The sedimentary response to a rapid change in lake level in Lake Tanganyika

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Highlights:

Elevated carbonate concentrations coincide with maximum lake level

Negative excursions in both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ coincide with maximum lake level

Results imply changes in lake chemistry during lake level oscillation centered at 1879

Abstract

We present records of sedimentary organic carbon, nitrogen, and carbonate, and stable isotope records of organic material and carbonate from a series of sediment cores that straddle the permanent chemocline in Lake Tanganyika. Sedimentation rates for these cores are consistent among the sites ($\sim 0.05 - 0.1 \text{ cm y}^{-1}$), and all records show an increase in sedimentary carbonate (aragonite) content centered at ~ 1879 . The mid-19th century coincides with a major ($\sim 10 \text{ m}$) lake level transgression. Throughout the period of lake level transgression and subsequent regression, the organic matter $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ records develop a prominent and coincident negative excursion followed by a return to values similar to those prior to the lake level transgression. This negative excursion in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ is also coincident with an increase in carbonate-corrected organic carbon. We interpret the $\delta^{13}\text{C}$ results as a decline in primary production during the transgression with the $\delta^{15}\text{N}$ results signaling a concomitant increase in the reliance on nitrogen fixation as the nitrogen source. The coincident peak in organic carbon is interpreted as being a result of enhanced preservation driven by the precipitation and burial of aragonite.

1.0 Introduction

Lake Tanganyika is a large, deep ($\sim 1470 \text{ m}$ maximum depth) tropical lake nestled within the East African Rift system between $\sim 3^\circ$ and 9° S (e.g., Coulter, 1991; **Figure 1**). The half-graben basins that cradle the lake create a bathymetry whereby the lake has relatively steep sides with a number of shelf platforms (e.g., Rosendahl et al., 1986). These platforms make exquisite localities for sediment coring for geochemical and paleo-climatological studies.

Like any aquatic system, lake chemistry is controlled by a combination of internal

54 physical, chemical, and biological processes as well as new inputs via the atmosphere and
55 rivers. Although many rivers contribute to the lake's chemical composition, Lake
56 Tanganyika's major element chemistry is likely dominated by inputs from the Ruzizi
57 River, which drains Lake Kivu (e.g., Craig, 1974; Cohen et al., 1997). Lake Kivu is
58 alkaline and is responsible for Lake Tanganyika's high Mg to Ca ratio (Craig, 1974;
59 Cohen et al., 1997). Lake Tanganyika also has a relatively high pH with the shallower
60 reaches having a pH of ~ 9 and an alkalinity of ~ 6.5 mM (**Figure 2**, Degens et al., 1971;
61 Edmond et al., 1993; Plisnier et al., 1999). These waters are supersaturated with respect
62 to calcite, aragonite, and magnesian calcite (Cohen et al., 1997), and this supersaturation
63 fosters a diverse assemblage of carbonate deposits (Cohen and Thouin, 1987; Haberyan
64 and Hecky, 1987; Cohen et al., 1997). The lake's carbonate system does seem to vary
65 significantly over time, however, and prior to ~ 2 - 4 ky, chemical conditions were such
66 that there seems to have been very little or no carbonate burial in contrast to the present
67 conditions (Degens et al., 1971; Haberyan and Hecky, 1987; Felton et al., 2007).

68 Because of its tropical setting, Lake Tanganyika has a permanent thermocline (e.g.,
69 Degens et al., 1971; Coulter and Spigel, 1991; **Figure 2**). This permanent stratification
70 provides a density barrier, which results in inefficient mixing between the lake's deep and
71 surface waters, and coupled with its internal biogeochemistry this physical stratification
72 generates persistent chemical stratification with dissolved oxygen generally restricted to
73 the upper ~100 meters in the north basin and sulfide present below ~ 150 meters
74 (Edmond et al., 1993, Plisnier et al., 1999; **Figure 2**). The macronutrients nitrate, silicic
75 acid, and phosphate have low concentrations in the shallowest water column depths (e.g.,
76 Edmond et al., 1993; Plisnier et al., 1999). As a consequence of its great depth,

77 stratification, and the presence of a chemocline, the deep waters are also rich in the
78 nutrients ammonium, phosphate, and silicon, and the upwelling of this water is the
79 primary nutrient supply mechanism for the lake ecosystem with nitrogen fixation also
80 being an important, if not the primary, source of fixed nitrogen (e.g., Hecky et al., 1991;
81 Verburg and Hecky, 2009). There is considerable temporal variability in the chemistry
82 of Lake Tanganyika's waters (Plisnier et al., 1999). This variability is driven by a variety
83 of biogeochemical and hydrodynamic processes, which varies on seasonal to interannual
84 timescales (Coulter and Spigel, 1991; Hecky et al., 1991; Plisnier et al., 1999).

85 There has been considerable interest in Lake Tanganyika's response to climate
86 change. Lake level has varied between roughly 700 and 785 m above sea level over the
87 past ~3000 years (Cohen et al., 1987; Alin and Cohen, 2003). These changes in lake
88 level are generally viewed as being forced by changes in regional climate, the elevation
89 of the lake's outlet, or a combination of these two factors (e.g., Alin and Cohen, 2003).
90 Germane to the current work, a lake low stand dominated the latter part of the Little Ice
91 Age (LIA, 1550 - 1850) followed by a relatively rapid lake level rise that reached its
92 maximum around 1879 (Nicholson, 1999; Alin and Cohen, 2003). Although increased
93 precipitation certainly played a role in lake level rise, the extent of the transgression,
94 followed by the abrupt regression, was regulated in part by the lake's present-day primary
95 outlet to the Lukuga River being blocked by vegetation and sediment (Nicholson, 1999).
96 Once this debris dam was breached, the lake regressed rapidly to a fairly stable level over
97 the next one to two decades (see a detailed review in Nicholson, 1999), and this lake level
98 stability has been relatively persistent up to the present (Alin and Cohen, 2003).

