Title: Separating ITCZ- and ENSO-related rainfall changes in the Galápagos over the last 3 kyr using D/H ratios of multiple lipid biomarkers

Keywords: paleoclimate, lipid biomarker, hydrogen isotope ratios, ENSO, ITCZ, tropical Pacific, Holocene

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Abstract: We present a 3,000-year rainfall reconstruction from the Galápagos Islands that is based on paired biomarker records from the sediment of El Junco Lake. Located in the eastern equatorial Pacific, the climate of the Galápagos Islands is governed by movements of the Intertropical Convergence Zone (ITCZ) and the El Niño-Southern Oscillation (ENSO). We use a novel method for reconstructing past ENSO- and ITCZ-related rainfall changes through analysis of molecular and isotopic biomarker records representing several types of plants and algae that grow under differing climatic conditions. We propose that δD values of dinosterol, a sterol produced by dinoflagellates, record changes in mean rainfall in El Junco Lake, while δD values of C34 botryococcene, a hydrocarbon unique to the green alga Botryococcus braunii, record changes in rainfall associated with moderate-to-strong El Niño events. We use these proxies to infer changes in mean rainfall and El Niño-related rainfall over the past 3,000 years. During periods in which the inferred change in El Niño-related rainfall opposed the change in mean rainfall, we infer changes in the amount of ITCZ-related rainfall. Simulations with an idealized isotope hydrology model of El Junco Lake help illustrate the interpretation of these proxy reconstructions. Opposing changes in El Niño- and ITCZ-related rainfall appear to account for several of the largest inferred hydrologic changes in El Junco Lake. We propose that these reconstructions can be used to infer changes in frequency and/or intensity of El Niño events and changes in the position of the ITCZ in the eastern equatorial Pacific over the past 3,000 years. Comparison with El Junco Lake sediment grain size records indicates general agreement of inferred rainfall changes over the late Holocene.
Response to the Editor

The authors would like to thank the editor for her constructive comments and time in reviewing this manuscript. Below are our responses to the editor’s comments. We hope the revisions are to your satisfaction.

Editor’s Comments:

As per EPSL data policy, a revised paper should include a supplemental table of the data shown in the figures that has not been previously published. Alternatively, you may submit the data to a publicly accessible database or repository such as PANGEA and indicate in the paper where the data can be accessed. If you decide to use a publicly accessible database, please provide a reference to this database in the results section of the revised manuscript.

Response: The data have been submitted to PANGEA (http://www.pangaea.de) and the database referenced in the results section of the manuscript (Lines 305 and 362-363) as well as at the end of the manuscript (Line 611).

EPSL has a limit of 10 figures and tables. You will need to move 3 of these display items to the supplement.

Response: The Supplemental Figures and Tables have been imbedded into the Supplementary Material so that the total number of display items in the main manuscript is 10.
Separating ITCZ- and ENSO-related rainfall changes in the Galápagos over the last 3 kyr using D/H ratios of multiple lipid biomarkers

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ABSTRACT

We present a 3,000-year rainfall reconstruction from the Galápagos Islands that is based on paired biomarker records from the sediment of El Junco Lake. Located in the eastern equatorial Pacific, the climate of the Galápagos Islands is governed by movements of the Intertropical Convergence Zone (ITCZ) and the El Niño-Southern Oscillation (ENSO).

We use a novel method for reconstructing past ENSO- and ITCZ-related rainfall changes through analysis of molecular and isotopic biomarker records representing several types of plants and algae that grow under differing climatic conditions. We propose that δD values of dinosterol, a sterol produced by dinoflagellates, record changes in mean rainfall in El Junco Lake, while δD values of C_{34} botryococcene, a hydrocarbon unique to the green alga *Botryococcus braunii*, record changes in rainfall associated with moderate-to-strong El Niño events. We use these proxies to infer changes in mean rainfall and El Niño-related rainfall over the past 3,000 years. During periods in which the inferred change in El Niño-related rainfall opposed the change in mean rainfall, we infer changes in the amount of ITCZ-related rainfall. Simulations with an idealized isotope hydrology model of El Junco Lake help illustrate the interpretation of these proxy reconstructions.

Opposing changes in El Niño- and ITCZ-related rainfall appear to account for several of the largest inferred hydrologic changes in El Junco Lake. We propose that these reconstructions can be used to infer changes in frequency and/or intensity of El Niño events and changes in the position of the ITCZ in the eastern equatorial Pacific over the past 3,000 years. Comparison with El Junco Lake sediment grain size records indicates general agreement of inferred rainfall changes over the late Holocene.
1. Introduction

The climate dynamics of the tropical Pacific play a fundamental role in climate variability across the globe. Two primary features of the tropical Pacific climate are the Intertropical Convergence Zone (ITCZ) and the El Niño/Southern Oscillation (ENSO). The ITCZ is the circum-global zone of convergence of the northern and southern trade winds that results in a band of heavy rainfall near the equator. Its position shifts latitudinally with the seasons following the zone of maximal solar insolation and sea surface temperature (SST). ENSO is characterized by irregular warming and cooling of the eastern equatorial Pacific (EEP) sea surface that typically lasts 1–1.5 years, recurs every 3–7 years and is associated with changes in sea level pressure and precipitation across the tropical Pacific. Associated atmospheric teleconnection patterns arise from ENSO events that influence climate on a global scale (Trenberth et al., 1998).

Some studies suggest that the ITCZ and ENSO have changed dramatically over the Holocene, however these changes are poorly constrained due to the low temporal and spatial coverage of paleoclimate records from regions where the fundamental dynamics of ENSO and the ITCZ operate. Paleo-rainfall records from the tropical Pacific can aid in these reconstructions, provided that the various climate features that influence local rainfall can be distinguished. El Junco Lake (Fig. 1) is located on San Cristóbal Island,
Galápagos (1° S, 89° W) in the heart of the EEP cold tongue with a climate highly sensitive to movements of the ITCZ and to ENSO. In this study we present a unique method of disentangling the ITCZ and ENSO rainfall signals from algal and plant lipid biomarkers and their hydrogen isotope ratios in this lake to infer changes in ENSO and the position of the ITCZ over the late Holocene.

