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2   Leaf Wax Hydrogen Isotopes as a Hydroclimate Proxy in the Tropical Pacific

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16 **Key Points:**

17   •   Leaf wax ^{2}H/^{1}H ratios are correlated with mean annual precipitation ^{2}H/^{1}H ratios
18   globally, but not in the tropical Pacific

19   •   Deviations from the global relationship between precipitation leaf wax ^{2}H/^{1}H ratios
20   cannot be predicted from palynological assemblages

21   •   Small range and large uncertainties in estimates of tropical Pacific precipitation ^{2}H/^{1}H
22   ratios likely account for poor correlations
23

24   **Abstract**

25   Hydrogen isotope ratios of sedimentary leaf waxes (δ^2H$_{Wax}$ values) are increasingly used

26   to reconstruct past hydroclimate. Here, we add δ^2H$_{Wax}$ values from 19 lakes and four swamps on

27   15 tropical Pacific islands to an updated global compilation of published data from surface

28   sediments and soils. Globally, there is a strong positive linear correlation between δ^2H values of

29   mean annual precipitation (δ^2H$_P$ values) and the leaf waxes *n*-C$_{29}$-alkane (R^2 = 0.74, n = 665)

30   and *n*-C$_{28}$-acid (R^2 = 0.74, n = 242). Tropical Pacific δ^2H$_{Wax}$ values fall within the predicted

31   range of values based on the global calibration, and the largest residuals from the global

32   regression line are no greater than those observed elsewhere, despite large uncertainties in δ^2H$_P$

33   values at some Pacific sites. However, tropical Pacific δ^2H$_{Wax}$ values in isolation are not

34   correlated with estimated δ^2H$_P$ values from isoscapes or from isotope-enabled general circulation

35   models. Palynological analyses from these same Pacific sediment samples suggest no systematic

36   relationship between any particular type of pollen distribution and deviations from the global

37   calibration line. Rather, the poor correlations observed in the tropical Pacific are likely a function

38   of the small range of δ^2H$_P$ values relative to the typical residuals around the global calibration

39   line. Our results suggest that δ^2H$_{Wax}$ values are currently most suitable for use in detecting large

40   changes in precipitation in the tropical Pacific and elsewhere, but that ample room for improving

41   this threshold exits in both improved understanding of δ^2H variability in plants, as well as in

42   precipitation.

43

44   **Plain Language Summary**

45   Past precipitation patterns are difficult to reconstruct, limiting our ability to understand

46   Earth's climate system. Geochemists reconstruct past precipitation by measuring the amount of

47 heavy hydrogen naturally incorporated into the waxy coating of leaves, which is preserved in

48 mud that accumulates in lakes, soils, and oceans. Heavy hydrogen in leaf waxes is strongly

49 correlated with local precipitation, allowing us to learn about rainfall intensity, temperature, and

50 cloud movement. However, no existing calibration studies include sites from the tropical Pacific,

51 home to the most intense rainfall on the planet and populations that rely on rain for drinking

52 water. We measured heavy hydrogen in leaf waxes from tropical Pacific islands and show that

53 although values are within the global calibration error, no precipitation relationship exists within

54 the region. Plant type distributions do not explain the lack of correlation, which is best attributed

55 to poorly constrained estimates of heavy hydrogen in local rain and the relatively small range of

56 variability within the region. At present, heavy hydrogen from ancient leaf waxes can show large

57 changes in past precipitation, but improved process-level understanding is needed to use this tool

58 to understand smaller changes in the tropical Pacific and elsewhere.

59 **1 Introduction**

60   As Earth warms, precipitation intensity, frequency, and spatial distribution are expected to

61 change over the tropical Pacific (Brown et al., 2011; Tan et al., 2015; Sharmila et al., 2018).

62 These predictions need to be constrained and validated by robust reconstructions of past changes,

63 which are unfortunately limited in this region, in part because of a lack of proxies and archives

64 suitable for producing high resolution, continuous records (Hassall 2017). Existing high-

65 resolution paleohydrologic records have been established from speleothems (Partin et al., 2013;

66 Maupin et al., 2014) and corals (Quinn et al., 1993; Quinn et al., 1998; Hendy et al, 2002;

67 Linsley et al., 2004; Linsley et al., 2006; Calvo et al., 2007; DeLong et al., 2012), but are

68 generally limited to the past 600 years in this region. Lacustrine and swamp sediments can

69 provide longer records with much higher temporal resolution than is possible from slowly

70   accumulating marine sediments, and are well-established archives of ecological, anthropogenic,

71   and broad climatic changes in the region (Southern, 1986; Hope & Pask, 1998; Stevenson et al.,

72   2001; Prebble & Wilmshurst, 2009; Prebble et al., 2019; Gosling et al., 2020). More recently,

73   such sediments have also been used to reconstruct past hydroclimate change in the western

74   tropical Pacific at higher temporal resolution (Sachs et al., 2009; Smittenburg et al., 2011;

75   Konecky et al., 2016; Richey & Sachs, 2016; Hassall, 2017; Sachs et al., 2018; Sear et al., 2020).

76   One hydroclimate proxy suitable for tropical lake and swamp sediments in the tropical

77   Pacific is based on the hydrogen isotopic composition of leaf waxes ($\delta^2H_{Wax}=$

78    $(^2H/^1H)_{Wax}/(^2H/^1H)_{VSMOW} - 1$) (Sachse et al., 2012; Konecky et al., 2016; Hassall, 2017). δ^2H_{Wax}

79   values are highly correlated with hydrogen isotopes of mean annual precipitation (δ^2H_P) on a

80   global scale and have been applied to reconstruct δ^2H_P values in diverse locations (Sachse et al.,

81   2012; McFarlin et al., 2019). δ^2H_P values are related to specific physical processes, and are a

82   chemical signal that can both be transferred to material preserved on geologic timescales, as well

83   as modeled in modern systems with increasing accuracy, making their reconstructions useful for

84   understanding past hydroclimate dynamics (Bowen et al., 2019).

85   As is typical for organic geochemical proxies, the relationship between δ^2H_{Wax} and δ^2H_P

86   has been established through empirical calibrations with surface sediments from lakes and

87   surface soils. These calibration efforts began in Europe (e.g., Sachse et al., 2004; Leider et al.,

88   2013; Nelson et al., 2018) and have been extended to the Americas (e.g., Hou et al., 2008;

89   Polissar & Freeman, 2010; Douglas et al., 2013), East Asia and the Tibetan Plateau (e.g., Jia et

90   al., 2008; Aichner et al., 2010; Bai et al., 2011), and Africa (e.g., Peterse et al., 2009; Garcin et

91   al., 2012; Schwab et al, 2015) (Figure 1). Recent compilations of δ^2H_{Wax} from surface sediments

92   (McFarlin et al., 2019) and from sediments and soils (Liu & An, 2019) placed these local

93   calibrations in a global context. However, existing global compilations do not include any

94   tropical Pacific δ^2H$_{Wax}$ values.

95   Two considerations make it important to include tropical Pacific data in the global

96   calibration. Firstly, different vegetation types can influence net community ^{2}H/^{1}H fractionation

97   between precipitation and leaf waxes, which is not constant among plant types or environments

98   (Feakins and Sessions, 2010; Sachse et al., 2012; Kahmen et al., 2013). The unique plant

99   communities on tropical Pacific islands include many endemic species (Gillespie et al., 2013).

100   Additionally, coastal regions or former lagoons on these islands are often covered by mangrove

101   swamps, which consist of trees and shrubs adapted to brackish to hypersaline water. Due to

102   salinity effects, mangrove δ^2H$_{Wax}$ values may have the opposite response to changes in

103   precipitation intensity as nearby freshwater plants (Ladd & Sachs, 2012; He et al., 2017).

104   However, the impact of mangrove contributions to sedimentary δ^2H$_{Wax}$ values has not been

105   assessed.

106   Secondly, there is large uncertainty associated with estimates of δ^2H$_P$ in the tropical

107   Pacific. Direct measurements of δ^2H$_P$ from the Global Network of Isotopes in Precipitation

108   (GNIP) are spatially and temporally limited compared to other regions, resulting in large

109   uncertainties for statistical interpolations of δ^2H$_P$ such as those used for isoscape products and

110   the Online Isotopes in Precipitation Calculator (OIPC; Bowen & Revenaugh, 2003). Estimates of

111   δ^2H$_P$ from general circulation models (GCMs) in which precipitation isotopes have been

112   incorporated offer another potential calibration target for δ^2H$_{Wax}$ measurements in the modern

113   tropical Pacific that has not yet been explored.

114   Here we measured δ^2H values of seven *n*-alkane and five *n*-alkanoic acid homologues from

115   surface sediments collected from lakes influenced by precipitation from the South Pacific

116   Convergence Zone (SPCZ) and from mangrove swamps influenced by the Intertropical

117   Convergence Zone (ITCZ). We add new surface sediment δ^2H_{Wax} measurements of two of these

118   compounds (n-C$_{29}$-alkane and n-C$_{28}$-acid) from 19 lakes and four mangrove swamps on 15

119   islands distributed throughout the tropical Pacific to an updated global compilation of δ^2H_{Wax}

120   values. We assess whether δ^2H_{Wax} values from tropical Pacific lake and swamp sediments are

121   consistent with the global relationship between δ^2H_{Wax} and modeled δ^2H_P values from a diverse

122   set of algorithms and models. Finally, we use pollen-based vegetation reconstructions to evaluate

123   the influence of plant communities on tropical Pacific δ^2H_{Wax} values.

