**Reconstructing precipitation in the tropical South Pacific from dinosterol 2H/1H ratios in lake sediment**

Ashley E Maloney1\*, Daniel B Nelson2, Julie N Richey3, Matthew Prebble4, David A Sear5, Jonathan D Hassall5, Peter G Langdon5, Ian W Croudace6, Atun Zawadzki7,Julian P Sachs1

1University of Washington, School of Oceanography, College of the Environment, Box 355351, Seattle, WA 98195, USA

2Department of Environmental Sciences – Botany, University of Basel, Basel, Switzerland

3U.S. Geological Survey, St. Petersburg, Florida 33701, USA

4Australian National University, Department of Archaeology and Natural History, School of Culture, History and Languages, Collage of Asia and the Pacific, Canberra ACT 0200, Australia

5University of Southampton, Geography and Environment, Highfield, Southampton SO17 1BJ, UK

6University of Southampton,School of Ocean and Earth Science, National Oceanography Center, Southampton SO14 3ZH UK

7Australian Nuclear Science and Technology Organization, New Illawarra Rd, Lucas Heights, NSW 2234, Australia

\*Corresponding author. E-mail address: amaloney@princeton.edu (A.E. Maloney)

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**Highlights:**

– δ2Hdinosterol in 21 lakes on 12 islands across the tropical South Pacific is significantly correlated with both δ2Hlakewater and mean annual precipitation amount.

– The δ2Hdinosterol vs. precipitation regression provides a means for quantitative reconstructions of past precipitation in the South Pacific Convergence Zone with an uncertainty of less than 3 mm d-1.

**ABSTRACT**

The South Pacific Convergence Zone (SPCZ) is the largest precipitation feature in the Southern Hemisphere supplying freshwater to 11 million people. Despite its significance, little is known about the location and intensity of SPCZ precipitation prior to instrumental records, hindering attempts to predict precipitation changes in a warming world. Here we use sedimentary molecular fossils to establish a tool for extending the historical record of precipitation. Freshwater lake sediments and water samples were collected from 30 lakes that span a 4.6 mm d−1 range in precipitation rates from the Global Precipitation Climatology Project (GPCP). δ2Hlakewater values from 29 lakes ranged from –29 to +23‰ and were inversely correlated (r= −0.51, R2=0.26, p<0.001) with precipitation rates, likely due to the combination of the amount of precipitation plus evaporation. δ2H values of the dinoflagellate sterol dinosterol in surficial sediments from 21 lakes ranged from −316‰ in the Solomon Islands to −245‰ in French Polynesia. These δ2Hdinosterol values were significantly correlated (r= −0.71, R2=0.50, p<0.001) with δ2Hlakewater and inversely correlated (r= −0.77, R2 =0.59, p<0.001) with mean annual precipitation rates with a sensitivity of −12.1±2.6‰ (mm d−1)−1. Fractionation between dinosterol and lake water (εdinosterol/lakewater) increased at the wettest lake sites (R2=0.48, p<0.001). The empirical relationship between δ2Hdinosterol and GPCP rainfall, although indirect, provides a means of quantitatively reconstructing past precipitation in the SPCZ region with an uncertainty of less than 3.1 mm d−1, which compares favorably to the 1.5 mm d−1 uncertainty for the satellite-gauge based GPCP precipitation data.

**1. INTRODUCTION**

The South Pacific Convergence Zone (SPCZ) is the Southern Hemisphere’s largest precipitation feature (Vincent, 1994) supplying freshwater to 11 million people on 3,975 islands (Power et al., 2011; WHO, 2016). The majority of coupled climate models suggest a larger and wetter SPCZ in the future with a more extreme zonal structure during ENSO events (Brown et al., 2011; Cai et al., 2012; Brown et al., 2012; Borlace et al., 2014). It is difficult to assess these model projections because little is known about the natural variability of precipitation in the SPCZ region owing to a short instrumental record of just 40 years from satellites and no more than 130 years from gauges. Efforts to extend the instrumental record of SPCZ precipitation with paleoclimate proxy data are needed to validate model hindcasts.

Speleothems from Vanuatu (Partin et al., 2013) and the Solomon Islands (Maupin et al., 2014) in conjunction with coral archives from Fiji and Rarotonga (Linsley et al., 2004; Linsley et al., 2006), Vanuatu (Quinn et al., 1993), New Caledonia (Quinn et al., 1998), and the nearby Great Barrier Reef (Druffel and Griffin, 1993; Hendy et al., 2002) and Coral Sea (Calvo et al., 2007) provide insights into pre-instrumental-era climate changes, but do not extend prior to 600 years ago in this region. Sediment archives have successfully recorded longer-term changes in tropical vegetation and human activity (Southern, 1986; Parkes, 1994; Hope and Pask, 1998; Stevenson and Hope, 2005; Wirrmann et al., 2006; Prebble and Wilmshurst, 2009; Prebble et al., 2013; Combettes et al., 2015; Rull et al., 2015), and large scale precipitation during the past 10,000 years (Hassall, 2017). However, additional proxy development and calibration work is required to develop quantitative rainfall records that can be compared to climate model output.

In the maritime tropics, the hydrogen isotope ratio (2H/1H, expressed as δ2H(= [Rsample/RVSMOW]− 1, where R is 2H/1H and VSMOW is Vienna Standard Mean Ocean Water) of precipitation is principally controlled by the precipitation rate when averaged over monthly and longer time scales (Dansgaard, 1964; Rozanski et al., 1993; Risi et al., 2008; Kurita et al., 2009). Tropical island lake water δ2H values reflect this signal, plus the added effects of evaporative enrichment (Kebede et al., 2009; Garcin et al., 2012; Issa et al., 2015). Lipids from microalgae have δ2H values that track the isotopic composition of their environmental water with R2 values typically > 0.9 (Paul, 2002; Englebrecht and Sachs, 2005; Schouten et al., 2006; Zhang and Sachs, 2007; Sachse et al., 2012), and their preservation in sediments offers a window into past lake hydrology, and by extension past precipitation. Despite this, limited region-specific calibration work has been conducted to understand and quantify the nature of the climate signal contained in the δ2H values of sedimentary lipids.

Phytoplankton produce a variety of taxon-specific lipids (Volkman et al., 1998; Volkman, 2005) that can be exploited to infer environmental conditions. Dinosterol (4α,23,24-trimethyl-5α-cholest-22E-en-3β-ol) is produced almost exclusively by dinoflagellates, although it has also been detected in a single marine diatom species (Volkman et al., 1993). The relative source-specificity of dinosterol offers an advantage over more generic lipids such as fatty acids and has been used to reconstruct pre-instrumental hydrology across the tropical Pacific (Sachs et al., 2009; Smittenberg et al., 2011; Atwood and Sachs, 2014; Nelson and Sachs, 2016; Richey and Sachs, 2016).

In addition to source water isotopic composition, algal lipid δ2H values are sensitive to environmental parameters including salinity (Schouten et al., 2006; Sachse and Sachs, 2008; Sachs and Schwab, 2011; Nelson and Sachs, 2014b; Maloney et al., 2016; Sachs et al., 2016; Weiss et al., 2017), growth rate and phase (Z. Zhang et al., 2009; Wolhowe et al., 2009; Chivall et al., 2014; Sachs and Kawka, 2015; Wolhowe et al., 2015), temperature (Z. Zhang et al., 2009; Wolhowe et al., 2009; Ladd et al., 2017), light (van der Meer et al., 2015; Sachs et al., 2017), and redox environment or metabolism (Schwab et al., 2015a). This highlights the need for careful lipid and site selection in attempts to use sedimentary biomarkers to reconstruct past environmental conditions.