Located at the southern edge of the ITCZ, seasonality at El Junco Lake is characterized by the annual migration of the ITCZ, which produces a wet season from January-May and a dry season from June-December (Fig. 2). The wet season occurs when the ITCZ is shifted southward–weakened trade winds and reduced ocean upwelling cause the EEP cold tongue to recede, sea surface temperature (SST) and air temperatures to increase, and the resultant convection results in increased rainfall. Conversely, the dry season occurs as the ITCZ shifts northward–stronger trade winds and upwelling result in lower SST and air temperatures and minimal convection. At elevations higher than approximately 250 m (including El Junco Lake at 670 m), the dry season is characterized by significantly higher annual rainfall than lower elevations as strong upwelling and cool SSTs result in cooler air at the surface than aloft, which creates a temperature inversion that produces persistent stratus clouds (called garúa) that blanket the highlands (Trueman and d’Ozouville, 2010). It is estimated that approximately 25% of rainfall in the Galápagos highlands comes from garúa (Pryet et al., 2012).

Rainfall in the Galápagos Islands also varies in response to SST anomalies (SSTAs) associated with ENSO. Annual rainfall is correlated to warm Niño 3.4 SSTAs, with the highest correlation observed during moderate-to-strong El Niño events (Niño 3.4 SSTAs ≥ 1.0 °C; r = 0.73, p < 0.05; Fig. 3). The historical record of station rainfall from
the Galápagos demonstrates that strong El Niño events are marked by annual rainfall that is 4–7 times as large as rainfall during non-El Niño years, while La Niña events have little influence on local rainfall (Fig. 4). The historical rainfall record also demonstrates that year-to-year variations in wet season rainfall in the Galápagos generally reflect the large-scale pattern of rainfall in the eastern Pacific along the equator (1S:1N, 110E:85E) irrespective of ENSO phase, which suggests that the ITCZ influences Galápagos rainfall on interannual timescales (in addition to driving the wet/dry seasons). Because rainfall in this region is highly dependent on both the position of the ITCZ and El Niño events, rainfall reconstructions from this region should take into account possible changes in both of these climate phenomena.

In this study we use algal biomarker distributions and hydrogen isotope ratios from El Junco Lake sediment to infer past changes in local rainfall. Hydrogen isotope ratios of lipid biomarkers from algae are a valuable tool with which to investigate past climate changes (Pahnke et al., 2007; van der Meer et al., 2008; Sachs et al., 2009; Smittenberg et al., 2011; Sachse et al., 2012). In closed (endorheic) lakes, lake water δD is driven by the ratio of precipitation to evaporation (Craig, 1961) and the isotopic value of the precipitation which, in the tropics, is strongly negatively correlated with the amount of precipitation on monthly or longer timescales (Dansgaard, 1964; Risi et al., 2008). The combination of these processes amplifies the response of lake water δD to changes in rainfall with lower (higher) δD values occurring during periods of wetter (drier) conditions. Changes in rainfall patterns can therefore be reconstructed from δD values of sedimentary algal lipids, due to the near perfect correlation between water δD and algal lipid δD values (Sauer et al., 2001; Sachse et al., 2004; Englebrecht and Sachs,
In regions where multiple climate phenomena influence local rainfall patterns, paleo-rainfall records can be difficult to interpret. In this study we present a novel method to distinguish changes in mean rainfall from changes in El Niño-related rainfall from the Galápagos Islands using a suite of molecular and isotopic records from El Junco Lake.

Concentration and δD profiles of dinosterol, a lipid biomarker produced by dinoflagellates, are compared to concentration and δD profiles of C$_{34}$ botryococcoene, a biomarker produced by the B Race of the green algae, Botryococcus braunii (Fig. S1). In addition, abundances of C$_{30}$ α20-keto-1-ol produced by ferns are compared to abundances of the algal C$_{30}$ α16-keto-1-ol in the sediment record. Biomarker concentrations and δD values derived from the same sediment samples are compared, thus eliminating potential age differences in the derived climate signals. These biomarker records are used to reconstruct changes in mean rainfall and El Niño-related rainfall for the past 3,000 years.

2. Methods

2.1 Study site

El Junco Lake is located atop a caldera in the highlands of San Cristóbal Island, Galápagos (1° S, 89° W) at 670 m elevation. With the exception of possible overflow conditions and seepage, it is an endorheic lake whose hydrologic fluxes are dominated by precipitation and evaporation. A channel cuts through the lowest point of the crater rim.
that was located approximately 2–3 m above the lake level during the field outing in Sept. 2004. El Junco Lake is the only permanent freshwater lake in the Galápagos Islands and during our 2004 field outing, the lake had a diameter of 280 m, maximum depth of 6 m, and temperature of 19 °C. The lake chemistry is characterized by a conductivity of 20–27 µS/cm, pH ca. 5.5 and is mesotrophic (Colinvaux, 1968; Ferrington and Pehofer, 1996).

The presence of strong easterly trade winds combined with the relatively shallow depth result in the lake being well mixed, as indicated by nearly constant temperature, O₂, and δD depth profiles.

2.2 Field methods, age model, biomarker measurements, and idealized lake model

Details of the field methods, dating methods, and age model can be found in the Supplementary Methods (Table S1, Table S2) and Zhang et al. (subm.). The age model was constructed in OxCal 4.0.1 (Bronk Ramsey, 2009) using 15 ²¹⁰Pb measurements with a constant rate of supply model and 21 ¹⁴C measurements with linear interpolation between data points and the SHCa104 calibration curve. The age model predicts a basal age of the sediment cores of 9.1 kyr BP and a sedimentation rate that ranges from 2.3 mm/yr (1 cm ≈ 5 years) in the top 5 cm and declines to 0.2 mm/yr (1 cm ≈ 50 years) for the lower 1 m. No significant reservoir effect is thought to exist in El Junco Lake, an assertion supported by the presence of nuclear bomb-derived ¹⁴C in the upper 8 cm of sediment and the smooth transition from ²¹⁰Pb-derived- to ¹⁴C-derived-age models (Table S1, and Fig. 5 in Zhang et al., subm.). Estimates of age model uncertainty can be found in the Supplementary Methods and Tables S1 and S2.

Purification, quantification, and δD measurement methods for dinosterol can be
found in the Supplementary Methods and in Atwood and Sachs (2012) and for C\textsubscript{34}
bottyococcene can be found in Zhang and Sachs (2007), Zhang et al. (2007), and Zhang
et al. (subm). Details of the idealized model of El Junco Lake can also be found in the
Supplementary Methods.