124   **2 Materials and Methods**

125   2.1 Site description and sample collection

126   Surface sediments were collected from 19 lakes on 11 islands across the SPCZ region

127   (Figure 1, Table 1), ranging in elevation above mean sea level from 790 m (Lanoto'o, Samoa) to

128   1 m (Rimatu'u, Oroatera, and Onetahi ponds on Tetiaroa, French Polynesia). Lakes ranged from

129   shallow ephemeral water bodies to an 88 m-deep volcanic crater lake (Lake Lalolalo, Wallis).

130   Most lakes were freshwater systems, except for the brackish (salinity = 17) coastal Lake

131   Dranoniveilomo (Fiji) and Lake Lalolalo (Wallis), which has a freshwater surface lens above

132   saline water (Sichrowsky et al., 2014). Mangrove trees surrounded Lake Dranoniveilomo, while

133   many other sites were located in forested regions, some impacted by human activity, particularly

134   horticulture. Aquatic vegetation covered the surface of some lakes (Table 1). Additional samples

135   were obtained from mangrove swamps located within the ITCZ throughout the Federated States

136   of Micronesia and Guam. All swamps were located at sea level and submerged at high tide. Four

137   or five surface sediment samples were collected from each swamp along a transect from the

138   inland edge to the coast.

139   Maloney et al. (2019) described the collection of most lake samples from the SPCZ region.

140   New samples include those from Lake Dranoniveilomo, which was cored in 2010 with a

141   Universal Percussion Corer (Aquatic Research, Hope ID, USA) fitted with a 6.6 cm diameter

142   polycarbonate core tube. Vesalea and Nopovois were cored in 2017 with a percussion corer

143   (UWITEC, Mondsee, Austria) equipped with a 6.3 cm diameter polycarbonate tube.

144   Unconsolidated upper sediment from these cores was subsampled at 1 cm intervals in the field

145   and stored frozen in Whirl-Pak plastic bags (Nasco, Fort Atkinson, WI, USA). Analyses of lake

146   surface sediment were restricted to the uppermost 1 or 2 cm of material. A hand trowel was used

147   to collect the upper 1 cm of mangrove sediments in 2012. Samples were stored frozen in Whirl-

148   Pak bags.

149   Water was collected from the surface of each lake and stored in screw-cap glass vials.

150   Additional water samples were collected from adjacent streams and long-term precipitation

151   integrators such as wells and rain cisterns when available. Samples were stored at 4 °C prior to

152   analysis.

153

154   2.2 Leaf wax extraction and purification

155   Maloney et al. (2019) described lipid extraction, saponification, and column

156   chromatography for all lake surface sediments except Dranoniveilomo, Vesalea, and Nopovois.

157   Samples from Dranoniveilomo were processed following the protocol of Maloney et al. (2019).

158   For all these lake sediments, the acid-containing fraction was eluted with 6 ml 4% acetic acid in

159   diethyl ether from an aminopropyl gel column and the alkane-containing fraction was eluted with

160   6 ml hexane from a silica gel column. Lipid extraction, saponification, and column

161   chromatography from Vesalea was described by Krentscher et al. (2019), and was identical for

162   the sample from Nopovois. Lipids from mangrove surface sediments were extracted and divided

163   into compound classes using Si gel column chromatography as in Ladd and Sachs (2017).

164   For mangrove surface sediments, *n*-alkanes were purified from a Si gel hexane fraction by

165   eluting 8 mL of 100% hexane over 0.5 g of $AgNO_3$-impregnated Si gel (10% by weight). For

166   lake samples, the alkane fraction was urea adducted to isolate unbranched compounds. Fatty

167   acids from lake sediments were methylated with 5% HCl in methanol for 12 hours at 70 °C, and

168   saturated fatty acid methyl esters (FAMEs) were isolated by elution in 8 mL of 4:1 Hex/DCM

169   over 0.5 g of $AgNO_3$-impregnated Si gel (10% by weight). Acid fractions were not analyzed

170   from mangrove surface sediments. Purity and concentrations of *n*-alkanes from mangrove

171   samples were assessed by gas chromatography – flame ionization detection (GC-FID) using the

172   GC program and instrumentation described in Ladd and Sachs (2017). For lake samples, *n*-

173   alkane and *n*-acid homologues were quantified using the same GC program and instrumentation

174   described in Ladd et al. (2018).

175

176   2.3 $\delta^2 H_{Wax}$ measurements

177   Samples were dissolved in hexane at a concentration suitable for hydrogen isotope

178   analyses of *n*-C_{29}-alkane or *n*-C_{28}-acid when those compounds were sufficiently abundant for

179   analysis by gas chromatography – isotope ratio mass spectrometry (GC-IRMS). $\delta^2 H$ values of

180   other baseline-resolved homologues with peak areas >15 Vs are also reported. For mangrove

181   sediment samples, GC-IRMS analyses were conducted with the same GC program and isotopic

182   referencing described in Ladd and Sachs (2017). Lake sediment samples were analyzed with the

183   same GC program and isotopic referencing described in Ladd et al. (2018). Phthalic acid of

184   known isotopic composition (Shimmelmann, Indiana University) was methylated to determine

185   δ^2H values of H added during methylation, which was corrected for using isotopic mass balance

186   (Lee et al., 2017).

187

188   2.4 Estimates of δ^2H$_P$ values

189   Estimates of δ^2H$_P$ values were extracted from different model products using latitude,

190   longitude, and elevation of each site. Model products included the Online Isotopes in

191   Precipitation Calculator (OIPC) version 3.2 (Bowen & Ravenaugh, 2003; IAEA/WMO, 2015;

192   Bowen, 2020), as well as isotope-enabled climate model contributions to the second Stable

193   Water Isotope Intercomparison Group (SWING2) from the CAM, ECHAM, GISS ModelE,

194   HadAM, isoGSM, LMDZ, and MIROC models (Sturm et al., 2010). OIPC estimated values were

195   obtained manually from the web interface ("OIPC mean annual δ^2H"), and also by extraction

196   from the high-resolution spatial gridded data set using a bilinear smooth function to

197   accommodate the proximity of a given location to neighboring pixels and the δ^2H values from

198   those pixels ("OIPC extracted mean annual δ^2H"). The multi-model mean annual precipitation

199   δ^2H value was calculated by averaging predicted values for all climate models that employed

200   spectral nudging (Yoshimura et al., 2008), which includes the ECHAM, GISS (nudged),

201   isoGSM, and LMDZ products.

202

203   2.5 Water δ^2H and δ^{18}O analyses

204   The isotopic composition of most lake and stream water samples were previously

205   analyzed and reported by Maloney et al. (2019). Water samples from the 2012 Micronesian field

206   campaign and from Lake Dranoniveilomo were analyzed by Cavity Ring Down Spectroscopy

207   (CRDS; Li-2130i, Picarro, Santa Clara, CA) using the same conditions and standards as in

208   Maloney et al. (2019). Additional water samples from the 2017 Vanuatu field campaign were

209   analyzed by Thermal Conversion/Elemental Analysis – Isotope Ratio Mass Spectrometry

210   (TC/EA-IRMS; ThermoFisher Scientific, Bremen, Germany) using the same conditions and

211   standards as in Newberry et al. (2017).

212

213   2.6 Pollen counts

214   Core samples for palynomorph analyses (including pollen and spores) were taken from

215   within the upper portion of the sediment core to determine modern baseline vegetation

216   differences among lakes. Each 1 cm^3 sample was processed using standard procedures (10%

217   HCl, hot 10% KOH, and acetolysis) (Moore et al. 1991). Samples were spiked with exotic

218   *Lycopodium clavatum* L. tablets to allow the palynomorph and charcoal concentrations to be

219   calculated. Counts continued until reaching at least 100 terrestrial palynomorphs. Reference

220   palynomorphs held in the Australasian Pollen and Spore Atlas (apsa.anu.edu.au/) assisted with

221   identification. The vegetation types (primary, secondary, dryland herbs, wetland herbs, etc.) were

222   determined from a regional synthesis of Pacific Island plant ecology (Mueller-Dombois &

223   Fosberg 1998).

224   **3 Results**

225   3.1 δ^2H_{Wax} values in the tropical Pacific

226   δ^2H_{Wax} values from surface sediments in the tropical Pacific were not correlated with mean

227   annual δ^2H_P values as calculated by the OIPC, nor with mean annual precipitation amount as

228   estimated by the Global Precipitation Climatology Project (GPCP) (Adler et al., 2003) (Figure

229   2). The only lipids with significant correlations with δ^2H_P values were dinosterol (data from

230   Maloney et al., 2019), n-C$_{16}$-acid, and n-C$_{18}$-acid, and the only significant correlations with the

231   amount of mean annual precipitation were dinosterol, n-C$_{18}$-acid, n-C$_{17}$-alkane, and n-C$_{33}$-alkane.

232 In almost all cases, correlation coefficients were negative for the relationship between $\delta^2 H_{Wax}$

233 and $\delta^2 H_P$ values, and positive for the relationship between $\delta^2 H_{Wax}$ values and mean annual

234 precipitation. An exception was n-C$_{17}$-alkane, which, similarly to dinosterol, had $\delta^2 H$ values that

235 are negatively correlated with mean annual precipitation (R = -0.95; p = 0.049) and positively

236 correlated with $\delta^2 H_P$ values (R = 0.95; p = 0.051) (Figure 2). However, n-C$_{17}$-alkane was only

237 abundant enough to measure its $\delta^2 H$ values in four samples, making any assessment of these

238 correlations tentative.