Previous surficial lake sediment studies have demonstrated that δ2H values of sedimentary molecular fossils from a variety of terrestrial and aquatic photoautotrophs largely reflect modern spatial climate gradients. Sampling regions include global (Nelson and Sachs, 2014b) and continental transects across Europe (Sachse et al., 2004), the Americas (Polissar and Freeman, 2010), North America (Sauer et al., 2001; Huang et al., 2004; Nelson and Sachs, 2014a), southwest United States (Hou et al., 2008), Cameroon (Garcin et al., 2012; Schwab et al., 2015a; Schwab et al., 2015b), and the Tibetan Plateau (Mügler et al., 2008; Xia et al., 2008; Aichner et al., 2010). However, the fidelity with which lacustrine photoautotrophic lipid δ2H values track climate in lakes across the vast expanse of the tropical South Pacific Ocean has not yet been tested, nor has a calibration for rainfall been established that can be used to quantitatively reconstruct past precipitation rates.

The present study was therefore undertaken to develop and validate a quantitative rainfall proxy based on the δ2H value of dinosterol from recent lake sediments across the tropical South Pacific Ocean. Although variable secondary influences on 2H/1H fractionation have been used to enhance the level of paleoclimate information recovered from sedimentary biomarkers (Nelson and Sachs, 2016), in the present study we sought to limit potential impacts of salinity or temperature by targeting only freshwater lakes spanning a small range of seasonally stable temperatures (<4°C, **Table A.1**, Appendix A). To compensate for less controllable parameters such as species composition, growth rate, metabolism, and light which may influence fractionation, as well changes in moisture source, temperature, and secondary evaporation which may influence lake water, multiple lakes were sampled in each region. The results demonstrate that δ2Hdinosterol values in lake sediments can be used to reconstruct mean annual precipitation rates in the tropical South Pacific.

**2. METHODS**

**2.1. Sample Collection**

Sediment core top samples were collected from 21 lakes (**Fig. 1**; **Table B.1**, Appendix B). Most core top samples came from 6.6 cm diameter sediment cores collected in 2011–2012 with a universal corer device (Aquatic Research, Hope, ID), but the Lake Wanum sample was collected in 1999. The uppermost portion (typically 20–50 cm) was sectioned in the field at 0.5 or 1 cm intervals to a depth at which the sediment was sufficiently consolidated to transport and store without disturbance to stratigraphy. Lake Emaotul was sampled in 1995 during a dry interval with a D-section peat borer and again in 2015 with an 8.6 cm diameter UWITEC (UWITEC, Mondsee, Austria) gravity corer. Samoa (Lake Lanoto’o) and New Caledonia (2 lakes) were sampled in 2013 and Samoa (Lake Lanoto’o) again in 2014 with the UWITEC gravity corer. Vanuatu, New Caledonia, and Samoa 2013 samples were subsampled in the field at 0.5 or 1 cm intervals, whereas Samoa samples obtained in 2014 were subsampled in the laboratory; interface integrity was maintained using Zorbitrol (sodium polyacrylate polymer gel, Ulster Scientific Incorporated, New Paltz, NY) in the field. Samples from Tetiaroa were collected in March and/or October 2015 by hand. Sediment cores from the deepest (88 m) part of Lac Lalolalo (Wallis) had disturbed sediment water interfaces (15–20 cm) due to vigorous degassing upon recovery, therefore additional cores were collected from shallower depths (24 m) to obtain surface sediments that did not experience mixing.

Water samples were collected at all lakes, with the exception of Lake Wanum and the 1995 visit to Emaotul lake. Most samples were from the lake surface, some lakes also had deeper samples (**Table B.2**, Appendix B). Samples and stored in glass or plastic screw-cap vials and sealed with electrical tape to prevent evaporation. Water samples were collected from nine additional lakes in Nauru, Vanuatu, Wallis, and the Solomon Islands from which no sediment samples were obtained. Several non-lake water samples were collected for hydrogen and oxygen isotope analysis from streams, channels, lagoons, ocean, tap water, rain barrels, and cave drip water (**Table B.2**, Appendix B). Salinity was measured in the field with a handheld refractometer which was calibrated against distilled water with a precision of 1 ppt. Local precipitation events were sampled whenever possible with a rain water collector containing mineral oil to prevent evaporation.

Lake water temperature, pH, conductivity, and dissolved oxygen (DO) were measured on-site using a portable data sonde (Hydrolab, Loveland, CO) at most lakes (**Fig. A.1**, Appendix A). Lake areas were determined with the Google Earth Pro polygon tool or from literature if available. Lake elevation was determined by topographical maps, GPS, and literature if available (**Table A.1**, Appendix A).

**2.2. Water isotope analyses**

Water δ2Hand δ18O values were measured at the University of Washington, School of Oceanography 6–14 times per sample (with the first three measurements discarded to avoid memory effects) on a Picarro L2130-i Isotopic Liquid Water Analyzer (Picarro, Inc., Santa Clara, CA) in high precision mode and normalized to VSMOW using three in-house lab standards with hydrogen isotopic compositions of −107.3 ± 0.5‰, −76.1 ± 0.3‰, and 14.8 ± 1.0‰. A subset of 32 samples were reanalyzed on 2 to 4 subsequent occasions between 2014 and 2017 resulting in a long-term analytical uncertainty of 1.3‰ for δ2H and 0.3‰ for δ18O.

Water samples collected from Wallis Island were measured at University of Hawaii on a Picarro L1102-*i* WS-CRDS with analytical precision of 0.5‰ for δ2H and 0.04‰ for δ18O determined using an in-house standard run after every 10th sample. Water samples collected from New Caledonia and Samoa Islands were measured at the Natural Environmental Research Council Isotope Geosciences Facility in Keyworth, UK with a GV Isoprime mass spectrometer connected to Multiprep and EuroPyrOH sample preparation and inlet devices and had an analytical precision of <0.05 for δ2H and <0.05 for δ18O. All samples from a single lake were averaged for a lake-average value (**Table 1**).

**2.3. Dinosterol hydrogen isotope analyses**

Lipid extracts were obtained from freeze-dried sediment samples with an Accelerated Solvent Extractor (ASE-200, Dionex Corp., Sunnyvale, CA, USA) using 9:1 dichloromethane:methanol (DCM:MeOH) at 1500 psi and 100 °C for three 5 min cycles. Samples were base-hydrolyzed with 1N potassium hydroxide (KOH) at 70 °C for 6–12 h followed by liquid-liquid extractions in hexane (HEX). Column chromatography was used to separate dinosterol from other major lipid classes. A solid phase of 0.5 g aminopropyl-silica gel (Supelco) and 8 mL of 3:1 DCM:isopropyl alcohol eluted the neutral fraction containing dinosterol. This fraction was further purified using 1.0 g deactivated silica gel with 6 mL of HEX, followed by 6 mL 1:1 HEX:DCM, and finally 8 mL 4:1 HEX:Ethyl acetate to elute an alcohol fraction containing dinosterol.

Dinosterol was purified from neighboring sterols by High Performance Liquid Chromatography (HPLC) according to procedures described in Nelson and Sachs (2013). Prior to HPLC, samples were acetylated using acetic anhydride with a known hydrogen isotopic composition (−123.8 ± 8.2‰) by dissolving in 40 μL of 1:1 pyridine:acetic anhydride and heating at 70°C for 0.5 h. Lipid identifications were performed on a Gas Chromatography Mass Spectrometer (GCMS) using an Agilent GC 6890N connected to an Agilent quadrupole MSD5975 detector and equipped with an Agilent 7683 autosampler, a split-splitless injector operated in splitless mode, and an Agilent VF-17ms capillary column (60 m × 0.25 mm × 0.25 μm) used with helium carrier gas at 1.5 mL min−1. The oven temperature was ramped from 110 °C to 280 °C at 15 °C min−1 and held for 40 min, then ramped to 320 °C at 3 °C min−1 and held for 10 min. HPLC-purified dinosterol samples were quantified against a 5α-cholestane internal standard on an Agilent 6890N GC equipped with a Flame Ionization Detector (FID), using methods similar to the GCMS.