3. Results and discussion

3.1 Botryococcene and dinosterol sources and their response to changing hydrologic
conditions in El Junco Lake

Botryococcenes, in particular C\textsubscript{34} botryococcene, represent some of the most
abundant lipids in El Junco Lake sediment and are produced by a single race of the green
alga, Botryococcus braunii (Metzger et al., 1985; Zhang et al., 2007). Although the
growth conditions of B. braunii have not been documented in El Junco Lake, this algae
has been found to thrive in oligotrophic conditions (e.g. in oligotrophic lakes and during
summer stratification of meso-eutrophic temperate lakes), while its abundance has been
found to decrease with increasing NH\textsubscript{4} and total nutrient concentration (Huszar and
Caraco, 1998; Huszar et al., 2003; Smittenberg et al., 2005). This evidence suggests that
B. braunii blooms during periods of oligotrophic conditions in El Junco Lake.

Dinosterol is another highly source-specific and abundant biomarker found
throughout El Junco Lake sediment. The sterol composition of the sediment indicates that
a dinoflagellate of the genus Peridinium has been the source of dinosterol in El Junco
Lake throughout the sedimentary record (Atwood et al., 2014). Dinoflagellates of this
genus have been found to thrive in a wide range of nutrient conditions (Trigueros et al.,
suggesting that the dinoflagellate and *B. braunii* adopt different ecological niches and thus grow under differing environmental conditions in El Junco Lake.

The sedimentary biomarker profiles in El Junco Lake further support the theory that the organisms producing dinosterol and C$_{34}$ botryococcene grow in differing environmental conditions. Firstly, the C$_{34}$ botryococcene and dinosterol concentration profiles are highly dissimilar (Fig. 6A). In addition, the log of C$_{34}$ botryococcene concentration is anti-correlated to its δD value ($r = -0.62$, $p < 0.05$) and is highly variable, with a concentration range that spans five orders of magnitude and falls below the detection limit of 0.1 μg/g in several intervals in the sedimentary record (Fig. 5, Fig. 6A).

These characteristics suggest that *B. braunii* thrives during periods of higher rainfall and that blooms are highly sensitive to changing environmental conditions. In contrast, the concentration of dinosterol is far more uniform through the 3 kyr sediment record (Fig. 6A), suggesting that the dinoflagellate thrives in a wider variety of environmental conditions as compared to *B. braunii*. Furthermore, its concentration is positively correlated to its δD value ($r = 0.51$, $p < 0.05$), although with a lower slope and correlation coefficient than the C$_{34}$ botryococcene data (Fig. 5), from which we infer a preference for drier conditions than *B. braunii*.

Variations in nutrient concentration and water column stability are the most probable sources of the dissimilar relationships between the concentrations and δD values of C$_{34}$ botryococcene and dinosterol in El Junco Lake. In particular, conditions favorable for *B. braunii* blooms are thought to occur during El Niño events in association with three distinct processes that promote oligotrophic conditions and lake stratification. Firstly, the
heavy rains associated with moderate-to-strong El Niño events are thought to dilute the nutrient pool and cause oligotrophic conditions. El Junco Lake is an endorheic lake (with the exception of occasional overflow conditions and possibly seepage) located within a caldera with a small catchment area consisting only of the narrow crater rim (Conroy et al., 2008). In such lakes the concentration of nutrients tend to decrease as lake levels increase through dilution of the incoming nutrients (Smol et al., 2001; Magyari et al., 2009). Because monthly rainfall in this area can be an order of magnitude higher during strong El Niño events as compared to non-El Niño periods (c.f., Fig. 4) and the lake has been reported to overflow during strong El Niño events, considerable dilution and flushing of nutrients likely occur during these times. Secondly, reduced nutrient input to the lake exacerbates oligotrophic conditions during strong El Niño events. A major source of nutrients to the lake is thought to be guano deposited directly into the lake by the large number of seabirds that frequent it (Colinvaux, 1968). Such a predominance of avian-derived nutrients has been documented in a number of lake systems (Manny et al., 1994; Marion et al., 1994). However, during strong El Niño events, this nutrient source is diminished as the Galápagos seabird population plummets in response to the massive fish die-offs associated with a deep thermocline and low ocean productivity in the EEP (Gibbs, 1987). Finally, El Niño events in the EEP are associated with conditions that increase water column stability and lake stratification, including warmer air temperatures, reduced cloud cover, and reduced trade wind strength (Lewis Jr., 1983; O’Reilly et al., 2003; Ndebele-Murisa et al., 2010; Cózar et al., 2012), conditions which have been shown to exacerbate nutrient depletion in the surface of oligotrophic lakes (O’Reilly et al., 2003).
Increases in water column stability during El Niño events should further promote favorable conditions for the oil-rich, colony-forming *B. braunii* due to their buoyancy. In various locations across the globe, *B. braunii* blooms have been strongly linked to increases in water column stability due to both their tolerance of oligotrophic conditions and their anti-sinking strategy (Kebede and Belay, 1994; Souza et al., 2008; Winder and Hunter, 2008). In particular, in a Columbian lake with conditions (including depth, pH, conductivity, and nutrient concentration) comparable to El Junco Lake, *B. braunii* blooms have been found to occur during the annual flood stage in association with nutrient flushing, oligotrophic conditions, and lake stratification (Pinilla, 2006), conditions that closely mimic moderate to strong El Niño events in El Junco Lake.

These lines of evidence strongly suggest that conditions favorable for *B. braunii* growth occur during moderate-to-strong El Niño events and thus that C$_{34}$ botryococcene δD records the hydrologic conditions of the lake during these events. In contrast, the available evidence suggests that the dinoflagellate population persists throughout the varying hydrologic conditions experienced by El Junco Lake and thus that sedimentary dinosterol δD records the long-term (decadal-to-centennial) mean lake conditions.

While we believe that our interpretation of the biomarker data is the most straightforward interpretation given the available data, we acknowledge the existence of alternative interpretations. In particular, as the modern dinoflagellate population dynamics have not been documented in El Junco Lake, it is possible that the dinoflagellates do not grow during El Niño events, and thus that sedimentary dinosterol δD values reflect the mean non-El Niño conditions of the lake. However, the construction of the climate indices and fundamental conclusions should be insensitive to such a
modified interpretation of the proxy data. In addition, while there is substantial evidence
to suggest that both physical and trophic conditions restrict *B. braunii* blooms to
moderate-to-strong El Niño events, we cannot rule out the possibility that oligotrophic
conditions that accompany a substantially wetter mean climate (e.g. that give rise to a
perpetually overflowing lake) are sufficient to drive *B. braunii* blooms. If this were the
case, during such periods we would expect to see the following two changes in the
biomarker record relative to the modern period: 1) a decrease in dinosterol δD and 2) an
increase in the C₃₄ botryococcene concentration. This consideration is built into our
method of reconstructing El Niño conditions in Section 3.2.