239

240 3.2 Tropical Pacific $\delta^2 H$ values in the global context

241   Tropical Pacific $\delta^2 H$ values of n-C$_{29}$-alkanes and n-C$_{28}$-acids (the most commonly

242 measured leaf waxes in the literature) were in the range expected based on the global relationship

243 between $\delta^2 H_{Wax}$ and $\delta^2 H_P$ values (Figure 3). Tropical Pacific n-C$_{29}$-alkane $\delta^2 H$ values ranged

244 from −177 to −139‰, while those of n-C$_{28}$-acid ranged from −175 to −119‰ (Table 1; Figure

245 3). Adding these new measurements to an updated global compilation of $\delta^2 H_{Wax}$ values from all

246 available surface sediment and soil data sets in non-marine settings (compilations from Liu &

247 An, 2019 and McFarlin et al., 2019, as well as data sets from Nelson, 2013; Bakkelund et al.,

248 2018; Feng et al., 2019; Goldsmith et al., 2019; Li et al., 2019; Wu et al., 2019; Lu et al., 2020;

249 Struck et al., 2020; van der Veen et al., 2020) has minimal impact on the slope, y-intercept, or

250 correlation coefficients for the global linear regression (Figure 3).

251   To compare new measurements from the tropical Pacific to the relationship defined by

252 previously published values, we calculated their residual values from the global linear regression

253 between $\delta^2 H_{Wax}$ and $\delta^2 H_P$ (excluding new tropical Pacific data). At most tropical Pacific

254 locations, residuals from the global linear regression line were within ± 20‰, but were greater

255 than this at 5 sites for n-C$_{29}$-alkane and 6 sites for n-C$_{28}$-acid (Table 1). Only two sites (Lake

256   Tagamaucia in Fiji and White Lake in Vanuatu) had residuals greater than 20‰ for both

257   compounds.

258   We also contextualized variability in the tropical Pacific data relative to the global data set

259   by randomly subsampling 17 values from the compiled data 4,000 times and comparing the

260   correlation coefficient between δ^2H_{Wax} and δ^2H_P to the range in δ^2H_P values (Figure 4). None of

261   these subsampled data sets had ranges in δ^2H_P values that were as small as the range in the

262   tropical Pacific (smallest range for n-C$_{29}$-alkane = 59‰, for n-C$_{28}$-acid = 118‰), so we

263   subsampled the compiled data again while restricting the maximum range to 100‰ (1,000

264   iterations each for the highest, lowest, and middle δ^2H_P values), 50‰ (200 iterations for each

265   possible 50‰ range with maximum δ^2H_P values shifted by 10‰), and 35‰ (100 iterations for

266   each possible 35‰ range with maximum δ^2H_P values shifted by 5‰). Correlation coefficients

267   were typically high (>0.5) when the range of δ^2H_P values was greater than 100‰, and became

268   increasingly scattered below this threshold (Figure 4). The relationship between the correlation

269   coefficient and the range of δ^2H_P values in the tropical Pacific plotted within the range generated

270   by random subsets. However, tropical Pacific correlation coefficients for both compounds were

271   more than one standard deviation below the mean value of random sample sets with a δ^2H_P range

272   between 30 and 35‰ (0.43 ± 0.28 for n-C$_{29}$-alkane; 0.22 ± 0.41 for n-C$_{28}$-acid) (Figure 4).

273   In addition to the OIPC, several water-isotope-enabled GCMs also provide estimates of

274   mean annual δ^2H_P values. We extracted δ^2H_P values for all sites with surface sediment or soil

275   δ^2H_{Wax} values from each climate model included in the Stable Water Isotope Intercomparison

276   Group, Phase 2 (SWING 2) model comparison. For all models, global n-C$_{29}$-alkane and n-C$_{28}$-

277   acid δ^2H values were positively correlated with δ^2H_P estimates (Figure 5). For n-C$_{29}$-alkane,

278   δ^2H_{Wax} values were most highly correlated with δ^2H_P values obtained manually from the OIPC

279   (R = 0.86). The lowest correlation was with δ^2H_P values from HadAM (R = 0.57) (Figure 5). For

280   *n*-C$_{28}$-acid, global δ^{2}H$_{Wax}$ values were most highly correlated with δ^{2}H$_P$ values extracted from the

281   CAM model (R = 0.91). The lowest correlation was with δ^{2}H$_P$ values extracted from the high-

282   resolution spatial gridded OIPC data (R = 0.82) (Figure 5). The correlation coefficient obtained

283   manually from the OIPC (R = 0.86) was intermediate among the different models (Figure 5).

284

285   3.3 Pollen and spore spectra

286   Palynomorphs from most sites were indicative of human disturbance to the catchment

287   vegetation, as the dominant pollen types are from secondary forest taxa (Figure 6; Table 2).

288   When secondary forest vegetation was not most abundant, fern spores contributed more to the

289   palynomorph sum than any other plant group, except at Lake Hut, where primary forest taxa

290   were most abundant (Figure 6; Table 2). Although wetland plants covered more than 50% of the

291   surface water at three lakes (Onetahi Pond, Lake Tagamucia, and Veselea Pond), wetland herbs

292   and aquatic plants never contributed more than 23% of the observed pollen.

293   Pollen data are only available for three of the five sites where *n*-C$_{29}$-alkane residuals from

294   the global δ^{2}H$_{Wax}$ vs. δ^{2}H$_P$ relationship were less than −20‰ (Figure 6). In two of these

295   (Tagamaucia and White Lake), fern spores were abundant (59% and 39%, respectively) (Figure

296   6; Table 2). However, at the third site with an *n*-C$_{29}$-alkane residual less than −20‰ (Lake Hut),

297   fern spore concentrations were low and primary forest taxa palynomorphs were most abundant

298   (Figure 6; Table 2). Three sites had *n*-C$_{28}$-acid residuals from the global δ^{2}H$_{Wax}$ vs. δ^{2}H$_P$

299   relationship less than −20‰, of which two have pollen data (Tagamaucia and Dranoniveilomo).

300   Each of these had a high abundance of ferns and wetland plants (80% and 37%, respectively)

301   (Figure 6, Table 2). Three sites had *n*-C$_{28}$-acid residuals from the global δ^{2}H$_{Wax}$ vs. δ^{2}H$_P$ relations

302   that were greater than 20‰ (Figure 6). One of these, White Lake, had relatively high

303   contributions from ferns. The second, Harai Lake #1, does not have recent pollen data (the most

304   recent pollen sample is from 11 – 12 cm), but historically had high contributions from ferns

305   (Table 2). The third, Nopovois, is dominated by secondary forest vegetation (54%) and does not

306   have palynological features that clearly distinguish it from sites where n-C$_{28}$-acid δ^2H values

307   adhere more closely to the global relationship (Figure 6; Table 2). Additionally, some sites with

308   high contributions from ferns and wetland plants – Harai Lake #3, Lanoto'o, and Lake Otas –

309   have δ^2H$_{Wax}$ values close to the global relationship (Figure 6).

4 Discussion

310

311   Although the relationship between δ^2H$_{Wax}$ and δ^2H$_P$ values lacks any correlation for the

312   tropical Pacific sites in isolation, the values fall within the global scatter around the linear

313   regression of compiled literature values from surface sediments and soils (Figure 3; Table 1). In

314   addition to adding recently published data, our global data set differs from two recent

315   compilations by excluding marine sediments (in contrast to Liu and An, 2019), and including

316   both soils and surface sediments (in contrast to McFarlin et al., 2019). Although leaf waxes in

317   soils and sediments might have different sources and represent different timescales and

318   catchment areas, there is no significant difference in the relationship between δ^2H$_{Wax}$ and δ^2H$_P$

319   for either compound (Figure 3c and 3d). The similarity between the soil and sediment

320   compilations suggests that the transit history of leaf waxes from plant to deposition and

321   subsequent preservation varies as much within archive type as it does between them.

322   The positive linear relationship between δ^2H$_{Wax}$ and δ^2H$_P$ values in the global compilation

323   remains robust, with R^2 values of 0.74 for both n-C$_{29}$-alkane (n = 665) and n-C$_{28}$-acid (n = 242)

324   (Figure 3). However, considerable scatter around the regression line exists globally and within

325   the tropical Pacific. Large residuals are due to both uncertainty in the y-axis (variable ^{2}H/^{1}H

326   fractionation between leaf waxes and water among plant types and environments, discussed in

327   section 4.1) and in the x-axis (mean annual δ^2H_P values and the water source used by plants,

328   discussed in section 4.2).

329

330   4.1 Variable hydrogen isotope fractionation during leaf wax synthesis

331   Although global δ^2H_{Wax} values are well correlated with δ^2H_P values of mean annual

332   precipitation (Figures 3, 5), several well-established factors contribute to variability in the net

333   $^2H/^1H$ fractionation between plant waxes and precipitation (α_{Wax-P}) (Sachse et al., 2012).

334   Variations in α_{Wax-P} occur among plant functional types (Liu et al., 2006), between leaves and

335   other plant organs (Gamarra & Kahmen, 2015), and with relative humidity (Tipple et al., 2015).