δ2Hdinosterol values were measured via gas-chromatography isotope-ratio mass spectrometry (GC-IRMS). Samples were injected into a Thermo Trace Ultra GC II (Thermo Fisher Scientific, Waltham, MA, USA) gas chromatograph via a Thermo TRIPLUS autosampler operated in splitless mode at 330 °C with a constant flow (1.1 mL min−1) of He carrier gas. Eluting compounds were pyrolyzed in a 1400 °C ceramic reactor for introduction on a Thermo DELTA V PLUS IRMS. The GC was equipped with a VF-17ms capillary column (60 m × 0.25 mm × 0.25 μm), the oven was programmed to hold at 120 °C for 2 min, increase to 260 °C at 20 °C min−1, increase to 310 °C at 1 °C min−1, increase to 325 °C at 20 °C min−1, and hold 325 °C for 15 min. The H3+ factor (Sessions et al., 2001) was measured prior to every sequence and was 2.00 ± 0.27 ppm nA−1 during the 38 months that 27 sequences were run for this study. External standards of known hydrogen isotopic composition including *n-*C21-alkane (−214.7‰ CAS #629-94-7), *n-*C23-alkane (−48.8‰ CAS #638-67-5), *n-*C26-alkane (−54.9‰ CAS #630-01-3), *n-*C32-alkane (−212.4‰ CAS #544-85-4), *n-*C34-alkane (−231.8‰ CAS #14167-59-0), and *n-*C38-alkane (−102.6‰ CAS #7194-85-6) (Dr. Arndt Schimmelmann, Indiana University, http://mypage.iu.edu/~aschimme/compounds.html) were injected throughout each run. Any peak areas less than 11 Vs were disregarded to avoid size dependent δ2H effects (Polissar and D’Andrea, 2014) and isotopic compositions were evaluated in the Isodat 2.0 software relative to calibrated H2 reference gas. Dinosterol δ2H values were corrected using the regression of known versus Isodat-reported *n*-alkane standard δ2H values. Dinosterol δ2H values were then corrected for hydrogen added during acetylation by a mass balance calculation as in Nelson and Sachs (2014a).

All 44 core top sample dinosterol δ2H values and the standard deviations of multiple injections are reported in **Table B.1**, Appendix B. The pooled uncertainty for replicate analyses was 6.2‰ (Polissar and D’Andrea, 2014) and represents the overall analytical uncertainty. Included in this table is a cyanobacterial mat sample growing atop floating peat sedge on lake Tagimaucia (Taveuni, Fiji) that was not included in the lake-averaged sediment core top value. For lakes with multiple core top samples, lake-averaged δ2Hdinosterol values were determined by averaging all core top samples and the standard deviation of multiple lake core tops represents lake-averaged δ2Hdinosterol uncertainty (**Table 1**). A pooled uncertainty of 10.4‰ was determined from lakes with multiple core tops and applied to lakes with only a single core top sample and used for all graphs and statistics.

**2.4. Statistics, data, and models**

Relatively large uncertainties on independent variables in this study required the use of a maximum likelihood estimate method (York et al., 2004) incorporating bivariate analytical uncertainty for all linear regressions (York, 1969; Reed, 1989; York et al., 2004; Cantrell, 2008; Thirumalai et al., 2011), which were performed using published MatlabTM code (Thirumalai et al., 2011). Since most previous studies of algal lipid isotopes with respect to environmental variables did not have large x-errors (culture studies) or the x-errors were not accounted for (field studies), the results of ordinary least squares regressions are also provided in the figure captions. In all cases the biased ordinary least squares regressions slopes are smaller than the maximum likelihood method slopes, as was the case in a rigorous study of calibration techniques for corals as sea surface temperature proxies (Xu et al., 2015).

Monte Carlo error propagation, t-test statistics, mapping, data extraction, and imaging was done in R (v3.4.0) (R Core Team, 2017) with RStudio (v1.0.143, www.rstudio.com). Smoothed point data extraction from netcdf files was done in R using a bilinear method to average the four grid cells nearest to the lake sites.

The Global Precipitation Climatology Project (GPCPv2.3) dataset (Adler et al., 2003; Adler et al., 2017) was used to compute long-term mean annual precipitation rates for the 1979–2016 period (**Table 1**). The dataset is based on gauge and satellite observations and is gridded at a 2.5° x 2.5° scale. Estimates of precipitation uncertainty (Adler et al., 2012) accompany precipitation data. Data are provided by the National Oceanic & Atmospheric Administration/Oceanic & Atmospheric Research/Earth System Research Laboratory Physical Sciences Division (NOAA/OAR/ESRL PSD), Boulder, Colorado, USA, from their website at <http://www.esrl.noaa.gov/psd/>.

Long term mean air temperature data for the 1948–2016 period are from the National Centers for Environmental Protection (NCEP) reanalysis data (Kalnay et al., 1996) gridded at a 2.5° x 2.5° scale (**Table A.1**, Appendix A) and provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their website at <http://www.esrl.noaa.gov/psd/>.

Long term mean evaporation data for the 1958–2016 period and evaporation error estimates averaged over the 1985–2016 period gridded at a 1° x 1° scale (**Table A.1**, Appendix A) are from the Woods Hole Oceanographic Institution Objectively Analyzed air-sea Fluxes (OAFlux) for the Global Oceans project ([http://oaflux.whoi.edu](http://oaflux.whoi.edu/)) funded by the NOAA Climate Observations and Monitoring (COM) program (Yu et al., 2008).

Mean annual amount weighted station values of observed precipitation isotopes (δ2Hrain) and mean annual local gauge precipitation rates (**Table A.2; Fig. A.2**, Appendix A) are from the Global Network of Isotopes in Precipitation (GNIP) (IAEA/WMO, 2006) (https://nucleus.iaea.org/wiser/) and from the Japan Agency for Marine-Earth Science and Technology (Kurita et al., 2009) (<http://www.jamstec.go.jp/iorgc/cgi-bin/database/v01/browse_summary.cgi?program=hcorp&group=CRHCG&cat=Isotope&id=Isotope_Palau> ).

Online Isotope Precipitation Calculator (OIPC) precipitation δ2H estimates (δ2HOIPCrain) (**Table B.3**, Appendix B**; Fig. A.2**, Appendix A) are from the OIPC which uses the IAEA database and interpolation algorithms with a set of predictor parameters to calculate precipitation isotopes (Bowen and Revenaugh, 2003). The OIPC is accessible at http://wateriso.utah.edu/waterisotopes/pages/data\_access/oipc.html.

Precipitation δ2H estimates from the second Stable Water Isotope Intercomparison Group (SWING2) (Sturm et al., 2010) (δ2HSWING2rain) (**Table B.3**, Appendix B**; Fig. A.2; Fig. A.3**, Appendix A) were calculated from nudged isotope enabled global circulation models (Hoffmann et al., 1998; Schmidt et al., 2007; Yoshimura et al., 2008; Risi et al., 2010). Mean annual results for LMDZ, ECHAM, and GISS models are from <https://data.giss.nasa.gov/swing2/>. Mean monthly results from IsoGSM are from <http://hydro.iis.u-tokyo.ac.jp/~kei/?IsoGSM1#j9fb954c> and averaged. Model details are given in **Table A.3**, Appendix A.

**3. STUDY SITES**

The lakes in this study (**Fig. 1;** **Table 1; Table A.1**, Appendix A) were chosen to create a modern spatial calibration between δ2Hdinosterol and SPCZ precipitation. The lakes span 123.7° of longitude, 21.4° of latitude, and a narrow 23–27 °C range of NCEP/NCAR reanalysis mean annual air temperature where the largest (smallest) seasonal/inter-annual variability is 8.3 (3.7) °C in New Caledonia (Wallis) over the last 69 years (**Fig. A.4**, Appendix A). The lakes are all less than 250 m above sea level with the exception of Lake Lanoto’o, Samoa (762 m) and Tagimaucia Lake, Fiji (820 m) (**Table A.1**, Appendix A).