Additional uncertainties include the fact that factors other than the isotopic
composition of environmental water, including the growth rate of the organism,
temperature, and salinity, have also been shown to influence D/H fractionation in algal
lipids (Schouten et al., 2006; Zhang et al., 2009; Wolhowe et al., 2009; Sachs and
Schwab, 2011). However, due to the extremely low conductivity of El Junco Lake (20
µS/cm; Conlinvaux, 1968) and its tropical location, salinity and temperature are unlikely
to have varied significantly over the Holocene. Growth rate changes are difficult to
constrain in the paleo-environment but could have varied due to changes in nutrient
concentrations and/or light levels (associated with changes in cloud cover). However,
field studies of modern freshwater lake systems indicate that the primary determinant of
lipid δD values is water δD values (Sauer et al., 2001; Huang et al., 2004; Sachse et al.,
2004).
3.2 Inferring mean rainfall and El Niño-related rainfall changes from El Junco Lake

From the available evidence, we infer that sedimentary dinosterol δD reflects the long-term mean isotopic composition of El Junco Lake and that changes in mean lake δD reflect changes in long-term mean rainfall. We further infer that C₃₄ botryococcene δD reflects the mean isotopic composition of El Junco Lake during moderate-to-strong El Niño events and to first order, that changes in the mean lake δD during El Niño events are driven by changes in El Niño-related rainfall. Due to the decadal and longer temporal resolution of the biomarker data, inferred changes in El Niño-related rainfall are interpreted in terms of a change in “El Niño activity”, which we define as a change in the average frequency and/or intensity of El Niño events.

Based on these inferences, we define the following indices to represent past changes in the hydrologic conditions of El Junco Lake.

1. Δ(long-term mean rainfall): Mean Rainfall Index = - Δ(dinosterol δD)$_{\text{std}}$
2. Δ(El Niño-related rainfall): El Niño Rainfall Index = - Δ(botryo δD)$_{\text{std}}$

where the indices are standardized by subtracting the biomarker δD values from the average δD value of the last 100 years and dividing by the standard deviation of the full 9.1 kyr δD record (Fig. 5) and the sign is such that positive (negative) values of the indices indicate wetter (drier) conditions. We chose to standardize the δD values with respect to the 9.1 kyr record in order to be consistent with future reports of the full record. However, the indices are not changed substantially if the standard deviation of the 3 kyr
record is used instead, as the standard deviation of dinosterol δD values is 12‰ for the 9.1 kyr and 11‰ for the 3 kyr record, while that of C₃₄ botryococcene δD values is 17‰ for the 9.1 kyr and 16‰ for the 3 kyr record. For periods in which the abundance of C₃₄ botryococcene was too low for isotopic analysis, the C₃₄ botryococcene concentration data were used instead. This approach is justified by the negative correlation between the log of the C₃₄ botryococcene concentration and the C₃₄ botryococcene δD values (r = -0.62, p < 0.05). The biomarker records are shown in Fig. 6A-C and the rainfall indices are shown in Fig. 7A,B (data available online at PANGAEA; http://www.pangaea.de). A comparison between the El Niño Rainfall Index constructed from C₃₄ botryococcene concentration and δD data versus the El Niño Rainfall Index constructed from δD data alone is shown in Fig. S2.

Positive (negative) values of the Mean Rainfall Index indicate increased (decreased) long-term mean rainfall. Positive (negative) values of the El Niño Rainfall Index indicate increased (decreased) El Niño-related rainfall, which we infer as stronger (weaker) El Niño activity. It is important to note that inferred changes in El Niño activity are based on reconstructions of EEP rainfall, not SSTAs. This distinction is important since it has been suggested that the frequency and amplitude of extreme EEP rainfall anomalies during El Niño events can change without a corresponding change in the amplitude of SSTAs (e.g. due to changes in the mean zonal SST gradient of the tropical Pacific; Cai et al., 2014). In addition, the El Niño Rainfall Index should be more sensitive to changes in Eastern Pacific (EP) El Niño events as compared to Central Pacific (CP) events due to the location of El Junco Lake in the EEP (Ashok et al., 2007) and thus this index should be sensitive to changes in the ratio of EP/CP El Niño events.
We adopt a conservative subset of the El Niño Rainfall Index that only includes periods in which both dinosterol and C$_{34}$ botryococcene data exist, but excludes time periods when a decrease in dinosterol δD is coeval with an increase in C$_{34}$ botryococcene concentration relative to modern (see Section 3.1 for justification). These conditions occur in only two periods in the 3 kyr record, ca. 580–630 yr BP and ca. 2370 yr BP). During these periods oligotrophic conditions driven by a substantially wetter mean climate may have been sufficient to drive B. braunii blooms.

3.3 Inferring ITCZ-related changes from El Junco Lake biomarker records

Due to the sensitivity of the climate in this region to both El Niño events and the position of the ITCZ, we further propose that during periods in which the inferred change in El Niño-related rainfall opposes the change in mean rainfall, changes in ITCZ-related rainfall can be inferred:

\[ \Delta(\text{total rainfall}) = \Delta(\text{El Niño-related rainfall}) + \Delta(\text{residual/ITCZ rainfall}) \]

\[ \text{(4) Residual/ITCZ Rainfall Index} = (\text{Mean Rainfall Index} - \text{El Niño Rainfall Index})_{\text{std}} \]

\[ = [\Delta(\text{botryo } \delta D)_{\text{std}} - \Delta(\text{dinosterol } \delta D)_{\text{std}}]_{\text{std}} \]

This index is a measure of changes in the residual (i.e. non-El Niño-related) rainfall. Because this index compares relative changes in the differentially standardized dinosterol and botryococcene δD values, inferred changes in residual rainfall are robust only for periods in which changes in the Mean Rainfall Index and El Niño Rainfall Index are of opposite sign. Thus, the Residual/ITCZ Rainfall Index is constructed only for these
periods. We further propose that inferred changes in residual rainfall are associated with shifts of the ITCZ in this region since movements of the ITCZ and El Niño events drive the largest rainfall changes (on seasonal-to-interannual time scales) in the modern environment and because the mean position of the ITCZ is known to be sensitive to changes in the climate system that affect the energy budget of the atmosphere (Frierson and Hwang, 2012; Donohoe et al., 2013). Based on this inference, the Residual/ITCZ Rainfall Index will hereafter be referred to as just the ITCZ Rainfall Index. As with the other indices, the ITCZ Rainfall Index is standardized by subtracting the average value of the last 100 years and dividing by the standard deviation of the 9.1 kyr record.

