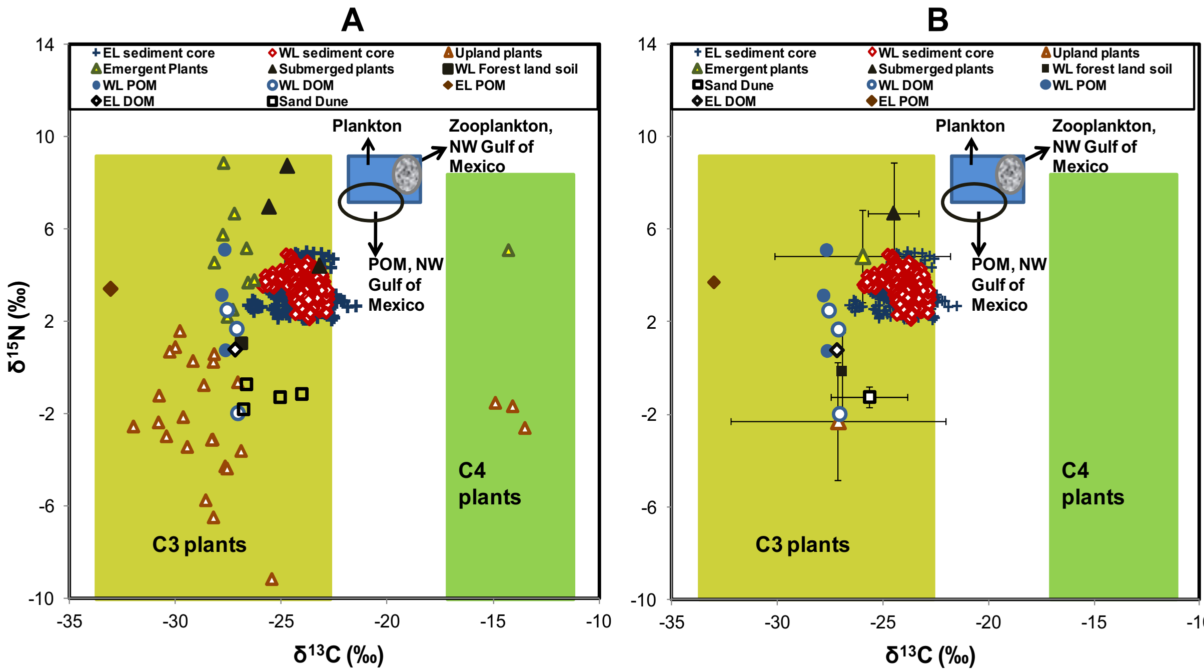

Fig 3. A. δ^{13}C and δ^{15}N values of organic matter in lake sediment, plants, POM and DOM from Eastern Lake and Western Lake in northwest Florida, in comparison with typical values of marine and land plants and other organic inputs in coastal areas. B. Comparison of δ^{13}C and δ^{15}N values of organic matter in lake sediment, POM and DOM with the mean δ^{13}C and δ^{15}N values of plants and soil in and around the lake area as well as the typical range of marine and terrestrial plants and organics (Macko et al., 1984; Lamb et al., 2006). Error bar indicates one standard deviation from the mean.

values of a DOM sample from Eastern Lake are −27.2‰ and 0.8‰, respectively (Fig. 3). The C/N ratios of POM and DOM from Eastern Lake (9.3 and 6.1, respectively) are comparable to those from Western Lake (Fig. 4). These data indicate that the POM and DOM in both lakes were derived primarily from aquatic and terrestrial C_3 plants and that C_4 plants, although present in the area, were not an important component of local biomass and thus were not a significant source of sedimentary OM in the lakes.

5.6. *Carbon and nitrogen isotopic characteristics of lake sediment*

The geochemical data from lake sediment cores display significant variations with depth (Figs. 5 and 6). The bulk organic C and N percentage contents (C% and N%) of Eastern Lake sediments varied from 0.4 to 7.4% and 0.03 to 0.71%, respectively, whereas the C% and N% of Western Lake varied from 0.4 to 10.4% and 0.04 to 0.81%, respectively (Fig. 5). The $(C/N)_{weight}$ ratios of the lake sediments are