Across the study sites the mean annual GPCP precipitation rates for the 1979–2016 period vary from to 8.1 mm d−1 in the Solomon Islands to 3.5 mm d−1 in New Caledonia (**Fig. 1, Table 1**). Seasonal variability (**Fig. A.5**, Appendix A) results in greater SPCZ precipitation extent during the austral summer (Vincent, 1994) and ENSO variability results in a northeast shift to a more zonal position during El Niño events (Widlansky et al., 2011). Without wind speed data, site-specific evaporation rates cannot be calculated. However, mean annual ocean evaporation rates for the 1958–2016 period from OAFlux indicate the lowest evaporation rates in the Solomon Islands, Papua New Guinea, and Nauru (3.5–3.6 mm d−1) and the highest evaporation rates in Vanuatu (4.9 mm d−1) (**Table A.1; Fig. A.6**, Appendix A).

The largest lake in this study is Lake Wanum in Papua New Guinea (405 hectares) and the deepest is Lac Lalolalo on Wallis (89 m). The lakes on Grande Terre, New Caledonia are large flat bottom doline formations (Jeanpert et al., 2016). Most other lakes are small coastal ponds or crater lakes. Seven of the shallowest lakes had extensive floating vegetation. These include five lakes from Vanuatu plus the artificial Onetahi Pond on Tetiarora, French Polynesia that had *Nymphaea* spp. and *Nymphoides* spp. covering up to >50% of the water’s surface, and Lake Tagimaucia (Fiji), which had extensive (92.5%) floating mats of sedge peat (Southern et al., 1986). Profiles of salinity, temperature, DO%, and pH were recorded for many of the sites in Fiji, Vanuatu, Wallis, Samoa, and the Solomon Islands (**Fig. A.1**, Appendix A). The bathymetry and some physio-chemical properties of other sites has been further described for Lake Wanum in Papua New Guinea (Garrett-Jones, 1979), Lake Tagimaucia in Fiji (Southern et al., 1986; Southern, 1986), and Lanoto’o in Samoa (Parkes, 1994; Schabetsberger et al., 2009; Hassall, 2017), and Lake Lalolalo and Lanutavake on Wallis (Schabetsberger et al., 2009; Sichrowsky et al., 2014).

All lakes were fresh at the time of sampling with the exception of Rimatu’u pond (Tetiaroa, French Polynesia) and Lac Lalolalo (Wallis). Rimatu’u pond was observed to be brackish (5 ppt) in 1997 (Che et al., 2001), however the shoreline that separates the pond from the sea water lagoon has been growing since 1997(Le Cozannet et al., 2013), presumably assisting the freshening of the pond. It was fresh when sampled in March 2015 but had a salinity of 2 ppt when sampled at the end of the dry season in October 2015. The effect of salinity on δ2H values of microalgal lipids is 1–2‰ ppt−1 (Schouten et al., 2006; Sachse and Sachs, 2008; Sachs and Schwab, 2011; Chivall et al., 2014; M’Boule et al., 2014; Nelson and Sachs, 2014b; Maloney et al., 2016; Sachs et al., 2016; Weiss et al., 2017), meaning that a 5 ppt increase in salinity could cause a 2H-enrichment of 5–10‰, within or slightly larger than the analytical error of δ2Hdinosterol measurements. Lac Lalolalo contained freshwater (<1 ppt) in the oxygenated photic zone (a secchi disk reading revealed that light penetrated only 3.4 m), but below 10 m salinity gradually increased reaching 3 ppt at 50 m and finally, 31 ppt at 80 m (Sichrowsky et al., 2014). Given the strong gradient in light and oxygen, it is unlikely that dinoflagellates lived outside the fresh region of Lac Lalolalo. Lakes near the coast may occasionally experience brackish salinities due to storm overwash events. After our 2012 visit, Lake Otas on Efate Island experienced a flooding event in 2015 during cyclone Pam (Hong et al., 2018), but it is unknown how this event affected the salinity or water isotope values.

Some lakes such as Barora Pond (Tetepare, Solomon Islands), Red lake (Thion, Vanuatu), and Lake Otas (Efate, Vanuatu) (**Fig. A.1**, Appendix A) were supersaturated in oxygen at the surface indicating high productivity rates at the time of sampling. Many lakes had low/zero oxygen at the sediment-water interface, particularly those with supersaturation at the surface. Most of the lakes in this study have not been examined for algal species composition but dinosterol was found in all sediments collected, confirming the existence of dinoflagellates in these lakes. Lalolalo and Lanutavke crater lakes on Wallis Island have been relatively well-studied and contain dinoflagellates of the genera *Gymnodinium* and *Peridinium* (Schabetsberger et al., 2009; Sichrowsky et al., 2014), the latter a known producer of dinosterol (c.f. Atwood et al., 2014). Lake Tagimaucia (Fiji) hosts at least four species of dinoflagellates from the genera *Gymnodinium*, *Gyrodinium*, and *Peridinium*, and Lake Lanoto’o (Samoa) has at least one species of *Peridinium* (Schabetsberger et al., 2009).

The sediment in most lakes was highly unconsolidated algal gytta. Sediment in Grand Lac (Grande Terre, New Caledonia) and Barora Pond (Tetepare, Solomon Islands) had high amounts of lithogenic material and the sediments in Harai Lake #2 (Rendova, Solomon Islands) contained sandy material. Onetahi Pond (Tetiarora, French Polynesia) was built in the early 2010s so sediment has only been accumulating for a few years. Radiometric dating in 15 lakes verifies that core top sediments are not older than 1960 (**Table A.4; Table A.5**, Appendix A). We estimate that the core top sediments in this study incorporate several years of accumulation ranging from 0.4±3 years cm−1 to 20±50 years cm−1, further details are provided in the Supplemental Information (**Table A.1**, Appendix A).

**4. RESULTS**

**4.1. Precipitation δ2H and δ18O values**

There are 15 IAEA/GNIP stations and 1 JAMSTEC station (Palau) (**Fig. 1**) on tropical Pacific islands between 30°N and 30°S with at least 23 δ2H measurements from at least 3 years (**Table A.2**, Appendix A). The 9 mm d−1 range in precipitation rates vs. the 44‰ range in mean annual amount-weighted δ2Hrain from the 16 stations yields a tropical Pacific amount effect with a slope of −4.5±0.6‰ (mm d−1) −1 [R2=0.42, p<0.001], note the Hilo station data fell well above the 95% prediction interval (**Fig. A.2a**, Appendix A). Even though most GNIP station sample collection dates (many in the 1960s) did not overlap with the GPCP 1979–2016 period, δ2Hrain was better correlated with GPCP precipitation data (**Fig. A.2b**, Appendix A**; Fig. 2**), and had a statistically indistinguishable (p=0.3, t-test) slope of −5.6±0.8‰ (mm d−1) −1 [R2=0.81, p<0.001].

At the 30 SPCZ sampling locations used in this study, multi-model mean δ2HSWINGrain data from four nudged isotope-enabled global circulation models (**Fig. A.3**, Appendix A**; Table B.3**, Appendix B) had a 22‰ range and a 7.3 mm d−1 range in multi-model mean precipitation rates. δ2HSWINGrain vs. multi-model mean precipitation rates gave a smaller amount effect slope of −2.9±0.6 (mm d−1) −1 [R2=0.66, p<0.001] (**Fig. A.2c**, Appendix A), about half the size as that between the GNIP/JAMSTEC observation data and the GPCP reanalysis data. δ2HOIPCrain values from the 30 sample sites in this study spanned 34‰, with very large uncertainties at several lake sites, especially sites in Vanuatu (**Table B.3**, Appendix B**, Fig. A.2d**, Appendix A). δ2HOIPCrain values vs. GPCP precipitation rates gave an amount effect slope of −8.6±2.4 (mm d−1) −1 [R2=0.72, p=0.002], ~two times larger than the GNIP/JAMSTEC – GPCP relationship.