Positive (negative) values of the ITCZ Rainfall Index indicate increased (decreased) non-El Niño rainfall, which we infer as a southward (northward) shift of the ITCZ. E.g. negative values of the El Niño Rainfall Index coincident with positive values of the Mean Rainfall Index imply weaker El Niño activity but increased mean rainfall, from which we infer increased non-El Niño rainfall and a southward shifted ITCZ relative to the modern period. In contrast, positive values of the El Niño Rainfall Index coincident with negative values of the Mean Rainfall Index imply stronger El Niño activity but decreased mean rainfall, from which we infer decreased non-El Niño rainfall and a northward shifted ITCZ. Records of the Mean Rainfall Index, El Niño Rainfall Index, and ITCZ Rainfall Index for the last 3,000 years are shown in Fig. 7 (data available online at PANGAEA; http://www.pangaea.de).

It is important to note that construction of the ITCZ Rainfall Index in this way results in inferred changes in ITCZ-related rainfall that are dependent on opposing changes in El Niño-related rainfall, and thus the El Niño Rainfall Index and ITCZ
Rainfall Index are anti-correlated. The ITCZ Rainfall Index thus highlights periods during which changes in El Niño- and ITCZ-related rainfall were opposing. The inferred opposing changes in El Niño activity and position of the ITCZ appear to account for several of the largest hydrologic changes in El Junco Lake over the last 3,000 years (Fig. 7).

Dynamical theory may support a link between the position of the ITCZ and ENSO variability in this manner. Using intermediate complexity models of the tropical Pacific and the seasonal cycle as an analog for mean state changes, a number of studies have shown that the mean state of the tropical Pacific governed by boreal spring (summer/fall) conditions, when the ITCZ is south (north) of its mean annual position, produces increased (decreased) stability of the coupled ocean-atmosphere system, a condition expected to favor decreased (increased) ENSO variability (Battisti, 1988; Battisti and Hirst, 1989; Thompson, 1998). Such a relationship between the long-term mean position of the ITCZ and ENSO variability should not be confused with their relationship on interannual timescales. For instance, during El Niño events, the ITCZ in the central and eastern equatorial Pacific shifts southward onto the equator in response to warm SSTAs there. In contrast, long-term shifts of the mean annual position of the ITCZ would be expected to be accompanied by substantial changes in the mean state of the tropical Pacific that would drive changes in ENSO variability through changes in the stability of the coupled ocean-atmosphere system.

Interpretation of the $C_{34}$ botryococcene $\delta D$ data are broadly consistent with that presented in Zhang et al. (subm.) with the following two exceptions. One, whereas Zhang et al. (subm.) propose a method of distinguishing between changes in the
amplitude and frequency of El Niño events we choose to adopt a conservative approach of interpreting the C\textsubscript{34} botryococcene data in terms of changes in El Niño “activity” (i.e. changes in mean rainfall associated with El Niño events that arise from changes in either the frequency or amplitude of El Niño events). Two, we limit our interpretation of changes in El Niño-related rainfall to periods in which both dinosterol and C\textsubscript{34} botryococcene data exist, but refrain from ascribing a climate interpretation to those times when a decrease in dinosterol δD coincides with an increase in C\textsubscript{34} botryococcene concentration, in order to eliminate periods in which a substantially wetter mean climate may have been sufficient to drive B. braunii blooms.

Though we believe that our interpretation of the biomarker data is the most straightforward interpretation, we acknowledge that other interpretations are possible. Because the modern-day ecology of El Junco Lake is not well studied, it is possible that the B. braunii and dinoflagellate communities vary in different or more complex ways than what is presented in our hypotheses. Further research on the ecology and limnology of El Junco Lake should aid in the evaluation of these hypotheses. In addition, δD values of lipid biomarkers can be influenced by factors other than the δD values of the environmental water, such as growth rate of the organism. These secondary factors are not easily constrained in the paleo-environment, but their impact on the lipid biomarker δD records cannot be ruled out. Thirdly, we interpret changes in the hydrologic conditions of the lake to result from changes in rainfall, while changes in the stratus cloud cover (garúa), through its impact on evaporation, could also play an important role. Finally, we interpret inferred changes in residual (i.e. non-El Niño-related) rainfall as meridional shifts in the ITCZ because such shifts are observed in the modern climate, and
the position of the ITCZ has been shown to be sensitive to a variety of changes in the climate system (e.g. Frierson and Hwang, 2012; Donohoe et al., 2013). However, other changes in mean climate are possible.

It should be noted that the Mean Rainfall Index, El Niño Rainfall Index, and ITCZ Rainfall Index are qualitative measures of rainfall changes (i.e. increased/decreased rainfall relative to the modern period). Uncertainties in the D/H fractionation factors, various parameters of the hydrologic and isotopic budget of the lake, and construction of the ITCZ Rainfall Index from the difference in biomarker δD values prohibit more quantitative measures of rainfall changes. However, such qualitative reconstructions provide valuable information from this region where few constraints on past changes in ENSO and the ITCZ exist.