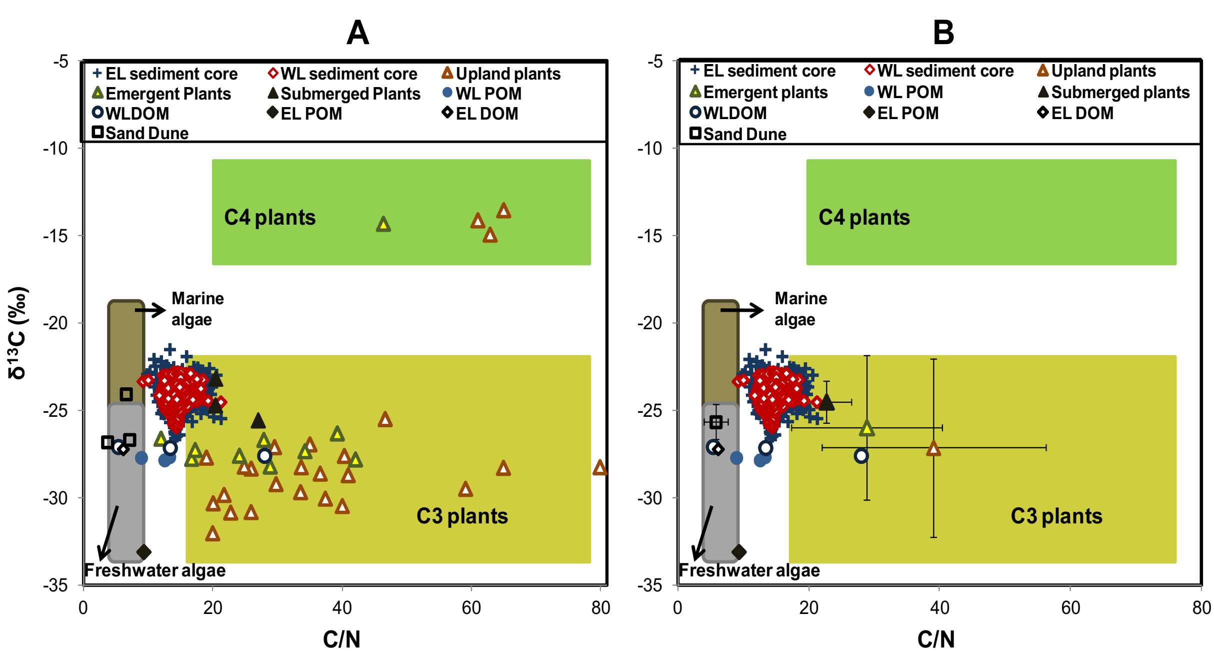

Fig 4. A. δ^{13}C and C/N values of organic matter in lake sediment, plants, POM and DOM from Eastern Lake and Western Lake in northwest Florida, in comparison with typical values of marine and land plants and other organic inputs in coastal area. B. Comparison of δ^{13}C and C/N values of organic matter in lake sediment, POM and DOM with the mean δ^{13}C and mean C/N values of plants and soil in and around the lake area as well as the typical values of marine and terrestrial plants and organics (Meyers, 1994; Lamb et al., 2006). Error bar indicates one standard deviation from the mean.

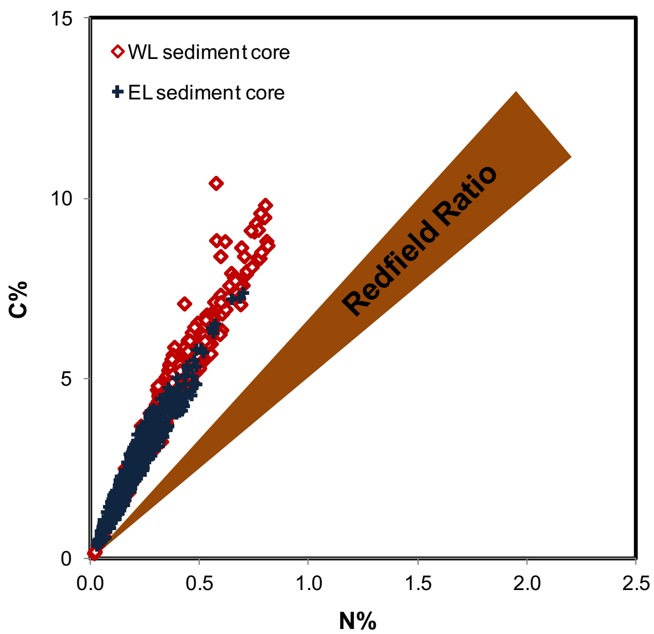

Fig 5. Relationship between C% and N% in lake sediment in comparison with marine field defined by Redfield ratio. $(C/N)_{weight}$ of Redfield ratio is 5.56 to 7.48 (Jørgensen, 2000). Note: $C/N_{atomic} = 1.17 \times (C/N)_{weight}$.

on average 12.9 for Eastern Lake and 13.2 Western Lake, and differ significantly from the marine field defined by the Redfield Ratio (Jørgensen, 2000; Fig. 5). The $\delta^{13}C$ and $\delta^{15}N$ values of Eastern Lake sediments varied from −21.5 to −26.6‰ and from 5.0 to 2.1‰, respectively (Fig. 6). Western Lake sediments have $\delta^{13}C$ values ranging from −22.8 to −25.8‰ and $\delta^{15}N$ values from 4.9 to 2.1‰ (Fig. 6). Temporal shifts in $\delta^{13}C$ and $\delta^{15}N$ values of the lake sediments reflect changes in the source of OM, likely induced by changes in climate and lake environment (e.g. Macko et al., 1984; Meyers, 1997; Lamb et al., 2006; Moy et al., 2011). The data including $\delta^{13}C$, $\delta^{15}N$ and C/N of the lake sediment have been detrended to remove any long-term trend by subtracting the next value from the current value (using first-order difference method) (Figs. 7–9).