99 It has also been pointed out that the lake has been warming and that this warming is

likely increasing thermal stratification (O'Reilly et al., 2003; Verburg et al., 2003). Increasing stratification in turn diminishes the exchange between the nutrient-rich deep waters and surface waters (Verburg et al., 2003; Verburg and Hecky, 2009). Because biological production relies on this nutrient exchange (e.g., Hecky et al., 1991) it can be reasoned that lake productivity can be expected to decline as thermal stratification increases (O'Reilly et al., 2003) and biogenic silica (Tierney et al., 2010) data from lake sediments have been used to support this inference.

In this paper we examine the sedimentary records of Lake Tanganyika to investigate changes in lake chemistry over the past ~ 200 years. Our work focuses specifically on five sites (ten cores) from a coring transect on the Luiche Platform that spans the current chemocline (**Figure 1**). We present carbonate, organic carbon, and nitrogen records that exhibit maximum concentrations that roughly coincide with the lake's most recent high stand followed by a decline to modern values. These results imply a close coupling between lake level and lake biogeochemistry and further imply that this high stand resulted in a significant shift in lake productivity and nitrogen supply.

2.0 Materials and methods.

Many of the sampling and analytical details have been presented previously (Brucker et al., 2011) but are briefly reiterated here. Sediment cores were collected using a four-position Hedrick/Marrs multi-corer (similar to the eight-position corer described in Barnett et al., 1984). At each site we sectioned two cores with one being sectioned for later analysis of ^{210}Pb and ^{137}Cs (cores indicated with an "A" in the figures). These radioisotopes were determined by γ -ray spectroscopy (e.g., Gilmore and Hemingway, 1995). Sediment samples were freeze-dried and ground before further analyses. The

other cores were sectioned under a nitrogen atmosphere and the sediment was centrifuged for pore water separation, and samples from these cores were presented previously (Brucker et al., 2011; cores indicated with a “B” in the figures).

We determined inorganic and organic carbon on the two cores in two separate laboratories, the University of California Riverside (UCR; B cores) and Oregon State University (OSU; A cores). The UCR analyses (B cores) were previously presented in Brucker et al. (2011), but are presented here for completeness. At UCR, total carbon was measured using an Eltra CS-500 elemental analyzer. Samples were acidified online for total inorganic carbon and total organic carbon was calculated by difference. Samples for total nitrogen were measured on these samples at the University of Akron on a Perkin Elmer Series II 2400 CHNS/O analyzer. The B cores were also used for pore water sampling and the sediment samples were taken from centrifuge tubes for solid phase analyses.

At OSU, carbonate, organic carbon, and nitrogen measurements were made on the same core as that used for ^{210}Pb and ^{137}Cs analyses (B cores). These analyses were not presented in Brucker et al. (2011). These cores were sectioned in the field and stored for later analyses. Total carbon and nitrogen were analyzed on a Carlo Erba Instruments NA 1500 and organic carbon was measured on samples that had been acidified for removal of carbonate. Inorganic carbon is calculated as the difference between these two values.

Ten samples were also analyzed on a Rigaku Ultima IV X-ray Diffractometer (XRD) located at the University of Akron. The analyzed samples ranged between 5% and 68% CaCO_3 and included samples from the 72, 107, 120, and 332 m water depths (see Appendix Table A1 for specific samples). Analysis of mineral components in finely

powered samples was achieved using the PDXL 2 version 2.1.3.4 integrated X-ray powder diffraction software. Selected samples from cores at 107 meters and 120 meters water depth were also imaged on an FEI Quanta 200 Environmental Scanning Electron Microscope (ESEM) at low vacuum at the University of Akron. Samples were freeze-dried and then loaded onto ESEM stubs using an adhesive tape. Pictures were taken of representative grains with a specific focus on the carbonate that was present in the samples.

Isotope analyses on the organic matter and the carbonate are presented for three of the B cores (72 m, 120 m, and 335 m). Carbon and nitrogen isotope analysis of organic matter ($\delta^{13}\text{C}_{\text{org}}$ vs VPDB, $\delta^{15}\text{N}$ vs air) were run at Oregon State University by continuous flow isotope ratio mass spectrometry on a Carlo Erba NA-1500 elemental analyzer connected to a DeltaPlus XL mass spectrometer. Prior to $\delta^{13}\text{C}$ analysis on the organic matter $\delta^{13}\text{C}_{\text{org}}$, samples were acidified using 10% HCl to remove carbonates while $\delta^{15}\text{N}$ was measured on unacidified samples and represents the isotope value of the total N. Organic carbon isotope data were calibrated with the international standards USGS 40 and ANU sucrose, and nitrogen isotope data were calibrated using USGS 40 and IAEA-N2. The Elemental Microanalysis low carbon isotopic standard (B2153) was used as a quality assurance standard. This standard has a reported $\delta^{13}\text{C}_{\text{org}}$ value of -27.5‰ and $\delta^{15}\text{N}$ value of +6.7‰ and we obtained values of $-27.61 \pm 0.05\text{‰}$ and $+6.56 \pm 0.07\text{‰}$ respectively. The isotopic composition of carbonate ($\delta^{13}\text{C}_{\text{carb}}$ and $\delta^{18}\text{O}$ both with respect to VPDB) was analyzed by dual inlet mass spectrometer using a Kiel III carbonate preparation system connected to a MAT252 mass spectrometer. C_{org} was not removed prior to the isotopic analysis of carbonate, and we assume that the analyzed CO_2 derives

solely from the carbonate phases. Carbon and oxygen isotopes of the carbonate fraction were calibrated using the internal laboratory calcite standard (Wiley standard, $\delta^{13}\text{C} = -0.41\text{‰}$ and $\delta^{18}\text{O} = -7.20\text{‰}$ VPDB). In addition, the international standard NBS19 ($\delta^{13}\text{C} = +1.95\text{‰}$ and $\delta^{18}\text{O} = -2.20\text{‰}$ VPDB) was used as a quality assurance standard and we obtained values of $1.96 \pm 0.01\text{‰}$ for carbon and -2.18‰ for oxygen.