3.4 Lake responses to climate change in an idealized model

To facilitate our interpretation of the El Junco Lake biomarker records, we developed an idealized model of the isotope hydrology of El Junco Lake forced with idealized changes in rainfall. Using this model, we simulated the response of lake δD to a wide range of climate conditions by varying the amount of El Niño- and ITCZ-related rainfall. Because a comprehensive sensitivity analysis of the model is outside the scope of this study, we present this model solely as a tool to aid in the illustration of our proxy interpretations outlined in Section 3.2-3.3. Three representative simulations of the lake model are shown in Fig. 8; the details of these simulations can be found in the Supplementary Discussion.
Analysis of the full suite of model simulations with imposed changes in El Niño- and ITCZ-related rainfall indicates that changes in El Niño lake δD are driven by changes in El Niño-related rainfall in the model (Fig. 9A) and a 1:1 relationship is observed between the El Niño Rainfall Index and the standardized changes in El Niño-related rainfall (Fig. 9B; see Supplementary Discussion for details). In addition, changes in mean lake δD are driven by changes in total rainfall in the model (Fig. 9C). Further, if we limit our analysis to periods in which the change in mean lake δD was of opposite sign to the change in El Niño lake δD (as done with the biomarker records for reasons outlined in Section 3.3), the model demonstrates a 1:1 relationship between changes in the standardized ITCZ-related rainfall and the ITCZ Rainfall Index (Fig. 9D; see Supplementary Discussion for details). Interestingly, while the presence of strong non-linearities cannot be ruled out in the real world, simulations with the idealized lake model suggest that the relationship between rainfall changes and lake δD may be fairly linear over a wide range of climate perturbations - even in the presence of overflowing conditions. This is likely attributable to the linearity of the relationship between rainfall rate and δD rain. These simulations with the idealized model of El Junco Lake demonstrate how we use lake δD reconstructions to infer past changes in mean, El Niño- and ITCZ-related rainfall.

3.5 Modern biomarker records

In order to assess if the sedimentary biomarkers accurately reflect the recent climate in the Galápagos, we compared the rainfall indices from the top 20 cm of sediment with local rainfall and ENSO records for the period during which
meteorological measurements are available (Fig. 10). Twenty-year running averages of rainfall and variance of Niño 3.4 SSTAs were taken to coincide with the average time slice represented by our sediment samples. While the low sampling resolution in the biomarker data, large error estimates relative to trends in the data, and short observational record prohibit a robust test of the proxy data, the behavior of the indices is broadly consistent with our interpretation. In particular, the correspondence between (1) the Mean Rainfall Index and average total rainfall (Fig. 10A), (2) the El Niño Rainfall Index, average El Niño-related rainfall (rainfall that fell during moderate-to-strong El Niño events) and the average variance of Niño 3.4 SSTAs (Fig. 10B), and (3) the ITCZ Rainfall Index and average ITCZ-related rainfall (taken as the difference between total rainfall and El Niño-related rainfall; Fig. 10C) is broadly consistent with our interpretation. The small variations in the modern lake δD and climate indices relative to those of the full 3 kyr record imply that large changes in ENSO and the position of the ITCZ may have occurred in the late Holocene in comparison to those that occurred in the 20th century.

3.6 Changing hydrologic conditions in El Junco Lake from keto-ols

As an independent assessment of past hydrologic changes of El Junco Lake, a record of mean lake level changes was developed based on the ratio of sedimentary keto-ol isomers derived from algae and ferns. Long chain keto-ols are abundant in El Junco Lake sediment and present additional indicators of paleo-environmental change due to their source-specificity (Atwood et al., 2014). C30 ω16-keto-1-ol (structure III in Fig. S1) is produced by algae of the class Eustigmatophyceae (Méjanelle et al., 2003; Volkman et
al., 1992; Volkman et al., 1999), while \( C_{30} \omega 20 \)-keto-1-ol (structure V in Fig. S1) is produced by Cyathea weatherbyana, an abundant tree fern around El Junco Lake (Atwood et al., 2014).

Because \( C_{30} 1,\omega 20 \)-diols and keto-ols are produced by terrestrial and shoreline ferns, while \( C_{30} 1,\omega 16 \)-diols and keto-ols are produced by aquatic algae, it is expected that as the lake level increases (decreases), the input of \( C_{30} \omega 20 \)-keto-1-ol relative to \( C_{30} \omega 16 \)-keto-1-ol would decrease (increase) due to both an increase (decrease) in the ratio of surface area to circumference of the lake and to an increase (decrease) in the ratio of lake area to catchment area (see Supplementary Discussion for more details).

In support of our interpretation of dinosterol \( \delta D \) as a proxy of mean rainfall changes, the ratio of \( C_{30} \omega 16 \)-keto-1-ol/\( \omega 20 \)-keto-1-ol is anti-correlated to the dinosterol \( \delta D \) values over the last 3 kyr (\( r = -0.54, \ p < 0.05; \) Fig. 6C,D). Of note, however, are the large differences in dinosterol \( \delta D \) and keto-ol records in the near-surface sediment. In particular, the \( \omega 16 \)-keto-1-ol/\( \omega 20 \)-keto-1-ol ratio decreases to near zero in the upper 5 cm of sediment while the dinosterol \( \delta D \) values are constant or decrease slightly. We hypothesize that these differences are due to a lower degree of diagenetic transformation of diols to keto-ols in the surface sediment relative to deeper sediment (see Supplementary Discussion).

Although we place more weight on the dinosterol \( \delta D \) record than on the keto-ol record as a record of mean rainfall changes (see Supplementary Discussion), the ratio of \( C_{30} \omega 16 \)-keto-1-ol/\( \omega 20 \)-keto-1-ol provides an independent record of past lake hydrologic conditions that supports many of the prominent trends in the dinosterol \( \delta D \) record and thus strengthens the paleoclimate interpretations of the biomarker records.
3.7 Comparison with other El Junco Lake paleohydrologic records

Although we defer an in-depth analysis of the ITCZ and El Niño reconstructions presented here to a subsequent paper, we include a basic comparison between our rainfall reconstructions and those based on El Junco Lake sediment grain size from Conroy et al. (2008). Variations in the volume ratio of silt to clay (where silt and clay are defined based on particle diameters of 3.9–62.5 µm and < 3.9 µm, respectively) were interpreted by Conroy et al. (2008) as a proxy for mean rainfall. We thus compare the Mean Rainfall Index from this study to the ratio of silt to clay in Fig. 7A,B. A visual comparison suggests general coherency in several of the major features present in the two records from ca. 1700 years BP–present. For instance, the maxima in the Mean Rainfall Index ca. 1400–1250 years BP (550–700 AD) and 650–600 years BP (1300–1350 AD), indicative of wet mean conditions, correspond to periods when the silt/clay ratio was also high. As the inferred El Niño-related rainfall was reduced ca. 1250–1350 years BP (700–600 AD), the ITCZ Rainfall Index suggests that a southward shift of the ITCZ may have been responsible for the wet conditions during this time. The period ca. 650–600 years BP (1300–1350 AD) corresponds to the wettest mean climate inferred from the Mean Rainfall Index. Coordinated anomalies in the dinosterol and botryococcene δD values indicate that the wet conditions during this time were recorded by both biomarkers, suggesting that either a southward shifted ITCZ and/or stronger El Niño activity could have been responsible for the wetter mean conditions. The Mean Rainfall Index and the silt/clay ratio also show agreement ca. 1200–1000 years BP (750–950 AD), when the driest mean conditions inferred from the Mean Rainfall Index correspond to a low ratio of
Comparison with the ITCZ Rainfall Index suggests that a northward shifted ITCZ ca. 1100–1000 years BP (850–950 AD) may have contributed to the dry climate during this time. Poorer agreement between the two records is observed prior to ca. 1700 years BP (250 AD).