6. Discussion

6.1. Proxy interpretation

Measurements of C%, N%, C/N, $\delta^{13}C$ and $\delta^{15}N$ values can provide valuable information about paleoenvironment and origins of sedimentary OM (e.g., Meyers, 1997; Lamb et al., 2006; Burnett et al., 2011). Variations of C% (and N%) in sediments probably result from changes in lake productivity, which is mainly controlled by temperature fluctuations and nutrient flux, although changing conditions of OM preservation could also have altered the C contents of sediments. All else being equal, high C% and N% values indicate warm conditions with high productivity in the lake and low influx of clastic material by flooding, overwash or wind erosion. Low C% and N% are indicative of colder conditions with low productivity in the lake and/or a relatively high influx of clastic material. Thus, an increase in C% may indicate increased productivity, increased preservation, and/or decreased dilution by clastic material.

Changes in C/N ratios likely reflect variations in the proportion of OM derived from terrestrial vs. aquatic environments. Terrestrial plants typically have C/N ratios greater than ~18, whereas the C/N ratios of aquatic plants (i.e., marine and freshwater plants) are lower, typically between 4 and 10 (Meyers, 1994; Lamb et al., 2006) (Fig. 4). The C/N ratios of the lake sediments from Eastern Lake and Western Lake are between 9 and 22, indicating that both algal

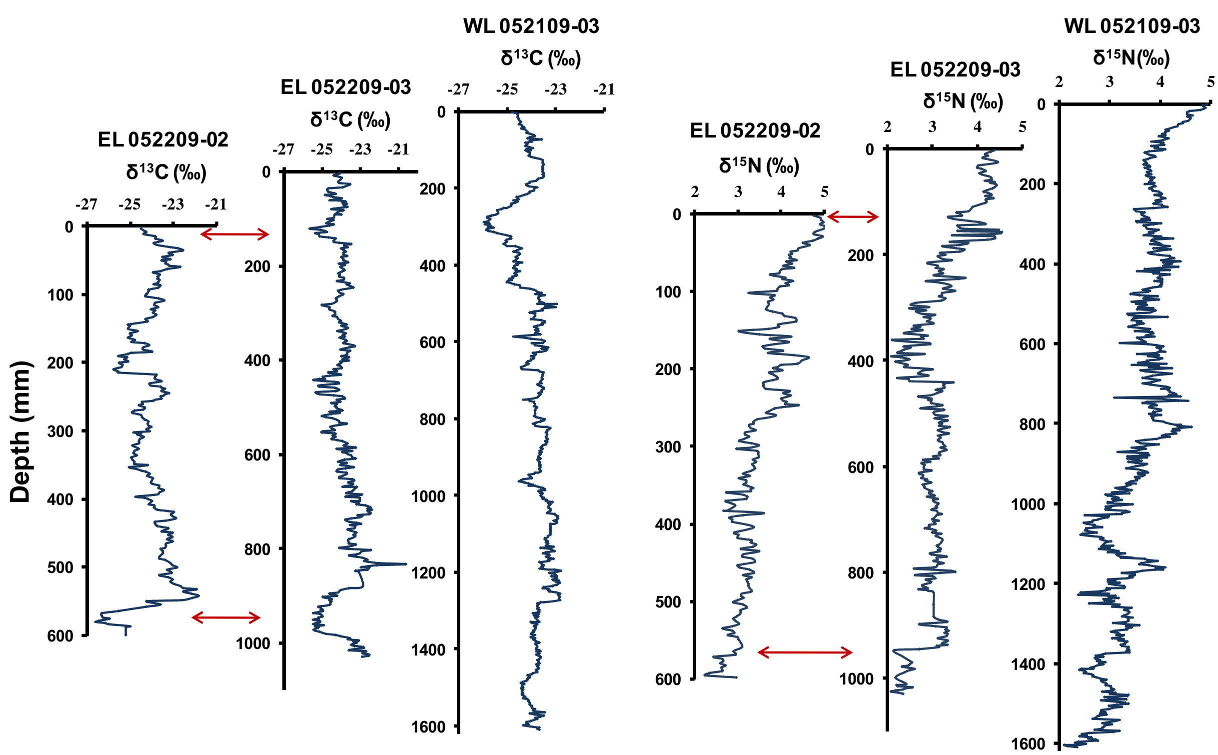

Fig 6. Comparison of $\delta^{13}C$ and $\delta^{15}N$ profiles of core 052209-02 and 052209-03 from Eastern Lake and core 052109-03 from Western Lake. Red arrows indicate likely identical shifts in different cores from Eastern Lake.