3.0 Results

3.1 Age Models

The ^{210}Pb - and ^{137}Cs -derived sedimentation rates for the cores discussed in this communication were presented previously (Brucker et al., 2011); however, here we add additional information on sedimentation rates and re-evaluate these previously published rates in more detail as they are central to our discussion. The rates reported here are similar to the values determined in that previous publication with modifications based on the reanalysis presented here. For the three shallowest sites, we estimated the depth of maximum ^{137}Cs and assigned an age of 1961, which is the average of two Cs maxima reported previously from the Southern Alps (Putyrskaya and Klemm, 2007; **Figure 3**). Neither of the deeper cores (232 m and 332 m sites) have sufficient data to estimate the ^{137}Cs maximum. For the ^{210}Pb fits we generally fit a single regression to calculate sedimentation rates for the upper portion of the sediment column and above the deepest few (low activity) samples. For the 120 m site we fit the data below the upper few centimeters because the surface layer may have been mixed during a core recovery or may have undergone some other disturbance (e.g. Nittrouer et al., 1979; **Figure 3, Table 1**). Alternatively the most recent sediments might be influenced by recent changes in sedimentation due to anthropogenic impacts on the watershed (e.g., McKee et al., 2005;

Alin and Cohen, 2002; O'Reilly et al., 2005) or may have undergone bioturbation. We recognize that for both the 72 m and the 107 m sites there is not an apparent change in the slope of the ^{210}Pb data within the upper-most sediment package (**Figure 3**), which makes these latter explanations for the upper few data points at 120 m unlikely. Despite that observation, the broad peak in Cs could imply some mixing at these sites, but we simply do not have the data to assess this further. For the 232 and 332 m sites, these are below the current chemocline depth and are bathed in sulfidic waters, thus the sediment distributions are unlikely to have been influenced by any bioturbation. For the 332 m site, we calculate two sedimentation rates determined above and below a disturbance at ~3 cm depth, and then average the two sedimentation rates (**Figure 3, Table 1**). For the 232 m site, both the ^{210}Pb and ^{137}Cs in the upper 10 cm of this core are lower than in neighboring cores. This suggests that the uppermost portion of the core (discussed further below) may have been lost upon sampling. Because of this issue, sedimentation rates at this locality harbors considerable added uncertainty relative to the shallower sites.

In addition to the radioisotope-based estimates of sedimentation rates, we also used the fact that all of the cores have a large dynamic range in their CaCO_3 contents as a tool for refining our choice of sedimentation rates (**Figure 4**). For this exercise, we visually overlapped the CaCO_3 profiles as much as possible, focusing on the various features throughout the core, including the maximum peak, the minimum values prior to the peak, and the change from the high values to the lower modern values (**Figure 4**). Because the sites at 107 and 120 meters have the most robust estimate of sedimentation rates, we effectively anchored the other cores to these cores for estimating the sedimentation rates. We recognize that this approach makes the assumption that changes in lake chemistry are

215 synchronous over the distances covered by our cores and that the sedimentation rates
216 remained constant over the interval covered by the sediment core. One potential issue
217 with this particular approach is that higher rates of epilimnetic warming, relative to the
218 hypolimnion (O'Reilly et al., 2003), would tend to lead to an acceleration of carbonate
219 accumulation in the shallower part of the lake relative to the deeper portion. We also
220 note some surprise at the apparently constant sedimentation rates despite the large drop in
221 carbonate contents from the mid-1800's to the present. For example, in Lake Kivu, a
222 change in carbonate sedimentation during the mid-1900's has been suggested as the cause
223 of a doubling of the sedimentation rate in that system (Pasche et al., 2005). In the absence
224 of changes in sedimentation rate, despite the fluctuations in carbonate contents, there
225 must have been a change in sediment composition at our sites, most likely because of
226 changes in biogenic opal or lithogenic material. Despite this compositional change, when
227 we align the carbonate records, the organic carbon and isotope records are synchronous
228 between the 72 m and 120 m sites, which bolsters our confidence in the relative age
229 model assumptions. We assumed that the radioisotope-based sedimentation rate of 0.09
230 cm y^{-1} for the two cores at ~100 meters is accurate, and we used these two cores as our
231 primary tie points for the rest of the cores. The carbonate peaks for these cores align with
232 the most recent high lake stand (~ 1879, **Figure 4, lower panel**). Based on alignment of
233 the chemical data from the cores, the estimated sedimentation rate for the 72 m site is
234 $\sim 0.07 \text{ cm y}^{-1}$, which is slightly less than the two sites at ~100 meters as well as the ^{210}Pb
235 value of 0.12 cm y^{-1} , but it is consistent with the ^{137}Cs -based value (**Figure 3**). Thus for
236 the three shallowest sites, the CaCO_3 -based sedimentation rate ranges from 0.07 - 0.09
237 cm y^{-1} . For the two cores from 232 meters water depth, aligning these cores based on