Variations in percent sand were interpreted by Conroy et al. (2008) as a proxy for changes in rainfall intensity and therefore changes in the frequency and/or magnitude of El Niño events. A comparison between the Conroy et al. (2008) sand record and the El Niño Rainfall Index from this study suggests several coherent features, including the absolute minimum in both records (indicating weak El Niño activity) ca. 1400–1200 years BP (550–750 AD). Leading up to this minimum, inferred El Niño activity declined from ca. 1900–1400 years BP (50–550 AD) in both records. In addition, following the period of lowest inferred El Niño activity, both records suggest increased activity beginning ca. 1200 years BP (750 AD) and culminating ca. 700 years BP (1250 AD).

However, while the El Niño Rainfall Index from this study suggests that El Niño-related rainfall remained higher from ca. 700–100 years BP (1250–1850 AD), the sand record suggests that the intensity of rainfall was similar to, or lower than, modern conditions between ca. 600–100 years BP (1350–1850 AD). Another notable dissimilarity between the records is the maximum in sand abundance ca. 2000–1600 years BP (50 BC–350 AD), which is not mirrored in the El Niño Rainfall Index.

Any lack of co-variation between our biomarker-derived climate indices and the sediment grain size records from Conroy et al. (2008) may result in part from complexities in the relationship between the biomarker records and rainfall on the one hand (as discussed in Section 3.3), and complexities in the relationship between sediment
grain size and rainfall on the other. The grain size of sediment in El Junco Lake will be
influenced by weathering, which is a complex function of rainfall, temperature,
vegetation type and cover, tectonics, among other things. Therefore, rainfall is just one
of several factors that will influence the grain size parameters, and any relationship
between rainfall and grain size could be non-stationary in time. Furthermore, the grain
size of sediment is likely to be insensitive to the source of the rain, and unable to
distinguish between ITCZ-related rainfall and El Niño-related rainfall, both of which
impact rainfall intensity. Nevertheless, climate changes that are inferred from both the
organic geochemical and grain size records should be more robust than interpretations
from either data set alone.

4. Conclusions

Past changes in long-term mean rainfall and rainfall associated with El Niño
events were reconstructed from the Galápagos Islands over the last 3 kyr using the
concentration and hydrogen isotope composition of four lipid biomarkers in the sediment
of El Junco Lake, San Cristóbal Island. Sedimentary dinosterol δD is interpreted as a
proxy for changes in long-term mean rainfall, while C_{34} botryococcene δD is interpreted
as a proxy for changes in mean rainfall associated with moderate-to-strong El Niño
events. A third hydrologic proxy was developed from the sedimentary ratio of C_{30} \omega 16-
keto-1-ol (produced by aquatic algae) to C_{30} \omega 20-keto-1-ol (produced by terrestrial and
shoreline ferns) that provides a record of past changes in mean lake level. Its co-variation
with the dinosterol δD record bolsters the utility of the latter as a record of mean rainfall
changes. These reconstructions are further used to infer periods when changes in ITCZ-related rainfall opposed changes in El Niño-related rainfall. Opposing changes in El Niño- and ITCZ-related rainfall appear to have characterized several of the largest changes in El Junco Lake hydrology over the last 3,000 years. Indices of past changes in mean, El Niño- and ITCZ-related rainfall were developed and illustrated with an idealized isotope hydrology model of El Junco Lake.

A unique method was presented that demonstrates the potential to reconstruct past changes in rainfall associated with disparate climate phenomena, here the ITCZ and ENSO, from the abundance and hydrogen isotope composition of sedimentary lipids in a single lake. Though the set of biomarkers used in this study may be specific to this environment, the general method can be broadly applied to any lacustrine system that experiences hydrological variability driven by multiple climate and/or environmental conditions and that hosts phytoplankton that respond to those conditions.

A comparison between the biomarker-based rainfall reconstructions presented in this study and rainfall reconstructions based on sedimentary grain size from Conroy et al. (2008) suggests that many of the prominent features in the biomarker-based reconstructions have counterparts in the grain size records, enabling more robust interpretation of rainfall changes in the Galápagos Islands over the past 3 kyr. That these records come from the core of the eastern equatorial Pacific where the fundamental dynamics of ENSO and the ITCZ operate and where few such records exist, represents an important contribution toward furthering our understanding of past climate change in the tropics.
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Data are available online at PANGAEA (http://www.pangaea.de).

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Figure Captions

Fig. 1. A) Map of the Galápagos Islands with location of El Junco Lake represented by the red dot; B) Photo of El Junco Lake taken in Aug. 2011. (Photo credit: Margaret Johnson)

Fig. 2. Seasonal cycle of the ITCZ in the tropical Pacific. Climatological rainfall from 1980–2010 AD averaged over the peak of the A) dry season (Aug-Oct) and B) wet season (Feb-April). The star indicates the location of San Cristóbal Island, Galápagos. Rainfall data is from GPCP Version 2.2 Combined Precipitation Data Set (http://www.esrl.noaa.gov).

Fig. 3. Galápagos annually-averaged rainfall (averaged from Aug–July and plotted on a natural log scale) versus positive Niño 3.4 SSTAs averaged over Dec-Feb. The 95% confidence interval for the true correlation coefficient (\( r \)) is 0.44 < \( r \) < 0.88 for a sample size of N = 22. No significant correlation was observed between annual rainfall and negative SSTAs (\( r = 0.30, N = 23 \)). Rainfall data is from the Puerto Ayora weather station on Santa Cruz Island (http://www.darwinfoundation.org) and spans the period from 1964–2011 AD.