**4.2. Regional meteoric water line**

δ2Hwater and δ18Owater of 38 rain events collected in Nauru, Fiji, Wallis, Solomon Islands, Vanuatu, and Tetiaroa between 2010–2016 (**Table B.2**, Appendix B**; Fig. 3**) gave a regional meteoric line [RMWL= 7.6(±0.3)x+11.5(±0.9), R2=0.93, p<0.001] with a slope similar to south Pacific tropical IAEA/GNIP data (station-specific GNIP slopes ranged from 6.4 to 8.1 (with a 7.3±0.1 mean)), but with a larger intercept than GNIP data (station-specific GNIP intercepts ranged from 0.0 to 10.4 (with a 6.7±0.3 mean) (**Fig. A.7**, Appendix A)). Water samples from Fiji, Wallis, and Vanuatu representative of long-term precipitation from rain tanks, bottled and tap water, streams, and cave drip water, (**Fig. 3**) in addition to published precipitation isotope data from Tahiti (Hildenbrand et al., 2005) and Papua New Guinea (Ferguson, 2007; Ferguson et al., 2011) agree with the RMWL (**Fig. A.8**, Appendix A). As expected for samples containing seawater, brackish and salty water samples from lakes, channels, lagoons, and ocean water with salinities between 2–35 ppt fell off the RMWL (**Fig. 3**).

**4.3. Lake water δ2H and δ18O values**

**Figure 3** shows individual δ2Hlakewater samples, largely to the right of the RMWL. Lakes on the Solomon Islands had the most isotopically depleted water and plotted closest to the RMWL while Tetiaroa lakes showed the greatest extent of evolution away from the RMWL due to the presumed greater extent of evaporation (and associated kinetic and equilibrium isotope effects) (Henderson and Shuman, 2009). Samples from higher-elevation lakes Lake Tagimaucia (Fiji), Lake Lanoto’o (Samoa), and both lakes on New Caledonia fell on/near the RMWL indicating these lakes experienced little evaporation at the time of sampling. Three lakes were sampled at two different time points. Lake Lanoto’o (Samoa) was sampled in July 2013 and September 2014 and surface δ2Hlakewaterdiffered by 4‰ (Hassall, 2017). Rimatu’u Pond (Tetiaroa) was sampled once during the wet season in March 2015 and once during the end of the dry season in October 2015. The October 2015 sample had a δ2Hlakewater value 7‰ higher and a salinity 2 ppt higher (2 vs 0 ppt) than the March 2015 sample. Emaotul Lake (Efate, Vanuatu) was sampled in September 2015 (−5‰) and again in May 2017 (−26‰) when it had a 21‰ lower δ2Hlakewater value (highlighted in green in **Fig. 3**). Because the seasonal variation in lake water isotopes is otherwise unknown, we adopte an estimated δ2H error value of 5‰ for each lake-averaged value, which incorporates the assumption that lake water integrates the δ2H value of precipitation over multiple events (Nelson and Sachs, 2014b). However, due to the large observed change in water isotopes in Lake Emaotul, the standard deviation of its two measurements (14.4‰) was used in all statistics and regressions. Lake Wanum (Papua New Guinea) was the only lake with a sediment sample but no water samples. All water sample data is available in **Table B.2**, Appendix B.

The lake-averaged δ2Hlakewater samples spanned a 52‰ range, from −29.4‰ to +23.0‰ (**Table 1**). δ2Hlakewater varied by 25‰ among eight Solomon Islands lakes, 20‰ among three Tetiarora lakes, 19‰ among ten Vanuatu lakes, and 10‰ among three Wallis Islands lakes. The 20‰ range among three nearby Tetiarora lakes was highly influenced by 2H-depleted Onetahi Pond with extensive (>50%) vegetation cover. Likewise, the 19‰ range in δ2Hlakewater among ten Vanuatu lakes is reduced to 8‰ after removing five lakes with >50% vegetation cover. Accordingly, >50% vegetation cover sites, in addition to Lake Tagimaucia (Fiji) covered in floating sedge peat (Southern et al., 1986), were not included in environmental regressions owing to their attenuated evaporative 2H-enrichment. Indeed, sites with >50% vegetation cover (highlighted with crosses in **Fig. 4**) are the most 2H-depleted lakes when compared to others from the same region.

δ2Hlakewater values (n=22) were inversely correlated with GPCP precipitation rates [r=−0.51, R2=0.26, p<0.001] (**Fig. 2**). δ2Hlakewater values did not show a strong correlation with OAFlux evaporation rates [r=0.23, R2=0.05, p=0.04] (**Fig. A.7c**), but were inversely correlated with P−E [r=−0.48, R2=0.23, p<0.001] (**Fig. A.7d**). δ2Hlakewater values were also correlated [r=−0.53, R2=0.28, p=0.02] (**Fig. A.7b**) with δ2Hrain values calculated from the tropical Pacific amount effect using GPCP rain rates. Note that these regressions did not include δ2Hlakewater values from seven lakes with vegetation cover >50% since the effects of local evaporation were inhibited.

**4.4. δ2Hdinosterol values**

All 44 δ2Hdinosterol values (**Table B.1**, Appendix B) from core top samples were 2H-depleted relative to δ2Hlakewater values. Lake-averaged δ2Hdinosterol core top values from (n=21 sites) spanned a 69‰ range, from −316‰ to −247‰ (**Table 1**) and had a statistically significant relationship with δ2Hlakewater values [r=−0.71, R2=0.50, p<0.001] (**Fig. 4**), where the regression includes vegetation-covered lakes to demonstrate the fidelity with which algal lipids track hydrogen isotopic changes in their source water. δ2Hdinosterol was inversely correlated with GPCP precipitation rates [r=−0.77, R2=0.59, p<0.001] (**Fig. 2**) with a slope (−12.1±2.6 ‰ (mm d−1)−1), similar to the δ2Hlakewater–GPCP relationship (−10.1±2.4 ‰ (mm d−1)−1). 2Hdinosterol was inversely correlated with OAFlux evaporation rates [r=0.48, R2=0.24, p=0.004] (**Fig. A.7c**) and GPCP precipitation minus OAFlux evaporation (P−E) [r=−0.73, R2=0.54, p<0.001] (**Fig. A.7d**). δ2Hdinosterol was also correlated with δ2Hrain values calculated from the tropical Pacific amount effect equation using GPCP rain rates at each lake site [r=0.77, R2=0.59, p=0.005] (**Fig. A.7b**). Once again, these regressions did not include δ2Hdinosterol values from three lakes with vegetation cover >50% since the effects of local evaporation were inhibited.

The offset between δ2Hdinosterol and δ2Hlakewater at 17 sites, expressed as fractionation, εdinosterol/lakewater = (α−1)\*1000, α = [(2H/1H)dinosterol / (2H/1H)lakewater] = [δ2Hdinosterol +1000 / δ2Hlakewater +1000], had a 58‰ range from −297 to −239 (**Table 1**). εdinosterol/lakewater was correlated with GPCP precipitation rate [r=−0.69, R2=0.48, p=0.0009] (**Fig. 5a**), δ2Hrain [r=0.68, R2=0.46, p=0.01] (**Fig. 5b**), evaporation [r=0.53, R2=0.29, p=0.01] (**Fig. 5c**), and P−E [r=−0.69, R2=0.47, p=0.0002] (**Fig. 5d**).

**5. DISCUSSION**

**5.1. δ2Hdinosterol values**

The slope of the δ2Hdinosterol–GPCP relationship (−12.1±2.6‰ (mm d−1) −1) was similar to the δ2Hlakewater–GPCP relationship (−10.1±2.4‰ (mm d−1) −1), both of which are steeper than the δ2Hrain vs GPCP rainfall (amount effect) relationship from GNIP/JAMSTEC stations (−5.6±0.8‰ (mm d−1) −1) (**Fig. 2**). This is due to the enriching effects of evaporation on δ2Hlakewater values that also manifests in δ2Hdinosterol values from dinoflagellates that use this lake water as a primary hydrogen source. **Figure 2** aptly demonstrates how hydrologic conditions are reflected in the dinosterol biomarker proxy via the links between precipitation and lake water evaporation.