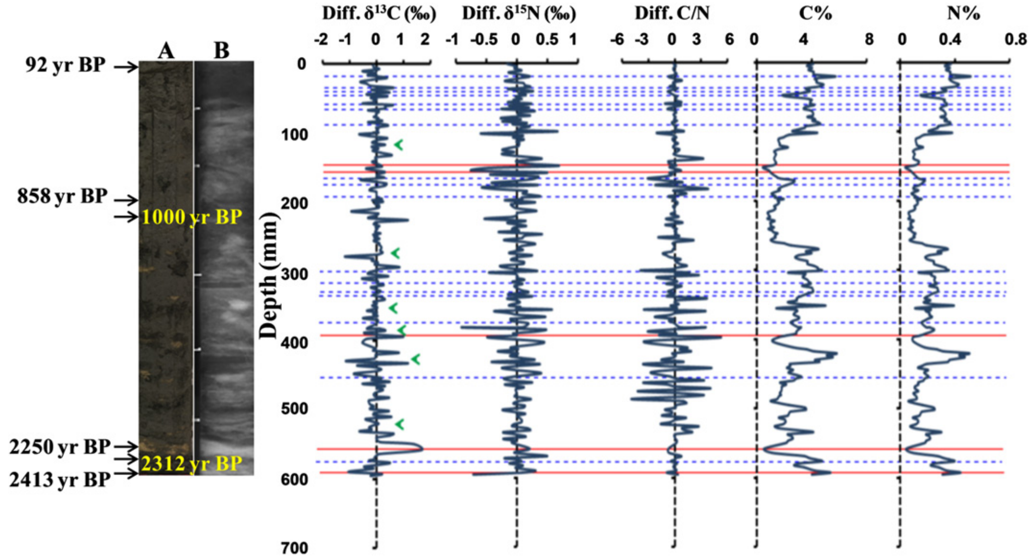

Fig 7. De-trended $\delta^{13}C$, $\delta^{15}N$ and C/N profiles, along with original C% and N% content profiles from core 052209-02 plotted against depth. The $\delta^{13}C$, $\delta^{15}N$ and C/N data are de-trended using the first-order difference method (see text for explanation). Horizontal lines mark concurrent positive $\delta^{13}C$ and $\delta^{15}N$ shifts and negative C/N shifts in the core, which are interpreted to be storm events. Red solid lines indicate positive excursion in $\delta^{13}C$ or $\delta^{15}N$ by $\geq 2\sigma$ ($1\sigma = 0.37$ for $\delta^{13}C$ and $1\sigma = 0.24$ for $\delta^{15}N$) and negative excursion in C/N. Blue dashed lines indicate positive excursion in $\delta^{13}C$ and $\delta^{15}N$ by $\geq 1\sigma$ and negative excursion in C/N. Green arrowheads indicate relatively higher productivity of C_4 emergent plants. A. Digital image of core; B. X-ray image of core. Also, shown calibrated model ages on bulk sediments. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

inputs and terrestrial OM are significant contributors to the sedimentary OM in these lakes. Thus, a negative shift in the C/N ratios of lake sediments (i.e., a shift to lower values) may indicate a change in the source of OM to more aquatic or marine input (Fig. 4).

The $\delta^{13}C$ and $\delta^{15}N$ values of sedimentary OM can also serve to differentiate OM of different origins (e.g., Deines, 1980; Macko et al., 1984; Meyers, 1994; Choi et al., 2001; Lamb et al., 2006; Lambert et al., 2008). Recently, Lambert et al. (2008) proposed a model based on OGPs preserved in coastal lake sediment for recognizing storms severe enough to cause coastal lakes being flooded by seawater. The OGPs (such as $\delta^{13}C$, $\delta^{15}N$ and C/N) are considered to be reliable and useful in recognizing storm events because they are sensitive to storm-induced changes in lake environment. When a storm hits a coastal lake area to cause seawater flooding in lakes, it has the potential to leave a signal preserved in the lake sediment. According to the Lambert et al. (2008) model, a coastal lake has two contrasting states: (1) an “isolated” state under which the lake system is isolated from the adjacent Gulf and the OGPs reflect the