Fig. 4. A) Galápagos lowland rainfall (solid; from the Puerto Ayora weather station at 2 m a.s.l.) and highland rainfall (dashed; from the Bellavista weather station at 194 m a.s.l.) from 1964–2011 AD in comparison with El Niño and La Niña events and B) satellite-
based rainfall data averaged over the eastern Pacific along the equator (1 °S to 1 °N, 110 °W to 85°W). Blue shaded bars at the top of the plot indicate La Niña events, while red shaded bars indicate El Niño events. Moderate to strong El Niño events are indicated by full-length bars. El Niño and La Niña events are defined by 3-month running mean Niño 3.4 SSTAs where El Niño (La Niña) events are defined as SSTAs ≥ 0.5 °C (≤ -0.5 °C) and moderate-to-strong El Niño events are defined as SSTAs ≥ 1.0 °C for three or more consecutive months. SST data was obtained from NOAA ERSST v3b. Rainfall data from Puerto Ayora and Bellavista weather stations on Santa Cruz Island was obtained from the Charles Darwin Research Station (http://www.darwinfoundation.org). Satellite rainfall data was obtained from GPCP Version 2.2 Combined Precipitation Data Set (http://www.esrl.noaa.gov). Designations of EP and CP El Niño events are from Kidwell et al. (2014).

**Fig. 5.** C$_{34}$ botryococcene and dinosterol concentration versus δD values for the full 9.1 kyr record (with concentration plotted on a log scale). The 95% confidence interval for the true correlation coefficient (ρ) is 0.47 < ρ < 0.74; N = 85 for the C$_{34}$ botryococcene data and 0.24 < ρ < 0.71; N = 41 for the dinosterol data.

**Fig. 6.** Lipid biomarker concentration and δD records from El Junco Lake. A) Dinosterol and C$_{34}$ botryococcene concentration; B) C$_{34}$ botryococcene δD; C) dinosterol δD; D) C$_{30}$ ω16-keto-1-ol/ω20-keto-1-ol ratio. The y-error bars on the C$_{34}$ botryococcene and dinosterol δD values represent ±1σ of the triplicate δD measurements.
Fig. 7. El Junco rainfall indices plotted with smoothed (70-year running mean) El Junco Lake grain size records from Conroy et al. (2008). A) Mean Rainfall Index plotted with the silt/clay ratio from Conroy et al. (2008); B) El Niño Rainfall Index (where the solid line connects all data points and the symbols with error bars are the conservative subset of the El Niño Rainfall Index (values for which both dinosterol and C\textsubscript{34} botryococcene data exist and a decrease in dinosterol δD is not coeval with an increase in C\textsubscript{34} botryococcene concentration- see Section 3.2 for details) plotted with percent sand from Conroy et al. (2008); C) ITCZ Rainfall Index (defined only for periods in which the Mean Rainfall Index and El Niño Rainfall Index are of opposite sign- see Section 3.3 for details). The y-error bars on the Mean Rainfall Index and the El Nino Rainfall Index are derived from the 1σ error of the dinosterol and C\textsubscript{34} botryococcene δD measurements, respectively. The y-error bars on the ITCZ Rainfall Index are calculated by propagating the 1σ error from the C\textsubscript{34} botryococcene and dinosterol δD data using Eqns (1)–(4).

Fig. 8. Response of the mean δD\textsubscript{lake} and El Niño δD\textsubscript{lake} (i.e. the mean lake δD during El Niño events) to prescribed climate changes in the idealized model of El Junco Lake. A) Modern simulation of El Junco Lake; B) simulation in which the El Niño-related rainfall was decreased and the ITCZ-related rainfall was increased- conditions that result in a negative El Niño Rainfall Index and a positive ITCZ Rainfall Index; C) simulation in which the El Niño-related rainfall was increased and the ITCZ-related rainfall was decreased- conditions that result in a positive El Niño Rainfall Index and a negative ITCZ Rainfall Index (see Supplementary Discussion for more details).
**Fig. 9.** Rainfall changes versus lake δD and climate indices as simulated by an idealized model of El Junco Lake (where changes are defined relative to the modern simulation). A) The change in El Niño-related rainfall versus the change in El Niño δD\textsubscript{lake} (i.e. the mean lake δD during El Niño events); B) the standardized change in El Niño-related rainfall versus the El Niño Rainfall Index; C) the change in total (ITCZ- plus El Niño-related) rainfall versus the change in mean δD\textsubscript{lake}; D) the standardized change in ITCZ-related rainfall versus the ITCZ Rainfall Index. Each data point represents output from a simulation with uniquely prescribed El Niño- and ITCZ- rainfall rates.

**Fig. 10.** El Junco rainfall indices plotted with instrumental rainfall and SSTA data. A) Mean Rainfall Index and 20-year running average of total rainfall; B) 20-year running average of the variance of Niño 3.4 SSTAs; C) El Niño Rainfall Index and 20-year running average of El Niño-related rainfall (taken as rainfall that fell during moderate to strong El Niño events); D) ITCZ Rainfall Index and 20-year running average of ITCZ-related rainfall (taken as the difference between mean rainfall and El Niño-related rainfall). Rainfall data was taken from the Puerto Ayora weather station on Santa Cruz Island (http://www.darwinfoundation.org), SST data was taken from iCOADS (http://icoads.noaa.gov/) and HadISST (http://www.metoffice.gov.uk/hadobs/hadisst/).
Highlights

- Novel proxies for tracking hydrologic changes of El Junco Lake, Galápagos
- Based on molecular and isotopic biomarkers from several types of plants and algae
- Proxies used to infer past changes in mean rainfall and El Niño-related rainfall over last 3 kyr
- Novel method used to infer changes in ITCZ-related rainfall during select periods
Figure

Click here to download Figure: Fig2.eps
Figure

Click here to download Figure: Fig3.eps

\[
\begin{align*}
\text{Annual rainfall (mm/day)} \\
\text{Niño 3.4 SSTA (°C)}
\end{align*}
\]

\[r = 0.73\]

\[0.44 < \rho < 0.88\]
Figure
Click here to download Figure: Fig4.eps
Figure

Click here to download Figure: Fig5.eps

- $C_{34}$ botryococcene
  - $y = -0.053x - 10.873$
  - $r = -0.62$

- dinosterol
  - $y = 0.022x + 6.584$
  - $r = 0.51$
Figure

Click here to download Figure: Fig7.eps

A) higher mean rainfall
   lower mean rainfall

B) stronger El Niño activity
   weaker El Niño activity

C) ITCZ south
   ITCZ north
$\Delta$ (El Nino $\delta D_{lake}) = +$

$\Delta$ (Mean $\delta D_{lake}) = -$
Figure
Click here to download Figure: Fig9.eps