their CaCO₃ profiles reveals that one of the cores has a significant offset relative to the other. It appears that the upper 8 - 9 cm of that core (232A) has been lost (**Figure 4**). This suspicion is supported by the fact that the ²¹⁰Pb and ¹³⁷Cs inventories are lower compared to the other cores (**Figure 3**). By aligning the CaCO₃ profiles from this core with the shallower cores, we obtain a sedimentation rate of 0.08 cm y⁻¹, which is both regionally consistent and consistent with the limited ²¹⁰Pb data for this site (**Figure 3**). For the 332 m site (**Figure 4**), there also seems to be some offset between the two cores. However, this offset is ~1 cm, which is considerably less of an offset than the larger one seen for the one core at 232 m. The sedimentation rate at this deepest site, based on the carbonate records, seems to lie between 0.05 and 0.06 cm y⁻¹, with a value of 0.05 cm y⁻¹ selected here. This value matches that derived from the ²¹⁰Pb profile if we pool all of the upper six data points and draw a single line through the data (**Figure 3**). For all of the sites, the sedimentation rates thus cluster quite closely between 0.05 - 0.09 cm y⁻¹, and appear to be consistent among the three approaches used here.

3.1.1 Limitations of the Age Model.

Our age models do not suggest large variations in sedimentation rates through time over the time periods considered. The possibility certainly exists that there have been recent increases in sedimentation, and the early parts of some of the records seem to indicate some inflections. Furthermore, the ²¹⁰Pb evidence for the 72 m core does imply a slightly higher sedimentation rate. Therefore, the ²¹⁰Pb data should be treated as being equivocal on whether or not our assumption of a constant sedimentation rate is reasonable, but we cannot address this question with the available data. Another related limitation is that prior to ~1850 we lack adequate age constraints and point out that

assigning dates prior to this point simply assumes a linear sedimentation rate, and that time-dependent interpretations prior to this point must be treated with extreme caution. It is clear however that there are differences among the sites. For example the 232 m site has a much tighter CaCO_3 peak and the detailed changes in % CaCO_3 do not line up particularly well with the other sites. This issue may just reflect a sampling problem with this particular core or errors in our age model; however, the essence of our cautionary statement is that the earlier part of the record is susceptible to large uncertainties in age modeling, as are earlier interpretations of sedimentary chemical change. Even with the noted caveats it is worth mentioning that these limitations do not substantively alter the major conclusions of the paper, which are concentrated in the younger portion of the record.

3.2 Carbonate, organic carbon, and nitrogen.

The most prominent feature of our results for CaCO_3 is a broad maximum in each core that exceeds ~60% CaCO_3 and roughly coincides with the lake's high stand (**Figure 4**). This time period is also marked by the initiation of a gradual increase in proxy-based lake temperature (e.g., Tierney et al., 2010, **Figure 4**). Based on ESEM and XRD results from selected samples, the carbonate phase appears to be aragonite (**Figure 5A-E**). This aragonite does not appear to be entirely shell debris; rather it appears to be, at least partially, precipitated via another mechanism (e.g., **Figure 5C, D**). Although our limited sample analyses certainly indicate a component of non-shell aragonite, our results are insufficient to clearly define the aragonite component throughout the transect.

Because of the large variation in CaCO_3 , we have normalized the organic carbon contents to account for variable dilution, i.e., the corrected $C_{\text{org}} = C_{\text{org(measured)}} / (1 - f_{\text{CaCO}_3})$,

where f_{CaCO_3} is the fraction of sediment that is CaCO_3 . Without such a normalization all of the other sedimentary component distributions would be heavily influenced by variations in dilution by CaCO_3 as these components vary from ~ 0 to over 70% in our sediments. The corrected organic carbon record (C_{org}) generally mimics the CaCO_3 record (**Figure 6**), but the peaks at the deeper sites, and in particular the 232 m site, are less pronounced than the carbonate peaks.

The sediment organic carbon to nitrogen ratio averages $\sim 13.1 \pm 0.7$ for the samples from the ^{210}Pb cores (core A) and 13 ± 2 for the core B samples. There is no obvious offshore trend in these data (Appendix Table A2); however, there is a slight tendency for lower values prior to 1879 in the A cores with the more elevated values occurring from that period up to ~ 1950 and values then decline slightly (**Figure 7**). Also, the deeper lower ratios tend to be in the 72A and 120A meter records (**Figure 7**). However, for the B core samples, these trends are not apparent in the data (**Figure 8**). The previously published down-core sulfur data (corrected for carbonate dilution) show little systematic variability within each core, but there is a general relationship of increasing sedimentary sulfur contents with increasing water depth, which is consistent with what we would expect based on the sediments being increasingly reducing with water depth (Soreghan and Cohen, 1996; **Figure 2**).

3.3 Stable isotope records of carbonate and organic carbon.

The carbonate $\delta^{13}\text{C}$ record shows consistent values among the three cores with a steady decline from values of $\sim 4\text{‰}$ during the late 1800's to the present values of nearly 3‰ in the most recent sample (**Figure 8A**). The carbonate $\delta^{18}\text{O}$ record also shows something of a small decline from as high as 3.2‰ to values of $\sim 2.3\text{‰}$ over the course

of the record (**Figure 8B**). Although this record exhibits considerable scatter, particularly in the earlier part of the record, values since ~1800 are fairly uniform at $2.44 \pm 0.07\text{‰}$ (**Figure 8**) and are lower than the prior period ($2.75 \pm 0.18\text{‰}$).

When corrected for the Suess effect (e.g., Verburg, 2007; Castañeda et al., 2011), the $\delta^{13}\text{C}$ records of organic matter, show generally more ^{13}C enriched values prior to ~1700 compared to the more recent record (**Figure 8C**). Superimposed on the general pattern of $\delta^{13}\text{C}_{\text{org}}$ values over time is a pronounced oscillation with a minimum near the timing of the lake's ~1879 high stand (**Figure 8C**). The anomalously low values in the $\delta^{13}\text{C}_{\text{org}}$ records are also apparent in the $\delta^{15}\text{N}$ records near the time of the maximum in lake level (**Figure 8D**). Following this sharp excursion in the isotope data both isotope systems increase toward the present.