It might be expected that the relationship between δ2Hdinosterol and P−E would result in an improved relationship compared to the δ2Hdinosterol–GPCP relationship. However, the r and R2 values for the δ2Hdinosterol–P−E relationship [r=−0.73, R2=0.54, p<0.001, n=21] (**Fig. A.7**) were lower than for the δ2Hdinosterol–GPCP relationship [r=−0.77, R2=0.59, p<0.001, n=21] (**Fig. 2**). This implies that: (1) the gridded ocean evaporation product does not adequately reflect evaporation rates at the studied lakes, (2) evaporation is not as important as precipitation in determining lake water isotopes, (3) lake water residence times are too short to match the long-term average evaporation rates, or a combination thereof. The OAFlux evaporation rates (3.5 to 4.9 mm d−1) have a smaller range compared to the GPCP precipitation rates (3.5 to 8.1 mm d−1) across the sample sites. Indeed, the P−E spatial pattern is dominated by P in this region (Schanze et al., 2010), with relatively constant evaporation rates among all sites, which partly explains why δ2Hdinosterol is not better correlated with P−E. Future improvements in gridded hydrological products could help clarify why the δ2Hdinosterol relationship with P−E was weaker than with precipitation alone. Given the uncertainties in applying an ocean evaporation product to the lake sites, we propose that the δ2Hdinosterol–GPCP relationship is most robust for quantifying precipitation in the SPCZ region but assumes that the relationship between P and E is constant through time.

The correlation between δ2Hdinosterol and δ2Hlakewater (**Fig. 4**) emphasizes that environmental water isotopes are the primary control on algal lipid isotopes, at least for dinosterol in the tropical Pacific. This is in agreement with numerous studies that show microorganism lipids track the isotopes of growth water in the lab (Paul, 2002; Englebrecht and Sachs, 2005; Zhang and Sachs, 2007; X. Zhang et al., 2009; Dirghangi and Pagani, 2013b; Dirghangi and Pagani, 2013a; Osburn et al., 2016) and field (Sauer et al., 2001; Huang et al., 2004; Sachse et al., 2004; Polissar and Freeman, 2010; Sachs and Schwab, 2011; Schwab and Sachs, 2011; Sachse et al., 2012; Schwab et al., 2015a).

The slope of the SPCZ δ2Hlakewater-δ2Hdinosterol relationship (1.9±0.3) is larger than slopes of various other (non-dinosterol) δ2Hlakewater–δ2Hlipid relationships determined from freshwater cultures of Chlorophyceae and Trebouxiophyceae, which ranged from 0.7 to 1.0 (Zhang and Sachs, 2007). It is also steeper than the slopes of δ2Hdinosterol–δ2Hlakewater relationships from suspended particle (0.7±0.2) and sediments (1.5±0.3) in freshwater lakes across Cameroon (Schwab et al., 2015a). The Cameroon sediment samples were hypothesized to be influenced by increased dinoflagellate heterotrophy or post-depositional δ2Hdinosterol alterations (Schwab et al., 2015a). We note however, that if errors in SPCZ δ2Hlakewater were disregarded and a simple ordinary least squares regression was used for the freshwater SPCZ lakes (as in the previously cited studies), the relationship would be within error of freshwater cultures and also Cameroon suspended particle data (1.1±0.3). Since sedimentary aquatic lipids incorporate several years of accumulation, better estimates of long-term mean values for δ2Hlakewater and knowledge about the timing of dinosterol production in relation to δ2Hlakewater could indicate if the SPCZ δ2Hlakewater–δ2Hdinosterol relationship is different from other freshwater algal lipids, or if it is a product of the regression technique used.

Non-hydrologic environmental factors including metabolism, growth rate or growth stage, irradiance, and species composition may contribute to the scatter in the SPCZ δ2Hlakewater–δ2Hdinosterol relationship (Fig. 4) and also to the relationship between fractionation and precipitation rates (Fig. 5a). Differences in metabolic state can have a large impact on 2H/1H ratios in lipids, at least in prokaryotes (X. Zhang et al., 2009; Heinzelmann et al., 2015; Osburn et al., 2016). Since heterotrophic dinoflagellates can contribute to the sedimentary record (Amo et al., 2007), the extent to which dinoflagellates utilize photoautotrophy versus heterotrophy to support cellular function could potentially influence δ2Hdinosterol values at some sites more than others. In addition, differences in growth rate/phase or productivity (Z. Zhang et al., 2009; Wolhowe et al., 2009; Chivall et al., 2014; Heinzelmann et al., 2015; Sachs and Kawka, 2015; Wolhowe et al., 2015; Ladd et al., 2017) and irradiance (van der Meer et al., 2015; Wolhowe et al., 2015; Sachs et al., 2017) are known to influence lipid-water 2H/1H fractionation so monitoring productivity and turbidity of lake waters could indicate if these factors play a role in SPCZ freshwater lakes. One possible explanation for the inverse relationship between εdinosterol/lakewater and precipitation rates or P−E (and normal relationship between and evaporation and δ2Hrain) is that increased precipitation rate could lead to increased nutrient delivery, and hence higher growth rates at wetter sites. Sachs and Kawka (2015) have shown that cultured algal sterols exhibit increased fractionation at higher growth rates and Ladd et al. (2018) found increased fractionation in a diatom sterol biomarker in more productive central Swiss lakes. Furthermore, given the large diversity in dinosterol producing dinoflagellates (Janouškovec et al., 2016), it is possible that the large spatial and environmental distributions in this study harbor distinct species assemblages, and that the changes in species assemblage along the studied rainfall gradient enhance the relationship between dinosterol δ2H values and rainfall amount.

Importantly, precipitation rate alone may not be only environmental factor responsible for the modern spatial relationship between δ2Hdinosterol and GPCP precipitation rates, and region-specific core top calibrations will benefit from future research into the ways lipid/water apparent fractionation can change. While impossible to fully characterize potential influences of metabolism, growth rate or growth stage, irradiance, and species composition in our data, many lakes with diverse physical and chemical properties were targeted to capture the range of species, heterotrophy, light, and nutrient-driven growth rate effects. Despite these potential influences on the resulting scatter, the SPCZ δ2Hlakewater–δ2Hdinosterol relationship, together with the robust relationship between δ2Hdinosterol and precipitation rates, which is the best empirical fit to the data available, supports the use of δ2Hdinosterol as a proxy for hydroclimate variations.

**5.2. δ2Hrain values and the amount effect**

Tropical precipitation isotope observations are sparse in space and time but characterizing spatial variability in modern δ2Hrain values is important for understanding the controls on the isotopic composition of lake water and dinosterol. Several studies have already established a strong inverse relationship between precipitation amount and δ2Hrain values in the maritime tropics (Dansgaard, 1964; Rozanski et al., 1993; Araguás-Araguás et al., 1998; Kurita et al., 2009; Conroy et al., 2013; Martin et al., 2017). This relationship is most robust over annual timescales (Conroy et al., 2013) and when isotope data are compared to regional (as opposed to local) precipitation rates (Kurita et al., 2009).

Recent advancements in isotope-incorporated general circulation models have greatly improved overall understanding of the behavior of water isotopes (Yoshimura, 2015) and improvements in interpolation algorithms have aided prediction of water isotopes in remote areas (Bowen, 2010). It is not clear why the amount effect obtained from SWING2 data was smaller (−2.3±0.6‰ (mm d−1) −1), while the amount effect generated using OIPC was larger (−8.1±2.2‰ (mm d−1) −1) than the observed amount effect from GNIP/JAMSTEC station data (−4.5±0.6‰ to −5.6±0.8‰ (mm d−1) −1)(**Fig. A.2**, Appendix A). These differences could be tied to the fundamental differences between GCMs and the real world in the case of the SWING2 approach, and data scarcity in the case of the OIPC approach. This comparison highlights the need for caution when using these tools to calculate precipitation isotopes in remote parts of the globe like the tropical southwest Pacific. Irrespective of the true cause of the amount effect and the differences between approaches to characterize the size of the amount effect, all three methods indicate that higher mean SPCZ precipitation rates are associated with 2H-depleted precipitation, and that the linear nature of this relationship is consistent across the SPCZ region.