336   Additionally, biosynthetic fractionation between leaf water and waxes can vary seasonally

337   (Newberry et al., 2015), with environmental stresses (Ladd & Sachs, 2015), and with changes in

338   plant metabolism (Cormier et al., 2018). Large differences in α_{Wax-P} can also exist among plant

339   species growing at the same site (Feakins & Sessions, 2010; Sachse et al., 2012; Eley et al.,

340   2014; He et al., 2020). With the current tropical Pacific data set we can only examine factors that

341   might relate to differences among different types of plants, and not factors that can occur within

342   a single plant, such as metabolic state. We examine three plant groups – mangroves, aquatic

343   plants, and ferns –whose contributions may impact community δ^2H_{Wax} values. By comparing

344   pollen distributions and information about surrounding vegetation at each site with the residuals

345   between δ^2H_{Wax} values and the global calibration line (Figure 6), we demonstrate that changes in

346   vegetation inferred from pollen cannot consistently explain anomalous δ^2H_{Wax} values in tropical

347   Pacific surface sediments.

348

349   4.1.1 Mangroves

15

350   Mangroves are woody plants that grow in brackish to hypersaline water. They contribute

351   large amounts of organic matter to coastal sediments in the tropics and subtropics (Alongi,

352   2014). Because mangroves discriminate more against ^{2}H as salinity increases (Ladd & Sachs,

353   2012; He et al., 2017; Ladd & Sachs, 2017), they should have lower δ^2H_{Wax} values than nearby

354   freshwater plants. This relationship was recently observed in the Florida Everglades, where

355   mangroves have δ^2H_{Wax} values ~50‰ lighter than those from nearby freshwater trees, despite

356   equivalent δ^2H_P values (He et al., 2020). Significant contributions of mangrove leaf waxes in

357   coastal areas in the tropical Pacific could result in sedimentary δ^2H_{Wax} values that fall below the

358   global calibration line.

359   Several of the lakes in our calibration set were located in coastal areas, but only one,

360   Dranoniveilomo, had brackish water and mangroves growing directly in its periphery. In this

361   lake n-C_{28}-acid is significantly depleted relative to the global calibration, but n-C_{29}-alkane is not

362   (Table 1). Despite the abundant mangroves around Dranoniveilomo, barely any mangrove pollen

363   was found in the sediment, which may reflect different transport mechanisms and catchment

364   areas for leaf waxes and pollen (Table 2). Two coastal lakes in Vanuatu (Otas and Waérowa

365   East) have the highest amounts of mangrove pollen observed in all examined surface sediments

366   (~15%; Figure 6; Table 2). In Lake Otas, δ^2H_{Wax} values are close to the values predicted by the

367   global relationship (Table 1). In Lake Waérowa East, they are slightly higher than expected

368   (Table 1), opposite to the expected impact of significant mangrove leaf wax contributions. These

369   data suggest that mangrove leaf waxes are not an important influence on δ^2H_{Wax} values in

370   tropical Pacific lake sediments included in this study.

371   Likewise, Micronesian mangrove swamp surface sediments had δ^2H_{Wax} values that were

372   consistent with the global linear regression (Table 1; Figure 3). Additionally, there was little

373   spatial variability in δ^2H_{Wax} values throughout each individual mangrove swamp, with 5‰

374   standard deviations among samples within a single swamp (Table 1). This homogeneity occurred

375   even though samples were collected from sites with surface water salinity ranging from 0-31 at

376   the time of collection. Surface water salinity was dynamic throughout these mangrove swamp

377   surveys, varying temporally and spatially with tides and rain events. Individual mangrove trees

378   with large root networks therefore had access to water with a wide range of salinities and may

379   have opportunistically taken up relatively fresh water that was ultimately used in leaf wax

380   synthesis. Preferential uptake of fresher water by mangroves has been observed previously

381   (Santini et al., 2015; Reef et al., 2015), and could result in all mangroves throughout a swamp

382   using water with similar salinity and isotopic composition, consistent with the surface sediment

383   δ^2H_{Wax} values observed in transects from Micronesian mangrove swamps.

384

385   4.1.2 Aquatic plants

386   Some of the lakes included in the tropical Pacific survey were partially or completely

387   covered by floating aquatic vegetation (Table 1). Since aquatic plants at diverse sites tend to

388   have lower alkane δ^2H_{Wax} values than nearby terrestrial plants (Chikaraishi & Naraoka, 2003;

389   Gao et al., 2011; Dion-Kirschner et al., 2020; He et al., 2020), differing relative contributions of

390   leaf waxes from aquatic plants could also reduce sedimentary δ^2H_{Wax} values. There are a few

391   reasons why aquatic plants may have relatively low δ^2H_{Wax} values. First, when a lake is mostly

392   covered by water lilies or similar aquatic vegetation, there is a physical barrier to evaporation of

393   lake water, and it may therefore maintain a δ^2H signal similar to that of precipitation, rather than

394   becoming enriched in ^2H due to transpiration, as is the case for leaves exposed to air (Kahmen et

395   al., 2013; Cernusak et al., 2016). Second, it is possible that aquatic plants may exhibit greater

396   biosynthetic fractionation between leaf water and leaf lipid. However, existing investigations of

397    $\delta^2 H_{Wax}$ values in submerged aquatic plants suggest this is only likely at high salinity, while plants

398   grown in freshwater display similar α_{Wax-P} values to other plants (Aichner et al, 2017).

399   High contributions from aquatic plants could explain why $\delta^2 H_{Wax}$ values at Tagamaucia in

400   Fiji, which is covered in floating sedge islands (Southern et al., 1986), were very [2]H-depleted

401   relative to the global calibration line (Table 1). However, this relationship was not consistent in

402   all lakes covered by aquatic plants. For example, Lake Veselea in Vanuatu is completely covered

403   by mats of aquatic plants (primarily *Persicaria* cf. *attenuata*, *Salvinia molesta*, and *Calystegia*

404   *soldanella*), yet $\delta^2 H_{Wax}$ values from its sediment fell close to the global calibration line (Table 1).

405   Additionally, pollen from wetland herbs and aquatic plants is not consistently associated with

406   large or small residuals from the global calibration line (Figure 6). Aquatic plants may have

407   minimal influence on sedimentary $\delta^2 H_{Wax}$ values because submerged plants are not at risk of

408   desiccation and therefore have little need for the moisture barrier provided by long-chain leaf

409   waxes. They therefore tend to have low concentrations of these compounds (Ficken et al., 2000;

410   Dion-Kirschner et al., 2020; He et al., 2020). Overall, our results suggest that increased presence

411   of aquatic plants does not unequivocally result in decreased $\delta^2 H_{Wax}$ values in tropical Pacific lake

412   sediments.

413

414   4.1.3 Ferns

415   Assessments of $\delta^2 H_{Wax}$ values from ferns are limited, but previous studies show that ferns

416   have similar α_{Wax-P} values to many other plant taxa, including lycopods, gymnosperms, eudicots,

417   and magnoliids (Gao et al., 2014). We therefore had no expectation that sites with large

418   sedimentary contributions of leaf waxes from ferns would diverge from the global $\delta^2 H_{Wax}$-$\delta^2 H_P$

419   linear regression. However, some of the sites with the largest residuals relative to the global

420   regression line had palynomorph spectra characterized by large contributions of fern spores. This

421 was particularly true at Lake Tagamaucia in Fiji and White Lake in Vanuatu, and to a lesser

422 extent in Fiji's Lake Dranoniveilomo (Table 1; Figure 6). However, high accumulation of fern

423 spores did not universally correspond to low δ^2H_{Wax} values, for example at Lake Lanoto'o in

424 Samoa and Harai Lakes #1 and #3 in the Solomon Islands (Tables 1 and 3; Figure 6). Of these,

425 n-C_{28}-acid δ^2H values from the Harai Lakes were much higher than the predicted values from the

426 global regression fit, in direct contrast to the Fijian lakes and White Lake (Table 1; Figure 6).

427 Additionally, Lake Hut in New Caledonia had the largest n-C_{29}-alkane residual for any site with

428 pollen data, but did not have much fern pollen (Figure 6). Overall, this suggests that relative

429 contributions of leaf waxes from ferns do not have predictable effects on sedimentary δ^2H_{Wax}

430 values. Nevertheless, a shift in δ^2H_{Wax} values that coincides with a change in the relative

431 abundance of fern spores in a down-core record may indicate a change in organic matter sources

432 rather than a change in δ^2H_P values.

433

434 4.2 Uncertainty in δ^2H_P values

435   Variability in α_{Wax-P} contributes to uncertainty in the y-axis in the relationship between

436 δ^2H_{Wax} and δ^2H_P (Figure 3). However, unlike most proxy systems, the x-axis calibration target is

437 poorly constrained and likely accounts for a large portion of the linear regression residuals

438 between δ^2H_{Wax} and δ^2H_P values in the tropical Pacific. δ^2H_P values are not constant throughout

439 the year, and water from different seasons has different residence times in soil, meaning that the

440 δ^2H values of water used by plants is typically not equal to mean annual δ^2H_P values (Brinkmann

441 et al., 2018). Additionally, δ^2H_P values from the OIPC represent climatological means, but δ^2H_P

442 values vary interannually and seasonally. The time period captured by a surface sediment sample

443 (typically a few years to two decades; see Maloney et al., 2019 for sediment accumulation rates

444 at most sites) may differ considerably from the long-term mean. Finally, the robustness of mean

445   annual estimates provided by the OIPC varies spatially among geographic settings due to the

446   uneven density of $\delta^2 H_P$ observations (IAEA/WMO, 2015).