4.0 Discussion

4.1 Luiche Platform sedimentation

Prior work on Lake Tanganyika's sedimentation rates noted a broad range of ^{210}Pb -based rates since the 18th century ranging between 0.08 and 0.9 cm y^{-1} (McKee et al., 2005). Lower sedimentation rates ($\sim 0.2 \text{ cm y}^{-1}$ and lower) are generally characteristic of the more undisturbed watersheds or the earlier portions of these sediment records, i.e., pre-1960's; however, for sites off the northern Tanzanian and Burundi coasts there are indications of earlier increases in sediment accumulation rates (McKee et al., 2005). The patterns are variable, but increasing sedimentation rates in Lake Tanganyika's more recent sediment records have been attributed elsewhere to local land-use enhanced erosion (Alin and Cohen, 2003; McKee et al., 2005; O'Reilly et al., 2005). Our sites at 72 and 120 meters do not show a significant change in sedimentation rates in the upper

part of the records. The inferred constancy of sedimentation is consistent with the fact that our study sites lie to the south of Kigoma but north of the Malagarasi River whereas the study of McKee et al. (2005), which showed increases in sedimentation rates, focused more specifically on river delta sites from 50 to 126 m water depth. It is important to emphasize that our data remain equivocal on whether or not sedimentation rate has varied. For example, the upper 3 cm of data from our site at 120 m could imply a more recent acceleration in sedimentation, but given that these cores are all closely located along a common transect, the balance of our sedimentation rate data from the three shallowest sites do not support large-scale changes in recent sedimentation. Furthermore, for the earlier parts of the record (>10 cm, light gray data points in Figure 3) there is some indication that sedimentation rates differed compared to present. Our general assertion of more recent constant sedimentation is also based on the general agreement of the carbonate-based sedimentation rates and the radioisotope-based estimates. Although we cannot rule out the possibility of recent or more variable changes in sedimentation rates along the Luiche Platform, the considerably higher sedimentation rates measured in some of the river deltas of the central and northern boarder of Tanganyika's eastern boundary are not apparent in our data, which suggests that the regional rate of sedimentation is relatively constant between ~ 0.05 and 0.1 cm y^{-1} , with the slightly lower sedimentation rates coming from our deepest site. These values, and in particular the deepest rate, are also consistent with the value derived by Tierney et al. (2010), who reported a ^{210}Pb -based value of 0.05 cm y^{-1} for a multi-core located at 309 m water depth. The location of this sediment core was to the south of our study region but on the Tanzanian side of the lake.

4.2 Lake level and changes in carbonate sedimentation

Lake Tanganyika, like many other African lakes, has a well-documented history of varying lake level (e.g., Finney et al., 1996; Johnson et al., 1996; Alin and Cohen, 2003; Cohen et al., 2005; Talbot et al., 2006; Gasse, 2000; and others). Germane to the current work, the Little Ice Age is regarded as a dry period for many parts of Africa (Verschuren et al., 2000; Brown and Johnson, 2005; Russell and Johnson, 2007) with Lake Tanganyika's lake level being uncertain but frequently lower than observed over much of the past 2000 years (Alin and Cohen, 2003; Cohen et al., 2005). Following that period (~1550 – 1850) there is a notable lake high stand, which peaks around 1879 followed by a rapid decline to values near the present lake level (see **Figure 4**, and Alin and Cohen, 2003 from Birkett et al., 1999; and Evert (1980)). As discussed earlier, the gradual rise and rapid fall of the lake level was likely generated by a combination of increasingly wet conditions that followed the LIA, causing the lake level to rise, followed by a breaching and subsequent erosion of the current lake outlet (see a detailed review in Nicholson, 1999). Coincidentally, this period is also when surface water temperatures began to increase from ~23 to 26°C (Tierney et al., 2010, **Figure 4**). We note here the caveat to this latter statement that Kraemer et al. (2015) argued that the TEX₈₆ proxy temperature record likely overestimates the magnitude and rate of warming but not that warming has occurred. Lake level does appear to have fluctuated from ~1880 to the present; however, those fluctuations were considerably smaller than the rise, peak, and decline coming out of the LIA (Alin and Cohen, 2003).

Tanganyika's shallow lake floor sediments are presently rich in a variety of carbonate deposits, including high magnesium carbonates and sedimentary aragonite

layers (Stoffers and Hecky, 1978; Cohen and Thouin, 1987; Cohen et al., 1997). The presence of these carbonate phases is consistent with the fact that the lake is saturated with respect to various carbonate phases ($\Omega = 6.5$ for calcite, 3.0 for 20% Mg, and 4.4 for aragonite, Cohen et al., 1997), and the lake's chemical composition (Müller et al., 1972). Over long timescales, the chemical conditions that generate the first-order relationship between carbonate sedimentation and lake level is likely caused by inflow from alkaline river systems, most notably the Rusizi River, which drains the Lake Kivu basin. The primary dependence on the Rusizi River stems from the fact that this river drains the alkaline Lake Kivu and supplies a significant fraction of many of Tanganyika's major elements and maintains the lake's carbonate chemistry at a level of supersaturation (Craig, 1974; Stoffers and Hecky, 1978; Haberyan and Hecky, 1987; Cohen et al., 1997; Vandellannoote et al., 1999). Supply from Kivu, however, requires wet climate conditions north of Lake Tanganyika (Stoffers and Hecky, 1978; Felton et al., 2007), which provides the somewhat counterintuitive linkage between lake level and carbonate chemistry. Given the large volume of Lake Tanganyika, it is unclear if a short-term increase in lake level regardless of the water source and its chemical composition would be the sole driver of the broad carbonate peak identified in this study, even though it is coincident with the lake's high stand. Despite that intuition, there is a fairly clear switch from low lake stand, low carbonate during the (dry) LIA followed by a rapid carbonate sedimentation event coincident with the lake stand maximum followed by a lowering of sedimentary carbonate contents with again a lowering of lake level (**Figure 4**).