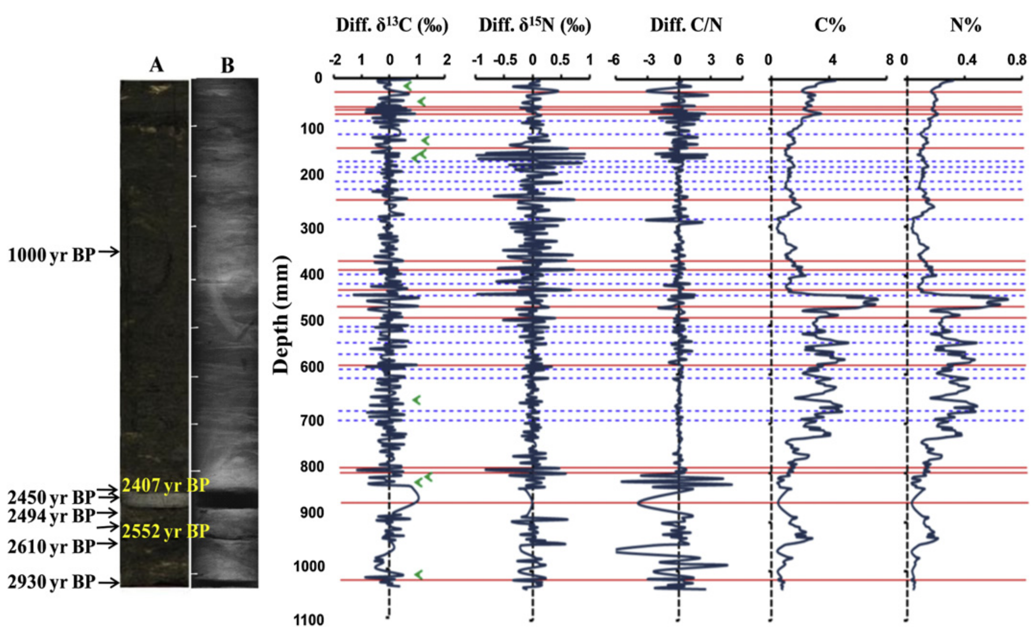

Fig 8. De-trended $\delta^{13}C$, $\delta^{15}N$ and C/N profiles, along with original C% and N% content profiles, from core 052209-03 plotted against depth. Horizontal lines mark concurrent positive $\delta^{13}C$ and $\delta^{15}N$ shifts and negative C/N shifts in the core, which are interpreted to be storm events. Red solid lines indicate positive excursion in $\delta^{13}C$ or $\delta^{15}N$ by $\geq 2\sigma$ ($1\sigma = 0.38$ for $\delta^{13}C$ and $1\sigma = 0.26$ for $\delta^{15}N$) and negative excursion in C/N. Blue dashed lines indicate positive excursion in $\delta^{13}C$ and $\delta^{15}N$ by $\geq 1\sigma$ and negative excursion in C/N. Green arrowheads indicate relatively higher productivity of C_4 emergent plants. A. Digital image of core; B. X-ray image of core. Also, shown calibrated model ages on wood fragments. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