447   Limited $\delta^2 H_P$ data from some sites in the tropical Pacific mean that $\delta^2 H_P$ values calculated

448   using OIPC have large uncertainties (Table 1). This is especially problematic for sites in the

449   southeastern portion of the region, since there are no observations of $\delta^2 H_P$ values from the

450   Solomon Islands, Vanuatu, or New Caledonia in the GNIP database. 95% confidence intervals

451   for $\delta^2 H_P$ values from sites in Vanuatu are as large as 76‰, considerably larger than the overall

452   range in $\delta^2 H_P$ values (~30‰) and the range in measured $\delta^2 H_{Wax}$ values (~40‰ for n-C_{29}-alkane

453   and ~55‰ for n-C_{28}-acid) in the Pacific (Table 1; Figure 3). For other sites, such as those in

454   Micronesia, OIPC uncertainties may add false confidence to predicted values, as these can be

455   based on a limited number of GNIP observations from several decades ago. For example, the

456   Yap GNIP station has 65 observations collected from 1968 to 1976, and the station on Chuuk

457   (Truk) has 72 observations collected between 1968 and 1977 (IAEA/WMO, 2015).

458   Climatological means calculated from these data may not be representative of conditions when

459   surface sediment leaf waxes in this study formed. This uncertainty in the independent variable

460   likely contributes to the lack of a regional correlation between $\delta^2 H_{Wax}$ and $\delta^2 H_P$ values (Figure 3).

461   An alternative approach for estimating mean annual $\delta^2 H_P$ values is to use water-isotope-

462   enabled GCMs (Sturm et al., 2010; Conroy et al., 2013; Steen-Larsen et al., 2017). These

463   estimates of $\delta^2 H_P$ values have not typically been used in $\delta^2 H_{Wax}$ calibration studies. Because

464   some of the western Pacific $\delta^2 H_P$ values derived from OIPC had such large uncertainties, we

465   assessed whether scatter in the global relationship between $\delta^2 H_{Wax}$ and $\delta^2 H_P$ could be reduced by

466   using isotope-enabled GCMs to estimate $\delta^2 H_P$ values (Figure 5).

467   In general, δ^2H_{Wax} values from *n*-C$_{28}$-acid are better correlated with δ^2H_P values generated

468   by isotope-enabled GCMs than δ^2H_{Wax} values from *n*-C$_{29}$-alkane. This difference may be due to

469   the distinct spatial distributions of the two calibration data sets, with fatty acid δ^2H values almost

470   exclusively limited to North America (Figure 1). Sites from which *n*-alkane δ^2H values are

471   available are more numerous, more globally distributed, and include many measurements from

472   the Himalayas and Tibetan Plateau (Figure 1). Here, steep elevation gradients may make δ^2H_P

473   vary on spatial scales that are smaller than the resolution of most models, and fluvial and aeolian

474   processes may transport waxes between regions with distinct δ^2H_P values. Overall, our analysis

475   does not suggest any structural limitations to using the OIPC to estimate δ^2H_P values for proxy

476   calibration. However, the limited δ^2H_P measurements from the tropical Pacific and resulting large

477   uncertainties in modern estimates from the OIPC or isotope-enabled GCMs remains a

478   considerable challenge for assessing the fidelity of δ^2H_{Wax} values in this region.

479   In particular, the orographic effects of mountainous islands may not be adequately captured

480   by either the OIPC or isotope-enabled GCMs, causing particular challenges determining the

481   appropriate calibration target for water isotope proxies. For example, the highest peaks on the

482   island of Espiritu Santo in Vanuatu are nearly 2000 m above sea level. With prevailing winds

483   from the east, the west coast sits in the rain shadow of the mountains and is significantly drier

484   than the eastern part of the island (Terry, 2011). However, the only long-term weather station for

485   the island is located at Pekoa airport in the southeast corner of the island, and there are no local

486   GNIP stations. Therefore, minimal local data is available to inform precipitation isotope models.

487   The OIPC predicts equivalent δ^2H_P values for sites on opposite coasts of Espiritu Santo, and

488   predicted δ^2H_P values from water-isotope-enabled GCMs on each coast are within a few ‰ of

489   each other. However, the leeward western sites should have lighter δ^2H_P values than the eastern

490   sites that receive rain from more marine air masses (Scholl et al., 1996, 2007). This expectation

491   is supported by the δ^2H values of lake and stream waters collected during our field campaigns,

492   which are depleted by 20 – 40 ‰ on the leeward western side of the island compared to the

493   windward east coast (Data Set S2).

494

495   4.3 Contrasts with δ^2H values of non-leaf wax lipids

496   In addition to the longer chain *n*-alkane and *n*-alkanoic acids that are primarily derived

497   from higher plant waxes, we also measured δ^2H values from several compounds of mixed or

498   primarily algal sources. These compounds are typically found in sedimentary records along with

499   leaf waxes, and the different controls on their δ^2H values offer the opportunity to more

500   completely resolve sources of down-core variability in δ^2H$_{Wax}$ values. Here we discuss lipids

501   from algal sources, and ubiquitous compounds produced by most organisms.

502

503   4.3.1 Algal lipids

504   Unlike δ^2H$_{Wax}$ values, δ^2H values of algal biomarkers are well correlated with tropical

505   Pacific δ^2H$_P$ values and mean annual precipitation (Figure 2). This is particularly the case for

506   dinosterol (Maloney et al., 2019), which is primarily produced by dinoflagellates (Volkman,

507   2003). The δ^2H values of *n*-C$_{17}$-alkane, which is primarily derived from algae (Cranwell et al.,

508   1987; Meyers, 2003), were also highly correlated with δ^2H$_P$ values (consistent with Sachse et al.,

509   2004) and inversely correlated with mean annual precipitation (Figure 2). An inverse correlation

510   between the amount of mean annual precipitation and δ^2H$_P$ values (and therefore δ^2H values of

511   lipids that track δ^2H$_P$) is expected in a low-latitude maritime regions where the amount effect

512   plays a strong role in determining the isotopic composition of rain (Dansgaard, 1964; Rozanski

513   et al., 1993; Kurita et al., 2009).

514   One reason why δ^2H values from the algal biomarkers are better correlated with δ^2H_P

515   values than δ^2H_{Wax} values might relate to the source water used by each type of organism. Leaf

516   waxes from higher plants growing on land may reflect a temporal bias, as monthly OIPC δ^2H_P

517   values can differ by up to 40‰ at the tropical Pacific sites. Higher plants primarily produce leaf

518   waxes soon after setting new leaves, meaning that there may be a seasonal bias in the δ^2H_P signal

519   that is transferred to their waxes (Tipple et al., 2013; Freimuth et al., 2017). If algae are

520   productive throughout the year, they may better integrate annual precipitation, therefore resulting

521   in algal lipids that more closely track mean annual δ^2H_P values.

522   Another reason why algal biomarkers may track δ^2H_P better than leaf waxes is that they

523   come from a more limited range of potential sources. In addition to the range of plant sources for

524   leaf waxes discussed in section 4.1, many of the mid-chain and relatively long-chained

525   acetogenic compounds, including n-C_{29}-alkane and n-C_{28}-acid, can be derived from a mix of

526   terrestrial and aquatic plants (Bush & McInerney, 2013; Andrae et al., 2020; Dion-Kirschner et

527   al., 2020). n-C_{28}-acid can also be partially derived from microalgal sources (Volkman, 1980; van

528   Bree et al., 2018). Therefore the leaf waxes may represent variable aquatic and terrestrial

529   contributions, while the algal compounds are always aquatically sourced.

530   Finally, the spatial variability integrated by each type of compound could explain the

531   different trends in their δ^2H_{Wax} values. Algal lipids are produced within the relatively confined

532   space of the lake or pond overlaying the sediments in which they accumulate. Leaf waxes can be

533   derived from plants growing adjacent to their depocenter, but also from further afield in the

534   catchment, and the relative size of the catchment area can differ among water bodies.

535   Additionally, > 20% of leaf waxes accumulating in sediment can come from aerosols, which can

536   be transported long distances and have δ^2H values distinct from local vegetation (Conte et al.,

537   2003; Gao et al., 2014; Nelson et al., 2017; Nelson et al., 2018). Leaf wax aerosols from very

23

538   distant sites may have more impact on lake sediments on islands than on continents, since there

539   is a relatively smaller contiguous land area to contribute regional and local waxes. On the other

540   hand, the overall contribution of local leaf waxes may be significantly higher on small islands

541   where all non-local leaf waxes must be carried great distances.

542

543   4.3.2 Generic fatty acids

544   In contrast to the algal specific biomarkers dinosterol and n-C$_{17}$-alkane, δ^2H values of n-

545   C$_{16}$ and n-C$_{18}$ fatty acids were positively correlated with mean annual precipitation and

546   negatively correlated with δ^2H$_P$ (Figure 2). These shorter chain fatty acids are synthesized by

547   most organisms, but are frequently attributed to algal sources in aquatic sediments (Huang et al.,

548   2004; Li et al. 2009). Heterotrophic and chemoautotrophic microbes produce short-chain fatty

549   acids that can have δ^2H values that differ by several hundred ‰ from those of photoautotrophs

550   grown in similar water (Zhang et al., 2009; Heinzelmann et al., 2015). However, other than in

551   microbial mats (Osburn et al., 2011), sedimentary n-C$_{16}$ and n-C$_{18}$ fatty acids typically have

552   fractionation factors consistent with values from photoautotrophs in culture (Li et al, 2009;

553   Zhang et al., 2009; Heinzelmann et al., 2018). In our tropical Pacific data set, fractionation

554   factors between lake water and n-C$_{16}$ and n-C$_{18}$-acids ($\alpha_{\text{Lipid-Water}} = (^2\text{H}/^1\text{H})_{\text{Lipid}}/ (^2\text{H}/^1\text{H})_{\text{Water}}$)

555   ranged from 0.773 to 0.920. This large range in $\alpha_{\text{Lipid-Water}}$ values is consistent with observations

556   of cultures of different types of algae (Zhang and Sachs, 2007; Zhang et al., 2009; Heinzelmann

557   et al., 2015). The δ^2H values of the n-C$_{16}$ and n-C$_{18}$-acids in our data set could be influenced by

558   variable contributions from non-photoautotrophs, but could also vary due to differing

559   contributions from different types of algae. In either case, it seems likely that δ^2H values of these

560   compounds reflect ecology more than hydroclimate, and their negative correlations with δ^2H$_P$

561   values in our sample set may be a coincidence. Dinosterol and n-C$_{17}$-alkane are sourced from a

562   smaller range of organisms than the near ubiquitous *n*-C$_{16}$ and *n*-C$_{18}$-acids (Cranwell et al., 1987;

563   Meyers, 2003; Volkman, 2003), which could make their δ^{2}H values more directly related to

564   those of lake water.