The XRD and SEM analyses both indicate that the dominant carbonate phase is aragonite, and the SEM results indicate that this aragonite is unlikely to be exclusively

biogenic shell material (**Figure 5**); at least for the limited samples that we have examined. It is certainly likely that there is ostracode debris as well within the carbonate layer, but our initial investigations are equivocal on this point. The carbonate peak occurs at all water depths presented here and that the maximum is nearly coincident at all of the depths within the resolution of the age models. Another observation of our work is that this change in lake chemistry is a prolonged event (decades), which dominates this particular region of Lake Tanganyika.

The carbonate $\delta^{13}\text{C}$ decline reflects the influence of the low $\delta^{13}\text{C}$ value of anthropogenic CO_2 (e.g., Quay et al., 1992). There are certainly a variety of chemical processes that could influence the magnitude of the $\delta^{13}\text{C}$ signature within the sediments, for example changes in pH, mixing with older deeper waters, or other non-equilibrium processes (Spero et al., 1997; Bauch et al., 2000). However, the important point for this manuscript is that the carbonate ^{13}C decline clearly reflects the impact in changing lake chemistry due to atmosphere-lake CO_2 exchange and that change is apparent in the lake record since the late 1800's. A more detailed treatment of this issue is beyond the scope of the paper and the available data, as well as being secondary to our primary interest in the impact of the lake transgression and regression.

The carbonate $\delta^{18}\text{O}$ and the $\delta^{13}\text{C}$ results indicate that there is little isotopic difference among the three cores that span the full depth range that we sampled, with perhaps the exception of some of the data from the mid-1600's (**Figure 8**). Because all three cores converge on common $\delta^{18}\text{O}$ and the $\delta^{13}\text{C}$ values, particularly over the past 200 years, it would appear that carbonate precipitation occurred at a common depth, most likely the lake's surface. This interpretation is based on the fact that there is a water

column depth gradient in both these isotope systems (**Figure 2**) that should be apparent if the carbonate is precipitating at the lake floor at each site. In combination our results indicate that during the extreme high stand at ~ 1879, lake surface waters became highly favorable to the precipitation of carbonate (**Figure 4**), at least regionally. The fact that the lake also began to warm in the late 19th Century (**Figure 4**) should further foster conditions favorable to carbonate precipitation. Even though these data imply chemical conditions favorable for carbonate precipitation beginning in the early 1800's with a maximum centered in the late 1800's, we do not know the full spatial extent of the carbonate-rich layer throughout the lake, whether this layer is confined to this particular portion of the lake or present throughout. However, prior work noted the presence of a distinct carbonate layer in the early part of their sediment record (Tierney et al., 2010, supplemental material). This added observation, coupled with prior observations of distinct carbonate layers and deposits, implies that the carbonate layer we observe is not unique to the Luiche Platform region.

We also note that previous high stands (< ~1750) do not appear to have coincident changes in carbonate sedimentation (**Figure 4**). However, these earlier high stands are much less well constrained as compared to the more recent instrumental record and none of those high stands appeared to reach the levels of the 1879 event.

4.3 Sedimentary organic matter

One possible explanation that we need to consider for the increase in carbonate sedimentation is that primary production increased with increasing lake level; although we will argue against this possibility below. High photosynthetic production could drive the lake's pH toward more alkaline conditions (e.g., see review in Dittrich and Obst,

2004) stimulating an increase in carbonate precipitation and burial. As noted above, the modern lake is indeed already primed for carbonate precipitation having alkaline surface waters of ~ 6.5 meq L⁻¹ and a high pH of ~ 9 (Craig et al., 1974; Edmond et al., 1993; **Figure 2**), and a further stimulus of a change in lake chemistry, regardless of the mechanism, could promote carbonate precipitation. This idea of an increase in productivity associated with a lake level high stand is consistent with the longer record of Tanganyika's relationship between productivity and lake transgressions (Talbot et al., 2006).

The question arises whether maxima in organic carbon are driven by increased productivity or enhanced preservation. Carbonate corrected organic carbon contents of the sediments also exhibit a maximum around the same time period as the maxima in CaCO₃, suggesting a concomitant productivity peak (**Figure 6**). Upon closer inspection, it appears that the carbonate peaks may actually lead the organic carbon peaks, at least for the 72 and 120 meter sites, which are the sites having the highest data resolution (**Figure 9**). Although this lag does not remove the possibility of a primary productivity driven forcing on the carbonate system, the order of the increase (carbonate first) seems to imply that enhanced organic carbon preservation may have been influenced by the increase in sedimentary carbonate. This idea is similar to that suggested by Hodell and Schelske (1998), who showed that calcite precipitation in Lake Ontario may be an effective transfer vector for shuttling organic matter to the lake floor, although we would still have anticipated that these two signatures would have varied in concert.