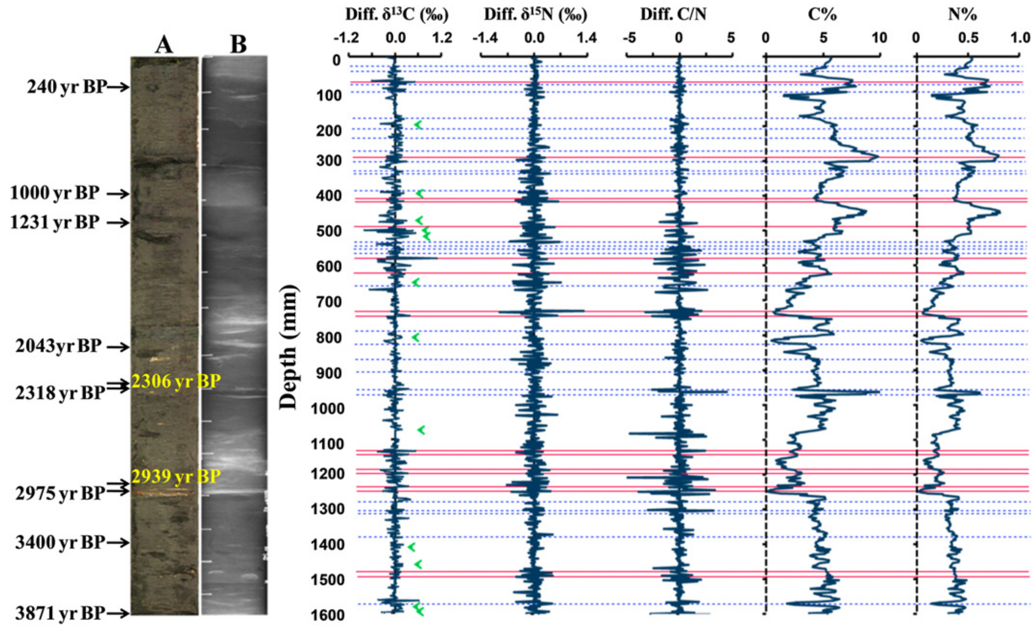

Fig 9. De-trended δ^{13}C, δ^{15}N and C/N profiles, along with original C% and N% content profiles, from core 052109-03 plotted against depth. Horizontal lines mark concurrent positive δ^{13}C and δ^{15}N shifts and negative C/N shifts in the core, which are interpreted to be storm events. Red solid lines indicate positive excursion in δ^{13}C or δ^{15}N by $\geq 2\sigma$ ($1\sigma = 0.16$ for δ^{13}C and $1\sigma = 0.21$ for δ^{15}N) or negative excursion in C/N. Blue dashed lines indicate positive excursion in δ^{13}C and δ^{15}N by $\geq 1\sigma$ and negative excursion in C/N. Green arrowheads indicate relatively higher productivity of C_4 emergent plants. A. Digital image of core; B. X-ray image of core. Also, shown calibrated model ages on wood fragments and shells. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

normal lake environment characterized by low nutrients and low δ^{13}C and δ^{15}N values; and (2) a "flooded" state under which the lake is subject to seawater flooding caused by hurricane-speed winds pushing the waves over the seaward barrier and characterized by a marine-like environment with higher δ^{13}C and δ^{15}N values. Thus, positive excursions in both δ^{13}C and δ^{15}N profiles in sediment cores would be indicative of large storm events (Lambert et al., 2008). This is because that OM of marine origin typically has higher δ^{13}C and δ^{15}N values than that derived from terrestrial C_3 vegetation (Figs. 3 and 4).

In addition, seawater in the Gulf of Mexico generally has much higher nutrients and DIC than coastal lake water (Lambert et al., 2008). For example, the average nitrate concentrations in Eastern Lake and Western Lake are 0.29 mg/l and 0.27 mg/l, respectively (ChoctaWhatchee Basin Alliance, 2009). In comparison, nitrate concentration in the Gulf of Mexico is ~1.6 mg/l and can reach up to 6 mg/l; and the average DIC concentration in the Gulf of Mexico is 147 mg/l and is also higher than in coastal lakes (Lambert et al., 2008). During the "flooded" state, high influx of fresh marine nutrients in the form of nitrate and DIC to coastal lakes due to seawater inundation, would lead to high aquatic productivity (i.e., a rapid eutrophication spike) in the lake immediately after the storm (Lambert et al., 2008). Not only are the marine-derived nutrients more enriched in ^{13}C and ^{15}N than freshwater nutrients, the high lake productivity immediately after the storm would also result in ^{15}N and ^{13}C enrichments in the remaining nitrate and DIC pool. As a consequence, OM formed and deposited on the lake floor under "flooded" state would be more enriched in ^{13}C and ^{15}N than OM formed under the "isolated" state (or normal condition) (Lambert et al., 2008).

Our analysis of potential OM sources in the study area supports the concept that concurrent positive excursions in δ^{13}C and δ^{15}N profiles, in conjunction with a negative shift or insignificant change in the C/N ratio, in sediment cores most likely indicate severe storm events (Table 2, Figs. 3 and 4) as proposed by Lambert et al. (2008). In addition, we recognize a few other OGPs patterns that can be interpreted as indicating changes in certain aspects of the lake environment (Table 2). As shown in Figs. 3 and 4, using any single geochemical parameter alone can lead to ambiguous interpretations, but examining the patterns of OGPs can help resolve the ambiguity (Table 2). For example, a negative shift in δ^{15}N could have resulted from input from dune erosion, or low lake productivity, or an increase in terrestrial OM input (Table 2). However, dune samples from the lake area have relatively lower C/N and δ^{15}N values relative to wetland and other terrestrial plants (Figs. 3 and 4). Thus, a negative shift in δ^{15}N, concurrent with negative shifts in C/N, C% and N% in coastal lake sediments, can be interpreted as indicating input from dune erosion (Table 2). Similarly, a positive shift in δ^{15}N profile of a coastal lake sediment core could indicate an overwash event, or increased algal productivity due to higher temperature, or increased algal productivity due to higher influx of nutrients resulting from seawater flooding, or a decreased production of emergent vegetation relative to submerged/floating plants, or increased productivity of the emergent C_4 wetland plant *Spartina* sp. at the lake margin resulting from wetland expansion due to drier conditions (Table 2). The ambiguity in the interpretation of δ^{15}N data, however, can be resolved by examining the patterns of the OGPs (Table 2). That is, positive shift in δ^{15}N, if concurrent with a positive shift in δ^{13}C and a negative shift or no change in C/N profile, can be interpreted as indicating seawater flooding or an overwash event (Table 2).