565

566   4.4 Implications for paleoclimate reconstructions in the tropical Pacific

567   Although δ^{2}H$_{Wax}$ values are strongly linearly correlated with δ^{2}H$_P$ values on a global scale

568   (Figure 3), the large residuals in this relationship indicate that caution should be applied before

569   interpreting relatively small down-core changes in δ^{2}H$_{Wax}$ values as hydroclimate changes.

570   However, our data do not suggest that there are clear links between vegetation source (as

571   indicated by palynological analyses) and residuals from the global δ^{2}H$_{Wax}$-δ^{2}H$_P$ relationship

572   (Figure 6). Rather, one of the largest challenges for interpreting sedimentary δ^{2}H$_{Wax}$ values in the

573   tropical Pacific are uncertainties associated with modern estimates of δ^{2}H$_P$ values, given the

574   limited spatial and temporal available of modern observations. Recent isotope modeling work

575   has helped constrain the processes that control δ^{2}H$_P$ values in this dynamically important region

576   (Conroy et al., 2016; Konecky et al., 2019). Continued effort in this regard is necessary to

577   robustly interpret proxies δ^{2}H$_P$ values, whether they are derived from δ^{2}H$_{Wax}$ values or from

578   other archives such as speleothems.

579   The expected inverse correlation between precipitation amount and OIPC-derived δ^{2}H$_P$

580   values in the tropical Pacific, and the correlations between these variables and algal lipid δ^{2}H

581   values (Maloney et al., 2019), suggest that the uncertainty in δ^{2}H$_P$ values cannot be solely

582   responsible for the poor correlations associated with δ^{2}H$_{Wax}$ values (Figure 2). Rather, factors

583   besides δ^{2}H$_P$ values that influence δ^{2}H$_{Wax}$ values (variations in seasonality, catchment scales,

584   contributions from organs other than leaves and/or from aquatic sources, or changes in

585   biosynthetic fractionation) result in residuals that are on the order of ±25‰ in the tropical Pacific

586 and elsewhere (Figure 3). Any calibration of δ^2H_{Wax} values spanning a relatively small range of

587 δ^2H_P values (like the 32‰ range studied here) is likely to have a poor correlation between the

588 two variables (Figure 4). Correlation coefficients for the tropical Pacific are within the

589 distribution generated by randomly subsampling the global data set while limiting the range in

590 δ^2H_P to 35‰, but fall towards the low end of this range, more than 1 standard deviation below

591 the mean correlation coefficient (Figure 4).

592   Regional calibrations are expected to have stronger than average correlations by

593 constraining some variables that contribute to scatter in the global relationship between δ^2H_P and

594 δ^2H_{Wax} values. However, the new data from the tropical Pacific exceed a regional scale in many

595 ways, spanning a distance of ~8500 km, larger than even continental-scale studies (e.g., Sachse

596 et al., 2004). The islands included range from low-lying atolls to mountainous volcanoes. The

597 sites differ in seasonality of precipitation, vulnerability to tropical storms, sensitivity to El

598 Niño−Southern Oscillation events, and biodiversity, all of which may impact δ^2H_{Wax} values. The

599 diversity of sites, uncertainty in local estimates of δ^2H_P values, and small δ^2H_P signal relative to

600 the noise in the global calibration, make it unsurprising that δ^2H_{Wax} values are not correlated with

601 δ^2H_P values within the tropical Pacific. However, it is encouraging that tropical Pacific δ^2H_{Wax}

602 values fall within the expected range, and do not have abnormally large residuals from the global

603 calibration line. Together, these results suggest that the processes determining δ^2H_{Wax} values in

604 tropical Pacific lake and swamp sediments are not fundamentally different than elsewhere.

605   The scatter in the global relationship between δ^2H_P and δ^2H_{Wax} (Figure 3) suggests that

606 down-core changes as large as ~50‰ at any location could be driven by factors other than δ^2H_P

607 values. In practice, at a single site where many variables that contribute to scatter among sites are

608 constant, the threshold for detecting changes in δ^2H_P values may be significantly smaller.

609 Detailed processed-based studies at the catchment scale (Freimuth et al., 2019, 2020; Dion-

610   Kirschner et al., 2020) may be more useful for constraining sedimentary δ^2H_{Wax} than additional

611   large-scale calibration efforts.

612   Hydroclimate-driven interpretations of changes in sedimentary δ^2H_{Wax} values will be

613   most robust when supported by independent lines of proxy evidence, such as δ^2H values of more

614   source specific biomarkers like dinosterol (Smittenberg et al., 2011; Nelson & Sachs, 2016;

615   Richey & Sachs, 2016; Sachs et al., 2018), changes in grain size distributions (Conroy et al.,

616   2008), or changes in the elemental composition of inorganic sediments (Sear et al 2020; Higley

617   et al., 2018). Continued refinement of a multi-proxy toolbox that includes sedimentary δ^2H_{Wax}

618   values offers the best path to confidently reconstructing past hydrologic change.

619   **5 Conclusions**

620   δ^2H_{Wax} values from surface sediments from 19 lakes and four swamps on 15 islands

621   throughout the tropical Pacific fall within the overall range of values expected based on a global

622   compilation surface sediment measurements ($R^2 = 0.74$ for both *n*-C$_{29}$-alkane (n = 665) and *n*-

623   C$_{28}$-acid (n = 242)), and the residuals around the global linear regression between δ^2H_{Wax} and

624   δ^2H_P are similar in the tropical Pacific and global data sets. Nevertheless, within the tropical

625   Pacific there is no significant correlation between δ^2H_{Wax} and δ^2H_P values. The lack of correlation

626   regionally is at least partly due to the large uncertainties in δ^2H_P values derived from reanalysis

627   data and cannot be ascribed to different vegetation sources within and surrounding the lakes in

628   this study, as deduced from pollen assemblages.

629   To a first order on a global scale, δ^2H_{Wax} values are clearly influenced by δ^2H_P values, but

630   the δ^2H_P signal spanning the tropical Pacific remains small relative to the noise in the current

631   global calibration. The global δ^2H_{Wax-P} calibration remains limited by uncertainties in both the x-

632   and y-axes, and could be improved by better constraints on δ^2H_P values. As in other locations,

633   large changes in δ^2H_{Wax} in sediments from tropical Pacific islands may be caused by variables

634   other than δ^2H_P, and could be improved by more catchment-scale, processed-based studies. In

635   particular, interpretations need to consider the possible effects of changing source, growth

636   conditions, and delivery of leaf waxes to sediments. When possible, δ^2H_{Wax} values should be

637   paired with δ^2H values of more source-specific compounds such as dinosterol, which can help

638   distinguish changes in water isotopes from changes in other factors that affect α_{Wax-P} values. As

639   is the case for all paleoclimate proxies, interpretations of δ^2H_{Wax} values are most robust in a

640   multiproxy context.

Acknowledgments, Samples, and Data

Figure captions

652   **Figure 1** Global distribution of leaf wax samples from surface sediments and soils. The left
653   panel shows the locations for n-C_{29}-alkane (665 sites); the right panel shows the locations for n-
654   C_{28}-acid (242 sites). Background shading represents annual mean δ^2H_P values from the Online
655   Isotopes in Precipitation Calculator (OIPC) (Bowen & Ravenaugh, 2003; IAEA/WMO, 2015;
656   Bowen, 2020). OIPC does not produce spatial data sets over marine areas, therefore shading is
657   limited to continents.

658

659   **Figure 2:** Correlation coefficients of linear regressions of δ^2H values of all analyzed compounds
660   relative to δ^2H_P values (blue circles) and mean annual precipitation (pink diamonds) in the
661   tropical Pacific. Filled symbols represent significant correlations at the 95% confidence level.
662   Mean annual precipitation is from the Global Precipitation Climatology Project (GPCP) and δ^2H_P
663   values are from the OIPC. Compounds are grouped by source (algal, general, or plant waxes,
664   with increasingly likely terrestrial plant sources associated with longer chain lengths). Dinosterol
665   δ^2H data is from Maloney et al. (2019), all other lipid δ^2H data from this study. Individual
666   measurements are included in Data Set S1.