**5.3. δ2Hlakewater values**

δ2Hlakewater values are correlated with GPCP precipitation rates with a steeper slope than δ2Hrain due to evaporative enrichment (**Fig. 2**). The effects of evaporation are also apparent from the evolution of paired lake water δ2H–δ18O values that fall to the right of the RMWL line (**Fig. 3**). However, δ2Hlakewater values are not well correlated with mean ocean evaporation rates (**Fig. A7**, Appendix A). The dinosterol samples were better correlated with OAFlux evaporation (r=0.48, R2=0.24, p=0.004, n=21) than lake water samples (r=0.23, R2=0.05, p=0.04, n=22), which could be a result of the greater timespan represented by dinosterol in core top sediment (years) than lake water (weeks–months).

Most of the lake water samples in this study represent snapshots in time and may not reflect mean annual δ2Hlakewater values. Additional sampling would clearly be of great value. For instance, two visits in 2015 and 2017 to Lake Emaotul (Efate, Vanuatu) revealed a surprisingly large 20‰ change in δ2Hlakewater values; the 2015 sample coincided with El Niño drought and the 2017 sample was collected shortly after Cyclone Donna, which increased lake depth from 6 to 6.4 m. While this range of values might only indicate contrasting extreme events in this case, only repeat field sampling or continuous monitoring would answer this question definitively.

Some of the local variability in δ2Hlakewater values is related to lake catchment area and residence time among lakes in close proximity to each other (Leng and Marshall, 2004; Henderson and Shuman, 2009). The δ2Hlakewater value from (aquatic vegetation-free) Red Lake (Thion, Vanuatu) was −16.3‰ but its larger neighbor White Lake (also aquatic vegetation-free) was relatively 2H-enriched with a value of −10‰ (**Table 1**). The difference may be due to Red Lake’s small surface area to volume ratio, and potentially shorter residence time, responding more readily to sporadic 2H-depleted rain events, especially cyclones, or evaporation. For the three neighboring lakes on Wallis Island, Lac Lalolalo, Lac Lanutavake, and Lac Lano, all were vegetation free and had a 10‰ range in δ2Hlakewater where the smallest lake (Lac Lano, 4m deep, 1.9 hectares) was −2.4‰ while the largest lake (Lac Lalolao, 89m deep, 18 hectares) was −12.3‰, reflecting the greater impact that evaporation has on lakes with larger surface area to volume ratios. Measured δ2Hlakewater values varied by ~25‰ among eight lakes in the Solomon Islands; the three Rendova Island Harai lakes to the north were more 2H-depleted compared to the large Lake Rano in the south. This is likely due to the highlands in the north that contribute to the watershed for many of the Harai lakes compared to the lower-altitude catchment for Lake Rano. Evidently, several secondary factors influence δ2Hlakewater values, and may partially obscure the relationship between the relatively coarsely gridded OAFlux product and δ2Hlakewater. Nevertheless, the strong correlation between precipitation rates and δ2Hlakewater values [r=−0.51, R2=0.26, p<0.001, n=22] (**Fig. 2**), and between δ2Hrain and δ2Hlakewater values [r=−0.53, R2=0.28, p=0.02, n=22] (**Fig. A.7b**), and the SPCZ lakes that fall to the right of the RMWL indicates that δ2Hrain (via precipitation rate) and evaporative enrichment are the primary influences on δ2Hlakewater.

**5.4. Paleoclimate applications**

The ultimate goal of this study is to understand the environmental controls on δ2Hdinosterol values in order to apply this knowledge to reconstructions of past environmental change using tropical southwest Pacific lake sediments. Despite the potential for a variety of secondary impacts on δ2Hlakewater and δ2Hdinosterol, δ2Hdinosterol values have the best empirical relationship with GPCP precipitation rates across the SPCZ compared to any tested environmental variable (**Fig. 2**). Notwithstanding the indirect connection, the major hydrological patterns of the SPCZ are clearly reflected in the dinosterol δ2H values from recent sediments, creating a useful empirical relationship that can be applied across most of the SPCZ region. Three steps are required to get from rainfall rates to dinosterol δ2H values: 1) precipitation rate to δ2Hrain, 2) δ2Hrain (and evaporation) to δ2Hlakewater, and 3) δ2Hlakewater to δ2Hdinosterol. While the exact nature of the isotope fractionations at each of these steps still requires additional study, the empirical SPCZ δ2Hdinosterol–GPCP relationship from freshwater lakes implies that it can be used in the SPCZ region to quantify past regional precipitation rates, with the acknowledged assumption of stationarity through time.

Notably, the lakes in this study do not have rain gauges on site, and the effects of topography on localized mean annual precipitation rate are not captured by the 2.5° x 2.5° grid scale used in GPCP data. Rain gauge data at nearby locations indicate that GPCP underestimates actual precipitation rates at some sites. For instance, a rain gauge on Tetepare, Solomon Islands recorded a mean annual precipitation rate of ~13 mm d−1 between July 2003 and July 2009 (Read et al., 2010) while the GPCP estimate is 8.4±1.1 mm d−1 for the same period. Likewise, precipitation at Lake Tagimaucia (Taveuni, Fiji) was estimated to be somewhere between 15–26 mm d−1 based on rain gauges from other parts of the island recorded in 1976–1983 and 1913–1974 (Southern et al., 1986) while the GPCP estimate is 4.6±1.2 mm d−1 for the 1979–2016 period. Despite these disagreements, GPCP offers the most temporally and spatially comprehensive data available for understanding modern precipitation in the maritime tropical Pacific. Therefore, reconstructions based on the calibration presented here must be interpreted as GPCP-like reconstructions of large-scale precipitation patterns.

The most straightforward approach to applying this calibration is re-arrangement of the modern δ2Hdinosterol–GPCP relationship (**Fig. 2**) to calculate paleoprecipitation down core:

Pp = δ2Hsample − b / m [eq. 1]

where Pp is the paleoprecipitation rate, δ2Hsample is a down core measurement of dinosterol, b is the intercept of the regression (−211±15), and m is the slope, or sensitivity, of the regression (−12.1±2.6).

**Figure 6** illustrates how uncertainty in reconstructed precipitation rates varies with δ2Hsample values. Slope and intercept errors from the calibration regression (eq. 1) and analytical uncertainty in δ2Hdinoterol measurements (±6.2‰ in this study) were propagated using a Monte Carlo approach with 100,000 iterations. Calculated precipitation errors are about twice as large (±1.5 to 3.1 mm d−1) as errors in the modern SPCZ GPCP precipitation rates (±0.3 to 1.5 mm d−1); importantly with minimum uncertainties in the former matching maximum uncertainties in the latter.

 The δ2Hdinosterol–GPCP calibration offers the opportunity to extend estimates of large scale mean SPCZ precipitation into the past. The best application would make use of sediment cores from multiple lakes from different areas of the SPCZ to map spatial patterns of precipitation through time. A network of coring sites would allow for differentiation between changes in intensity and position of the SPCZ feature. Furthermore, duplication of records from islands that have nearby lakes would ensure proper interpretation of hydrological changes.