6.2. Temporal variations in OGPs and identification of large storm events

Temporal variations in OGPs reflect changes in the source of OM and lake environment (Figs. 7–9). In the cores analyzed, significant positive and negative shifts occurred in the δ^{13}C and δ^{15}N profiles at various depths, which correspond to negative or positive shifts in the C/N, C% and N% records. In Eastern Lake core 052209-02, significant positive shifts in both δ^{13}C ($>2\sigma$) and δ^{15}N (1σ) at 558 mm depth are correlated to a 1-cm thick visible sand layer (presumably

Table 2
Interpretations of geochemical proxies.

$\delta^{13}C$ shift	$\delta^{15}N$ shift	C% shift	N% shift	C/N shift	Interpretation
Positive	Positive	Negative	Negative	Negative	Overwash event (overwash sand deposit)
Positive	Positive	Positive	Positive	Negative or no change	Higher lake productivity due to influx of nutrients from seawater flooding
Positive or Negative	Negative	Negative	Negative	Negative or positive	Input from dune or soil erosion by wind or rain, or decreased lake productivity
Negative	Positive	Negative	Negative	Negative	Decreased productivity of emergent plants relative to production of submerged plants or algae
Negative	Positive	Positive	Positive	Negative or no change	Higher lake productivity due to high temperature
Positive or Negative	Negative	Positive	Positive	Positive	High terrestrial input
Positive	Positive	Positive	Positive	Positive	High productivity of emergent C_4 plants such as *Spartina* sp. due to wetland expansion

an overwash deposit) as well as negative shifts in C%, N% and C/N (1σ) (Fig. 7), which we interpret as a severe storm event (Table 2) and thus supports the model proposed by Lambert et al. (2008). Similarly, in Eastern Lake core 052209-03, a 3-cm thick visible sand layer (presumably an overwash deposit) occurring at 850–880 mm depth exhibits positive excursions from the base values in both $\delta^{13}C$ ($>2\sigma$) and $\delta^{15}N$ (1σ) and concurrent negative shifts in C%, N% and C/N ($>2\sigma$) profiles (Fig. 8), also consistent with the OGP pattern expected from seawater inundation caused by a large storm event (Table 2). Thus, our OGP data from both modern C pools and ancient overwash sand deposits preserved in sediment cores all support the Lambert et al. (2008) model for identifying storms based on the OGPs. The same OGP pattern (i.e. concurrent positive shifts in $\delta^{13}C$ and $\delta^{15}N$ and a negative shift in C/N) will likely help identifying other storm events throughout the cores where overwash sand layer are not present (Figs. 7–9; Table 2; Supplementary Table A.2).

At a few places in the cores, all organic geochemical proxies (i.e., $\delta^{13}C$, $\delta^{15}N$, C%, N%, and C/N) have shown excursions in the positive directions, indicating increased productivity of C_4 emergent plant such as *Spartina sp*. around the lake, likely due to wetland expansion/lake contraction (Figs. 7–9; Table 2).

6.3. *Reconstruction of paleostorm frequency*

The model age of the thick sand layer is 2250 yr BP in core 052209-02 and 2450 yr BP in core 052209-03, respectively. Both overwash sand layers are assumed to be deposited by the same storm event because these two cores were collected within 1 m of each other, and the ^{14}C age difference between the two visible sand layers most likely reflect large uncertainties in the age models, especially the age model for core 052209-02, which is based on very limited ^{14}C dates of bulk sediment OM (Table 1, Fig. 2). In both cores, the visible sand layer was characterized by positive excursions in both $\delta^{13}C$ ($>2\sigma$) and $\delta^{15}N$ (1σ) that are correlated with a negative excursion in C/N (1σ) (Figs. 7 and 8). The positive shifts could be due to input of marine OM and/or OM formed from seawater-derived nutrients during and after the storm event. Both positive excursions in $\delta^{13}C$ and $\delta^{15}N$ also coincided with negative shifts in C% and N%, which are characteristic of organic-poor sand layers. There were a total of 22 identical patterns at different depths in the short core (052209-02) and 35 identical patterns in the long core 052209-03 from Eastern Lake (Figs. 7 and 8). Western Lake core 052109-03 revealed 45 such patterns at different depths (Fig. 9). If this OGPs-based pattern recognition of storm events and our age models are valid throughout the time period represented by the cores, our data suggest that a minimum of 45 large storms hit the northwest coast of Florida during the past 3871 years. The recurrence interval of large storms derived from Eastern Lake cores 052209-03 and 052209-02 is 84 years in the past 2900 years and 110 years over the last 2400 years. The recurrence interval calculated from the Western Lake core (052109-03) data is 86 years over the last 3900 years.