An important caveat to the idea that concomitant carbonate and organic carbon peaks are driven by a productivity change is that the longer-term biogenic silica record

does not closely mirror the organic carbon and carbonate records (**Figure 6**, Tierney et al., 2010). The first order interpretation of the biogenic silica record shows high values between ~ 1700 and the lake high stand at 1879, after which there is a decrease with values remaining generally lower. However, upon more detailed inspection, there is a minimum in normalized biogenic silica values around the early to mid-1800s followed by progressively higher values just prior to the maximum in lake level (**Figure 6**). Thus, the detailed expression of the biogenic silica record is one where there are multiple biogenic silica maxima prior to the high stand and that the most recent maximum appears to slightly precede the lake level high stand. At face value this observation would imply that the lake transgression may result in a lowering of biogenic silica production. However, differences in the age models, locations and depths of the study sites make a more nuanced comparison of these records imprudent at this time, but it is difficult to reach an unequivocal conclusion concerning the relationship among organic carbon, biogenic silica, primary productivity, and the carbonate record. Nevertheless, there is some evidence to support elevated productivity (elevated organic carbon and perhaps biogenic silica) near the time of the onset of the elevated carbonate record; however, as discussed further below, there is also strong evidence to suggest that primary production is actually lower during much of the lake level transgression.

Another possible mechanism that could foster organic carbon preservation is through changes in the redox conditions of the sediment (Hartnett et al., 1998; Hartnett and Devol, 2003). The idea is that degradation of organic material is retarded under more reducing conditions. Given the close proximity of the oxygen-sulfide boundary within the water column to our coring depths (**Figure 2**), it is conceivable that changes in the

intensity of the lake's reducing character might drive changes in organic carbon burial. One possible expression of such variation would be changes in sedimentary sulfur burial; however, there are no obvious oscillations in the carbonate corrected sulfur record that might imply shifts in the lake's redox conditions over time (**Figure 7**). The absence of a sulfur signature leads us to conclude that redox oscillations are not the driver of the organic carbon peak.

4.3 $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ of Organic Matter

The $\delta^{13}\text{C}$ record of the organic matter, corrected for the Suess effect, exhibits a strong negative excursion that reached its peak at the lake high stand. Our Suess effect correction follows the formulation outlined previously (Verburg, 2007). This $\delta^{13}\text{C}$ excursion is coincident with a decline in the $\delta^{15}\text{N}$ record as well as with the increase in $\%C_{\text{org}}$ (**Figure 8**). In terms of utilizing $\delta^{13}\text{C}$ as a productivity tracer, more ^{13}C enriched organic matter in lake systems is generally interpreted as less isotopic discrimination between the source CO_2 and the photosynthate, which occurs under higher rates of primary production because of a greater degree of CO_2 limitation (e.g., Hecky and Hesslein, 1995; Hodell and Schelske, 1998; O'Reilly et al., 2003; Torres et al., 2012). Thus, as primary production lessens, isotope discrimination increases (Hecky and Hesslein, 1995). Following this line of reasoning the $\delta^{13}\text{C}$ record is interpreted as a decline in primary production with a minimum centered at the lake high stand. Although the organic carbon record seems to be tracking the isotope records throughout that period quite closely (**Figure 8**), it is important to emphasize that the increase in organic carbon is opposite to what we would have expected with a decline in production.

There are likely to be alternative explanations for the $\delta^{13}\text{C}$ record as well, for

example one idea posits that a lake transgression could lead to enhanced delivery of light dissolved inorganic carbon from run off (e.g., Meyers and Teranes, 2001). It is also possible that the isotope signature reflects a greater fraction of terrestrial plants, which would be consistent with the $\delta^{15}\text{N}$ data (see discussion below). Although we cannot completely exclude this scenario the lack of a consistent increase in the C:N ratio within our full data set argues against a significant change in terrestrial C_{org} contribution (**Figure 8**), which we would expect if the isotope shift was driven by variable carbon sources rather than rates of primary productivity (e.g., Meyers and Ishiwatari, 1993; Meyers and Teranes, 2001). However, this somewhat classic view of C:N stoichiometry must be treated with caution as well because in lakes the C:N ratio is considerably more elastic than in marine environments, and there are multiple pathways to altering or maintaining this ratio (e.g., Corzo and Niell, 1991; Hecky et al., 1993).

The $\delta^{15}\text{N}$ record could be interpreted as a change in the primary nitrogen source to the lake. Assuming that the lake is probably highly susceptible to nitrogen limitation, and assuming that nitrogen fixation is an important source of this nutrient to the lake as previously suggested (Hecky et al., 1991) an increase in nitrogen fixation could result in a lowering of the $\delta^{15}\text{N}$ value (e.g., Leng et al., 2005). An alternative explanation is that there is an increase in the terrestrial organic matter contribution to the sediment (Meyers and Ishiwatari, 1993); however, as mentioned, we might have expected more variations in the C:N ratio if this were the case. At face value an increase in nitrogen fixation would probably be the favored interpretation of the $\delta^{15}\text{N}$ data. Indeed, we note that the phytoplankton community in Lake Tanganyika exhibits low $\delta^{15}\text{N}$ values during certain periods of time (-1 to 1‰, O'Reilly et al., 2004), and these low values are consistent with

a nitrogen fixation source. There are, however, exceptions to the lower $\delta^{15}\text{N}$ values in Lake Tanganyika in that the plankton can exhibit elevated $\delta^{15}\text{N}$ values as well (6 – 9‰, O'Reilly et al., 2004). These ^{15}N -enriched values are presumed to derive from an upwelling source of nitrogen (O'Reilly et al., 2004), which is consistent with similar interpretations from Lake Malawi (Francois et al., 1996) and Lake Kivu (Morana et al., 2015). During Lake Kivu's dry season, vertical mixing produces $\delta^{15}\text{N}$ signatures suggestive of an upwelling nitrogen source, whereas during the rainy season $\delta^{15}\text{N}$ signatures and cyanobacteria abundances reflect a dominant atmospheric nitrogen source (Morana et al., 2015). In light of these results, our data seem to imply that there was an increase in stratification rather than upwelling with a parallel overall increase in nitrogen fixation that resulted in the excursion to lower $\delta^{15}\text{N}$ values during the lake high stand.