**6. CONCLUSION**

 This work investigated the environmental controls on the 2H/1H ratios of SPCZ lake waters and of the dinoflagellate sterol, dinosterol, purified from recent freshwater lake sediments. The 30 lakes in this study represent diverse freshwater ecosystems with varying physical and chemical parameters. The main controls on δ2Hlakewater were precipitation rateand evaporation. Additional lake-specific environmental properties such as lake surface area-to-volume, water residence time, trajectories of source water, and catchment area likely contribute to δ2Hlakewater variability in each lake and between lakes in the same region. While environmental and growth parameters such as light, growth rate, metabolism, and species composition likely contribute to δ2Hdinosterol variability, we conclude that the main control on δ2Hdinosterol was δ2Hlakewater. The stronger relationship between GPCP precipitation rate and δ2Hdinosterol values, rather than with δ2Hlakewater values, suggests that secondary factors contribute to the strength of the correlation, or that measured δ2Hlakewater values do not accurately reflect the mean source water that was used for dinosterol production. The breadth of physical parameters that impact δ2Hlakewater, combined with the range of environmental/growth processes that impact δ2Hdinosterol, highlight the importance of sampling multiple lakes in each region. The empirical SPCZ 2Hdinosterol–GPCP relationship provides a method to quantitatively reconstruct past precipitation rates with uncertainties only about twofold larger than the best climatological rainfall estimates available in this region today.

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FIGURE AND TABLE CAPTIONS

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**Fig. 1.** Location of 29 SPCZ freshwater lake water samples (open white triangles) from nine island groups, 21 freshwater lake sediment samples (red squares) from nine island groups, and 16 tropical Pacific GNIP or JAMSTEC (Palau) stations (yellow circles). Contours and color scale show mean GPCPv2.3 precipitation rate (mm d−1) on a 2.5° x 2.5° grid for the 1979–2016 period ([http://www.esrl.noaa.gov/psd/)](http://www.esrl.noaa.gov/psd/%29).



**Fig. 2.** Maximum likelihood regressions for GPCPv2.3 mean annual precipitation rate (mm d−1) for the 1979–2016 period and δ2Hdinosterol,δ2Hlakewater, andδ2Hrain values. Lake-averaged δ2Hdinosterol values (red squares) from 18 lakes are plotted on the left axis. Also plotted are 3 lakes with >50% vegetation cover (dark red crosses) that are not included in the regression. On the right axis: lake-averaged δ2Hlakewater values (white triangles) from 22 lakes in addition to 7 lakes with >50% vegetation cover (black crosses) that are not included in the regression, and GNIP/JAMSTEC δ2Hrain values (yellow circles) from 16 stations (same as **Fig. A.2b**, Appendix A). X-axis errors are precipitation rate errors provided by GPCP. δ2Hlakewater error bars are the estimated standard error of tropical lake water (±5‰, except 14‰ at Lake Emaotul, see text), δ2Hrain error bars are GNIP/JAMSTEC weighted standard deviations, and δ2Hdinosterol error bars are the standard deviations of multiple core top values. When only one core top was measured, the δ2Hdinosterol pooled standard deviation is shown (±10.4‰ indicated by error bars with black dashes). The dark and light shaded bands around the regression lines are the 95% confidence and prediction intervals. *Not shown* ordinary least squares regressions:

δ2Hlakewater = −3.9(±1.4)\*P+11(±9), R2=0.27, p=0.01, n=22

δ2Hrain = −3.5(±0.7)\*P−6(±3), R2=0.67, p=0.0001, n=16

δ2Hdinosterol = −9.6(±2.0)\*P−223(±12), R2=0.58, p=0.0002, n=18

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**Fig. 3**. Maximum likelihood regression of hydrogen and oxygen isotope values from SPCZ rain events. The GMWL is shown for reference (thin black line y=8x+10). Samples from 38 local rain events (yellow circles) create a regional meteoric water line (RMWL) represented by the thick brown line (dark and light shading indicates the 95% confidence and prediction intervals, respectively). Water samples from 16 rain tanks, bottled or tap water, streams, and cave drip water (black dots) presumably average multiple rain events during longer timescales than discrete rain samples and generally fall along the RMWL. Gray diamonds represent 31 samples collected from brackish coastal ponds, inlets, lagoons, and the ocean at salinities from 2 to 35 ppt. Individual freshwater lake samples (n=61 triangles) from 29 lakes fall along the RMWL at sites with high precipitation rate or high altitudes, and away from the RMWL at sites with low precipitation rates. Green triangles represent Lake Emaotul (Efate, Vanuatu), with the sample collected in September 2015 2H-enriched by 20‰ compared to the sample collected in May 2017. Analytical uncertainty in hydrogen isotope values (up to 1.3‰, Table B.2) and oxygen isotope values (up to 0.3‰, Table B.2) is not shown for clarity.

*Not shown* ordinary least squares regression:

RMWL: y=7.2(±0.3)x+10.4(±1.0), R2=0.93, p<<0.001, n=38

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**Fig. 4.** Maximum likelihood regression of δ2Hlakewater vs. δ2Hdinosterol values from 20 SPCZ lakes. Lakes in French Polynesia (pink) have the most 2H-enriched δ2Hlakewater and δ2Hdinosterol values. New Caledonia (light blue), Wallis Island (blue), Fiji (open purple), Samoa (yellow), and Vanuatu (green) lakes are less 2H-enriched. Lakes in the Solomon Islands (red) have the most 2H-depleted δ2Hlakewater and δ2Hdinosterol values. δ2Hlakewater error bars are the estimated standard error of tropical lake water (±5‰, except 14‰ at Lake Emaotul, see text), whereas δ2Hdinosterol error bars are the standard deviations of multiple core top values. Where only one core top was measured, the δ2Hdinosterol pooled standard deviation is shown (±10.4‰ indicated by error bars with black dashes). The dark (light) shaded regions represent the 95% confidence (prediction) intervals. Samples from lakes with >50% vegetation cover are indicated with crosses, and they are included in the regression (black line). *Not shown* ordinary least squares regression: y=1.1(±0.3)x−268(±5), R2=0.49, p=0.0005, n=20



**Fig. 5.** Maximum likelihood regressions of εdinosterol/lakewater on (a) GPCPv2.3 mean annual precipitation rate (mm d−1) for the 1979–2016 period ([http://www.esrl.noaa.gov/psd/)](http://www.esrl.noaa.gov/psd/%29), (b) δ2Hrain (δ2Hrain was calculated using the GPCP rainfall at each lake site and the Tropical Pacific amount effect relationship between GNIP/JAMSTEK station data and GPCP rainfall rates), (c) mean annual evaporation rate (mm d−1) for the 1958–2016 period from OAFlux (Yu et al., 2008) (<http://oaflux.whoi.edu/evap.html> ), and (d) P−E (mm d−1) (P−E = GPCP mean annual precipitation rate for the 1979–2016 period − OAFlux mean annual evaporation rate for the 1958–2016 period). Also shown are 3 lakes with vegetation cover >50% not included in regression (black crosses). X-axis error bars represent estimated systematic mean errors provided by (a) GPCP, (c) OAFlux, and (d) propagated errors provided by GPCP and OAFlux products, or in panel (b) were calculated using a Monte Carlo approach with 10000 iterations and account for errors from GPCP rainfall estimates plus errors in the slope and intercept of the “amount effect” relationship. εdinosterol/lakewater error bars are the propagated errors from standard deviations of multiple core top δ2Hdinosterol values and estimated standard error of tropical lake water δ2Hlakewater (±5‰, except 14‰ at Lake Emaotul, see text). When only one core top δ2Hdinosterol value was available, the pooled standard deviation calculated from lakes with multiple core tops was used (±10.4‰, indicated by error bars with black dashes). Regressions (solid or dotted black lines) are shown with 95% confidence and prediction intervals (dark and light shading, respectively). The dotted lines were significant below the p=0.05 level, not the p=0.01 level for the maximum likelihood regressions.



**Fig. 6.** Uncertainties in calculated precipitation using equation 1. Panel a) Calculated P (thin black line) for a range of modeled δ2Hdinosterol values with uncertainties from three sources. Panel b) summarizes the magnitude of uncertainty in calculated P. Uncertainties were calculated using a Monte Carlo approach with 100,000 iterations with normally distributed errors from analytical uncertainty in δ2Hdinosterol measurements (±6.2‰) (thick gray lines), plus calibration error in the slope (±2.6) (dark gray dashed lines) and intercept (±15) (thick black lines).

**Table 1.** Lake location, precipitation rate, lake-average isotope values, and fractionation.