6.4. *Comparison with other records*

Liu and Fearn (2000) reported (what was considered at the time to be) a high-resolution record of catastrophic hurricane landfalls for the past 7000 years based on analysis of overwash sand layers in sediment cores collected from Western Lake. According to Liu and Fearn (2000), twelve hurricanes (category 4 or 5) hit the Western Lake area during the past 3400 years with a long-term frequency of one hurricane in every 280 years. The frequency of catastrophic hurricane was low between 3400 and 5000 yr BP and also during the last 1000 yr BP, but increased between 1000 and 3400 yr BP to five hurricanes per 1000 years. The time interval of 1000–3400 yr BP was considered a “hyperactive” period by Liu and Fearn (2000). Especially, during the first millennium of 1000–2000 yr BP, catastrophic hurricanes struck five times per 1000 years, corresponding to a landfall probability of 0.5% per year (or a recurrence interval of 200 years), higher than the recent landfall probability of 0.1% per year (or a recurrence interval of 1000 years) (Liu and Fearn, 2000). In comparison, our Western Lake data show a total of 45 storms hit the area in the past 3900 years with an average recurrence interval of 86 years, amongst which 14 storms hit during the last 1000 years with a recurrence interval of 71 years, and 28 storms during the time period of 1000–3400 yr BP with a recurrence interval of 86 years (Figs. 2C and 9; Table 3). Eastern Lake core 052209-02 data revealed 12 storms in the past 1000 years with an average recurrence interval of 83 years, and 10 storms during the proceeding time period of 1000–2400 yr BP with a 141-

Table 3
Comparison of recurrence intervals derived from OGPs in sediment cores and also with those from Liu and Fearn (2000).

EL core	052209-02	EL core	052209-03	WL core	052109-03	Liu and Fearn	2000
Time period (yr BP)	Recurrence interval (yr)	Time period (yr BP)	Recurrence interval (yr)	Time period (yr BP)	Recurrence interval (yr)	Time period (yr BP)	Recurrence interval (yr)
0–1000	83	0–1000	71	0–1000	71	0–1000	1000
1000–2400	141	1000–2900	92	1000–3400	86	1000–3400	200
0–2400	110	0–2900	84	0–3900	86	0–3400	280

year recurrence interval (Figs. 2A and 7; Table 3). Core 052209-03 from Eastern Lake yielded 14 storms out of total 35 storms during the last 1000 years with a recurrence interval of 71 years, whereas 21 storms are found during 1000–2900 yr BP with a recurrence interval of 92 years (Figs. 2B and 8; Table 3). The data from these cores suggest similar or slightly higher storm activities (1.2–1.4% per year of landfall probability) during 0–1000 yr BP compared to the time period of ~1000–3400 yr BP (with a landfall probability of 1.2% per year), inconsistent with the "hyperactive period" suggested by Liu and Fearn (2000). This inconsistency of storm frequencies is likely due to large uncertainties in the age models, which are based on ^{14}C dating of bulk sediment OM and/or limited wood and shell samples (Fig. 2). Bulk sediment OM is a mixture of OM derived from aquatic and terrestrial sources and may not provide accurate age controls due to the possible influx of old carbon from erosion of soil and other old OM deposits. However, OGPs-based storm record suggests many more storm events than the proxy record based on counting overwash sand layers (Liu and Fearn, 2000).

7. Conclusions

The Florida panhandle is susceptible to hurricane landfalls. Strong storms affecting the area often leave their marks in coastal lake sediments by modifying the geochemical signatures of OM deposited in coastal lakes. The sources of OM in the sediment of dune lakes include a lacustrine source (consisting of phytoplankton, zooplankton, bacteria, and macrophytes), a terrestrial source derived from land surrounding the lakes, and a marine source derived from the seaward Gulf of Mexico sediments and nutrients associated with seawater flooding during large storm events. The isotopic signatures of POM and DOM from the lakes under normal "isolated" conditions indicate that C_3 plants (both aquatic and terrestrial) are the dominant source of sedimentary OM. Although some C_4 grasses are present in the lake area, they are not a significant source of OM in soil and lake systems in the study area that is currently dominated by C_3 plants (i.e., trees). The influx of marine OM and nutrients due to seawater flooding leads to a high lake productivity and consequently an enrichment in ^{13}C and ^{15}N contents of sedimentary OM formed during and after the seawater flooding. The positive C isotopic shifts in coastal lake sediment cores, if concurrent with positive shifts in $\delta^{15}N$ and negative shifts in C/N ratios, most likely indicate shifts to a marine-like environment in coastal lakes.

The storm recurrence intervals derived from OGP records from Eastern Lake are 84 years in past 2900 years and 110 years over the last 2400 years. The recurrence interval of severe storms calculated from the OGP record from Western Lake is 86 years in the past 3900 years. The radiocarbon ages derived from bulk sedimentary OM may not be reliable because of possible interference of older carbon from different sources entering into the lakes, leading to large uncertainties in reconstruction of the timing and frequency of identified storm events.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2012.11.046.

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