One possible explanation of the full data set is that there is an increase in organic carbon preservation at the time of the transgression, an increase in the proportion of nitrogen supply from nitrogen fixation, and a decline in primary production. There are clearly multiple additional possibilities that could be contributing to these signatures as well, and there is no particular reason that any single explanation need satisfy the entire data set. The use of any individual chemical signature presented here as a productivity proxy must be treated cautiously, however. As examples, both $\delta^{15}\text{N}$ and $\delta^{13}\text{C}_{\text{org}}$ can be influenced by changes in source materials, variations in the terrestrial contributions, and their isotope signatures, as well as diagenesis, and all of these specific processes could lead to variations in the isotope signature of organic matter that would produce the observed sedimentary signature (e.g., Fry, 1986; Hodell and Schelske, 1998; O'Reilly et al., 2002; Torres et al., 2012).

5.0 Conclusions

Although watershed activities have caused increases in sedimentation in some locations in Lake Tanganyika, sediments on the Luiche Platform are marked by generally near-constant sedimentation over the past 150 years ($0.09 \pm 0.02 \text{ cm y}^{-1}$) as measured at three sites between ~70 – 120 meters water depth. At ~1879 lake level is thought have been at a maximum and during or immediately following this period sedimentary carbonate concentrations were at a maximum. Although we relate these signals to lake level, it is important to emphasize that we do not know how the precipitation balance might have been altered following the peak in lake level. Carbonate corrected organic carbon also peaks around the same time period, but generally seems to lag the carbonate record. We note here the possibility that the organic carbon record actually reflects a preservation signature, likely because of a preservation effect from the carbonate. The $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ records of organic matter, which show a minimum around the time of the lake level maximum, although coincident with the organic carbon and carbonate records are likely to be separate from these processes. These isotope records seem to imply a lowering of primary production along with an increase in the reliance on nitrogen fixation as the source of fixed nitrogen to the lake.

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Figure Captions

Figure 1. Map of study area. Inset shows locations of multicore deployments on the Luiche Platform. Base map was generated using Demis Web Map (<http://www.demis.nl>) and modified after Brucker et al. (2011).

Figure 2. Water column temperature, chloride, dissolved oxygen, sulfide, alkalinity, pH, $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ plotted as a function of depth. Temperature, alkalinity, chloride, $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ data are from Craig et al. (1974), and pH, O_2 , and sulfide data are from Edmond et al. (1993). Dark dashed lines indicate water depths from which multicores were taken.

Figure 3. $\text{Ln } ^{210}\text{Pb}_{\text{xs}}$ (squares) and ^{137}Cs (diamonds) plotted as a function of sediment depth. Light gray symbols were not used in $^{210}\text{Pb}_{\text{xs}}$ fits to calculate sedimentation rate. Dark hashed lines represent depths of the ^{137}Cs maximum depths.

Figure 4. The $\%\text{CaCO}_3$ contents plotted as a function of calendar year (based on the age models described in the text). For the bottom panel, lake level data are from the compilation presented previously (Cohen et al., 2005; Alin and Cohen, 2003) and the proxy temperature data are from Tierney et al. (2010). Note that MC1 and KH1 are overlapping cores having the temperature proxy data (Tierney et al., 2010) and those core designations are simply maintained here for identification purposes.

Figure 5. Scanning Electron Microscope images for samples taken from 107 meters depth at 17 cm (A) and 32 cm (B) and from 120 meters depth at 16 cm (C), 21 cm (D) and 16 cm (E) at different magnifications.

Figure 6. (Top four panels) The % organic carbon contents corrected for carbonate dilution plotted as a function of calendar year (based on the age models described in the text). **(Bottom two panels)** Lake level data are from a compilation in Cohen et al., (2005) and Alin Cohen (2003) and the proxy temperature data and biogenic silica data are taken from Tierney et al. (2010).

Figure 7. (A) C:N ratio of the organic matter (core A) and **(B)** %S corrected for carbonate dilution (core B data) plotted as a function of calendar year. Note the dashed horizontal black line indicating 1879, and the light gray vertical line in the C_{org}:N plot indicates the average C_{org}:N ratio of all the sediments for the A cores.

Figure 8. (A) $\delta^{13}\text{C}$ of carbonate, (B) $\delta^{18}\text{O}$ of carbonate, (C) $\delta^{13}\text{C}$ of organic matter, (D) $\delta^{15}\text{N}$ of sediment, (E) % organic carbon (corrected for carbonate dilution), and (F) the organic carbon to nitrogen ratio of the organic carbon from three cores plotted as a function of time. Heavy dashed line indicates the approximate year of high lake level (1879). Note that the values of $\delta^{13}\text{C}$ of organic matter are corrected for the Suess effect as described in the text. The dashed gray line indicates 1879.

Figure 9. The calcium carbonate and organic carbon (carbonate corrected) concentrations

834 plotted as a function of time. Note that the increase in organic carbon lags behind that of
835 carbonate in all three cores.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5A

Figure 5B

Figure 5C

HV	Spot	Mag	Det	Sig	WD	Pressure	—10 μ m—	
30.0 kV	4.5	3787x	Lfd	SE	11.07 mm	80.0 Pa		

Figure 5D

Figure 5E

Figure 6

Figure 7

Figure 8

Figure 9

Table 1. Age model summary.

Core depth (meters)	^{210}Pb rate (cm y ⁻¹)	^{137}Cs rate (cm y ⁻¹)	$^1\text{Final}$ (cm y ⁻¹)
72	0.12	0.07	0.07
107	0.10	0.09	0.09
120	0.07	0.09	0.09
232	0.10	² na	0.08
335	0.06	² na	0.05

¹Final sedimentation rate results from aligning CaCO₃ peaks as described in text.

²na indicates that there was insufficient data to derive a sedimentation rate.