667

668   **Figure 3** δ^2H values of (a, c) *n*-C$_{29}$-alkane and (b, d) *n*-C$_{28}$-acid from surface sediments and soils
669   plotted relative to OIPC-derived δ^2H$_P$ values, color-coded by region (a, b), and sample type (c,
670   d). In panels a and b, red diamonds are lakes from the SPCZ region and green squares are
671   mangrove swamps in Micronesia (this study), both plotted with error bars. X-axis error bars
672   represent 95% confidence intervals of OIPC values. Y-axis error bars represent 1 standard
673   deviation of measurements from replicate samples from the same lake or swamp and are
674   typically smaller than the marker size. Circles are global values compiled from the literature,
675   color-coded by region. X-axis error bars are not shown for previously published data points, and
676   average 5.2‰ for sites outside the tropical Pacific. Regression statistics in (a) and (b) are shown
677   with and without new Pacific data. Globally compiled data (including tropical Pacific values) do
678   not differ significantly between soils and lacustrine sediments for either (c) *n*-C$_{29}$-alkane or (d) *n*-
679   C$_{28}$-acid. Shading around linear regressions represents 95% confidence intervals.

680

681   **Figure 4** Correlation coefficients of random subsamples of 17 values from the global
682   compilation surface sediment and soil of (a) *n*-C$_{29}$-alkane and (b) *n*-C$_{28}$-acid δ^2H values plotted
683   relative to range of δ^2H$_P$ values. Subsampled data were taken from the full data set and from
684   restricted δ^2H$_P$ ranges as described in the text. Correlation coefficients for the global compilation
685   and each continent are plotted for comparison.

686

687   **Figure 5** Correlation plots of δ^2H values from the global compilation of surface sediments and
688   soils relative to the δ^2H$_P$ values from various models (described in section 2.4). Numbers, colors,
689   and the widths of the ellipses correspond to correlation coefficients (R values).

690

691   **Figure 6** Pollen distributions from surface or near surface sediments in tropical Pacific lake
692   samples plotted relative to residuals from the global δ^2H$_{Wax}$ – δ^2H$_P$ calibration line for (a) *n*-C$_{29}$-
693   alkane and (b) *n*-C$_{28}$-acid. Square and triangle symbols are used to distinguish among multiple
694   sites with the same residual values.

695

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1135 **Table 1** Lake location, total number of samples analyzed per site, δ^2H values of local precipitation and
1136 leaf waxes, and measured δ^2H$_{Wax}$ values with residuals from predicted values based on linear correlation
1137 of compiled literature values

Site, Island, Country	Lat. (°N)[1]	Long. (°E)[1]	# of sam ples	δ^2H$_P$ (‰, VSMOW)[2]	δ^2H n–C$_{29}$ alkane[3] (residual from global relationship[4]) (‰, VSMOW)	δ^2H n–C$_{28}$ acid[3] (residual from global relationship[4]) (‰, VSMOW)
Lakes						
Rimatu'u Pond, Tetiaroa, French Polynesia	−17.0249	210.4417	2	−25 ± 26	−177 (−38 ± 12)	−160 ± 17 (−28 ± 19)
Oroatera Pond, Tetiaroa, French Polynesia	−16.9958	210.4591	1	−25 ± 26	−173 (−34 ± 12)	*N.A.*
Onetahi Pond, Tetiaroa, French Polynesia[#]	−17.0207	210.4081	1	−25 ± 26	−139 (−0 ± 12)	−126 (5 ± 10)
Lake Lanoto'o, Upolu, Samoa	−13.9109	188.1726	3	−34 ± 3	−159 (−12 ± 1)	−148 ± 6 (−9 ± 6)
Lac Lalolalo, Wallis, Wallis and Futuna	−13.3017	183.7662	3	−23 ± 1	*N.A.*	−150 ± 6 (−20 ± 6)
Lac Lanutavake, Wallis, Wallis and Futuna	−13.3212	183.7860	2	−24 ± 2	*N.A.*	−140 ± 9 (−9 ± 9)
Lake Dranoniveilomo, Vanua Balavu, Fiji	−17.1976	181.0441	2	−21 ± 11	−151 (−15 ± 5)	−173 ± 14 (−44 ± 14)
Lake Tagamaucia, Teveuni, Fiji[#]	−16.8163	180.0601	2	−34 ± 14	−170 ± 1 (−23 ± 7)	−175 ± 1 (−36 ± 5)
Otas Lake, Efate, Vanuatu	−17.6945	168.5850	1	−34 ± 73	−154 (−6 ± 35)	−136 (2 ± 27)
Emaotul Lake, Efate, Vanuatu	−17.7342	168.4151	3	−36 ± 76	−152 ± 9 (−3 ± 36)	−130 ± 3 (9 ± 26)
White Lake, Thion, Vanuatu	−15.0410	167.0892	2	−35 ± 70	−174 (−25 ± 34)	−119 ± 2 (21 ± 26)
Waérowa East Lake, Espiritu Santo, Vanuatu[#]	−15.5950	167.0788	1	−34 ± 71	*N.A.*	−155 (−16 ± 27)
Nopovois Pond, Espiritu Santo, Vanuatu	−15.4970	166.7357	1	−40 ± 71	−154 (−1 ± 34)	−122 (21 ± 27)
Vesalea Pond, Espiritu Santo, Vanuatu[#]	−15.1589	166.6549	1	−40 ± 70	−157 (−4 ± 34)	*N.A.*
Lake Hut, Grande Terre, New Caledonia	−22.2609	166.9526	2	−15 ± 54	−161 ± 2 (−31 ± 26)	−133 ± 1 (−8 ± 20)
Lake Tavara, Tetepare, Solomon Islands	−8.7029	157.4503	1	−46 ± 43	−162 (−4 ± 21)	−156 (−9 ± 16)
Lake Rano, Rendova, Solomon Islands	−8.6879	157.3243	2	−47 ± 42	*N.A.*	−135 ± 5 (13 ± 17)
Harai Lake #1, Rendova, Solomon Islands	−8.5622	157.3556	1	−47 ± 42	*N.A.*	−121 (27 ± 16)
Harai Lake #3, Rendova, Solomon Islands	−8.5648	157.3651	2	−47 ± 42	*N.A.*	−134 ± 11 (15 ± 19)
Mangrove swamps						
Sapwalap Swamp, Pohnpei, Fed. States of Micronesia	6.88	158.30	5	−33 ± 2	−150 ± 5 (−3 ± 5)	*N.M.*
Tol Swamp, Chuuk, Fed. States of Micronesia	7.35	150.60	4	−32 ± 1	−153 ± 5 (−2 ± 5)	*N.M.*
Sasa Swamp, Guam, United States	13.45	140.73	4	−29 ± 1	−145 ± 5 (−7 ± 5)	*N.M.*
Galal Swamp, Yap, Fed. States of Micronesia	9.50	138.08	5	−34 ± 1	−151 ± 6 (−3 ± 6)	*N.M.*

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1139 [1]Less precision is provided for latitude and longitude in mangrove swamps because swamp samples were collected
1140 along a transect typically spanning > 1 km.
1141 [2]Mean annual precipitation δ^2H values (relative to VSMOW) from OIPC ± 95% confidence intervals.

1142   [3]Mean value of multiple surface sediment measurements from same lake, relative to VSMOW. Uncertainties
1143   represent 1 standard deviation. When only one sample was analyzed no uncertainty is reported. Analytical
1144   uncertainty for compound specific δ^2H measurements is 4‰. *"N.A."* = compound was not present or was below
1145   detection limit for δ^2H measurements. "*N.M.*" = not measured.
1146   [4]Residuals are offsets from global calibration line of compiled leaf wax δ^2H values from the literature. Uncertainties
1147   are site specific standard deviations of OIPC δ^2H values, and are propagated with standard deviations of leaf wax
1148 δ^2H values when multiple samples are available from a site.
1149   [#]Lakes with greater than 50% vegetation cover
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1193 **Table 2** Pollen counts from near surface sediments, reported as a percentage of total palynmorphs counted. For each sediment sample, age ranges
1194 are presented for the top and bottom depth.

Site, Island, Country	Depth of pollen sample (cm)	Bacon age at top of interval (year C.E.)$	Bacon age at bottom of interval (year C.E.)$	Primary forest (%)	Secondary forest (%)	Mangroves (%)	Ferns (%)	Non-vascular plants (%)	Dryland herbs (%)	Wetland herbs (%)	Aquatic plants (%)	Unknown (%)
Lake Lanoto'o, Upolu, Samoa	1-2	2013 ± 1	2002 ± 1	3.0	24.6	0	62.8	0	2.7	6.9	0	0
Lac Lalolalo, Wallis, Wallis and Futuna*	1-3	2001 +8 -14	1991 + 14 -19	10.3	57.1	0	16.4	0	0.4	13.4	0.8	1.7
Lac Lanutavake, Wallis, Wallis and Futuna	3-4	1990 +20 -64	1983 +26 -86	13.1	69.1	0	4.3	0	6.2	4.6	0	2.7
Lake Dranoniveilomo, Vanua Balavu, Fiji	2-3	2010 ± 2	2009 ± 3	21.0	33.6	0.7	17.5	1.4	4.2	19.6	0	2.1
Lake Tagamaucia, Teveuni, Fiji#	2-3	1989 ± 7	1978 ± 10	5.1	13.4	0	58.6	0	0.6	21.3	0	1.0
Otas Lake, Efate, Vanuatu	2-3	N.A.	N.A.	4.7	52.1	15.6	2.7	0	0	21.8	0.8	2.3
Emaotul Lake, Efate, Vanuatu	1-2	2016 ± 3.4	2014 ± 3.4	4.9	55.8	0	12.7	0.3	11.4	8.4	5.2	1.3
White Lake, Thion, Vanuatu	3-4	1997 +23 -14	1991 +30 -19	1.3	42.3	0	39.3	0	2.1	11.7	0	3.4
Waérowa East Lake, Espiritu Santo, Vanuatu#	3-1	2010 ± 3	2009 ± 3	1.2	11.2	14.1	35.9	0	11.2	14.7	7.7	4.1
Nopovois Pond, Espiritu Santo, Vanuatu	0-1	2017	N.A.	16.3	53.5	0	15.0	0	8.9	3.1	0.3	2.6
Vesalea Pond, Espiritu Santo, Vanuatu#	0-1	2016 +1 -3	2005 +10 -14	6.5	46.2	0	18.8	1.5	11.4	6.8	4.6	4.3
Lac Hut, Grand Terre, New Caledonia	0-1	N.A.	N.A.	50.1	41.1	0	4.8	0	0	0.9	0	3.1
Lake Tavara, Tetepare, Solomon Islands	8-9	1996 ± 5	1993 ± 6	6.3	23.4	6.3	54.7	0	1.6	7.8	0	0.1
Lake Rano, Rendova, Solomon Islands	9-10	1969 +17 -16	1960 +21 -20	16.7	46.5	0	29.8	0	0	4.4	0.9	1.8
Harai Lake #1, Rendova, Solomon Islands	11-12	1716 +99 -123	1702 +103 -120	5.4	19.4	2.2	66.7	0	0	5.4	0	1.1
Harai Lake #3, Rendova, Solomon Islands	30-31	1871 ± 85	1866 +86 -84	3.1	5.2	0	91.8	0	0	0	0	0

1195 *Mean of 2 samples from different sites in these lakes. Age ranges presented represented the mean age for the top and bottom of each interval, and
1196 the full range of possible ages for both sites.
1197 # Lakes with greater than 50% vegetation cover
1198 $ Age ranges are provided from sites with existing age models, the details of which are provided by Maloney et al. (2019), Krentscher et al. (2019),
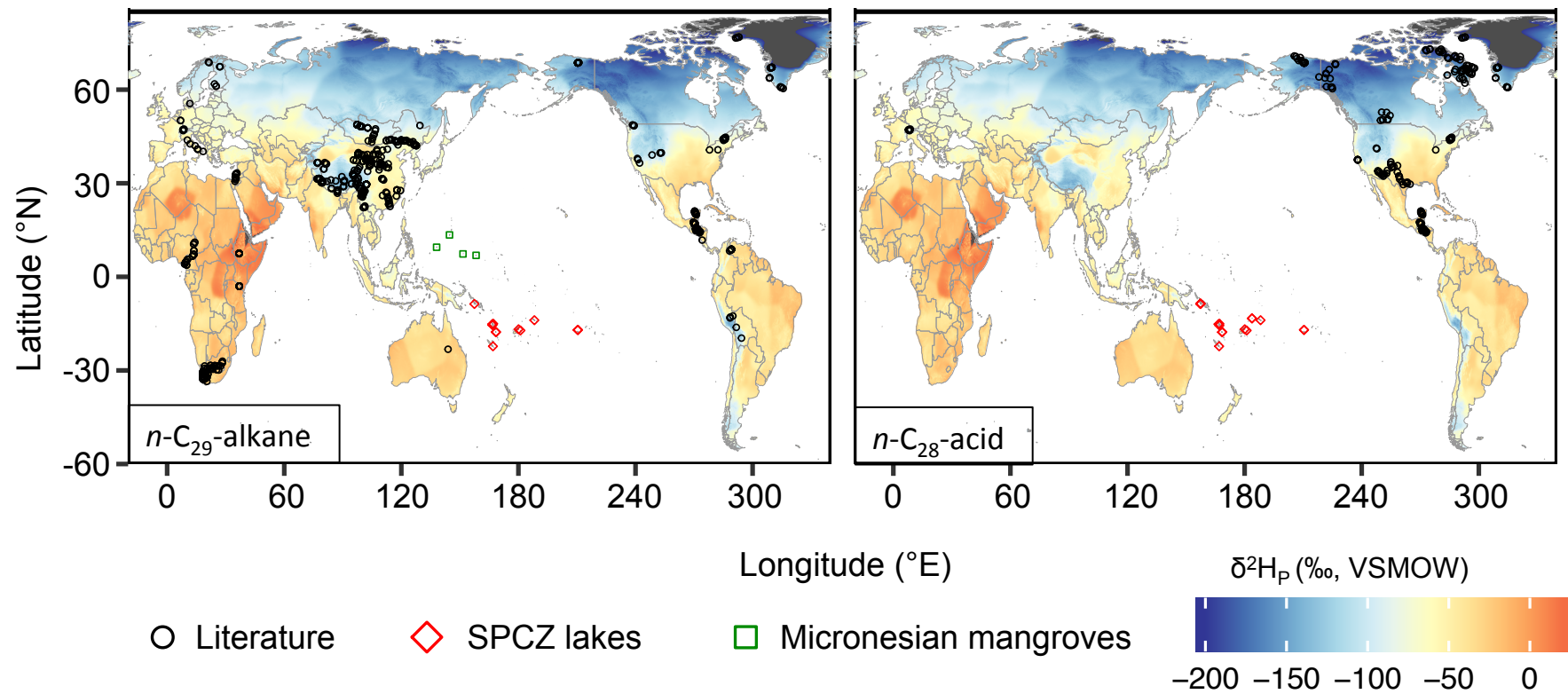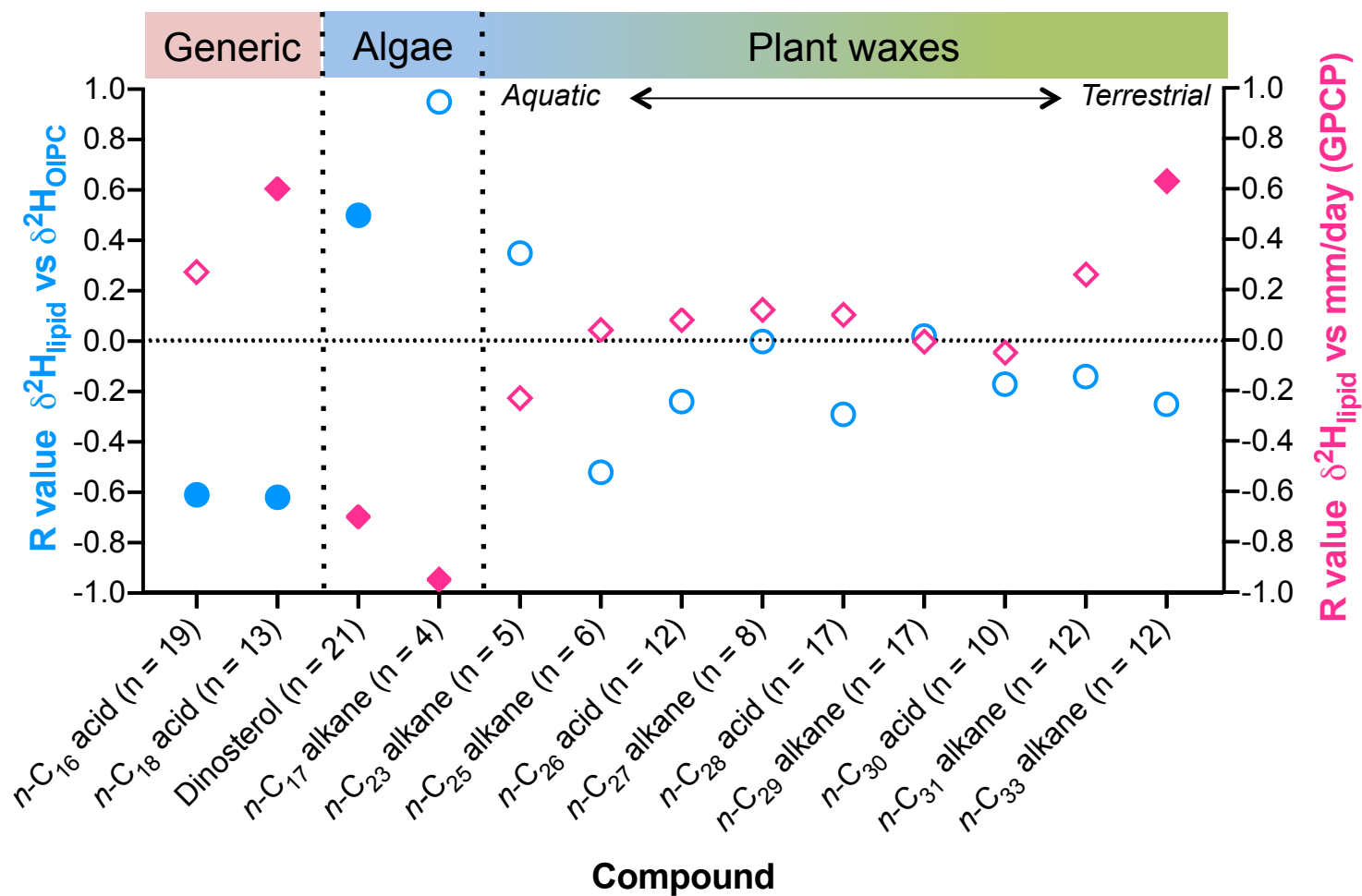1199 Gosling et al. (2020), and Sear et al. (2020)
1200
1201
1202

Figure 1.

Figure 2.

Figure 3.

a)

δ²H *n*-C$_{29}$-alkane (‰, VSMOW)

With Tropical Pacific:
m = 0.92 ± 0.02; b = -117 ± 2
R^2 = 0.74; p < 0.0001; n = 665
Without Tropical Pacific:
m = 0.93 ± 0.02; b = -116 ± 2
R^2 = 0.74; p < 0.0001; n = 648

SPCZ
Micronesia
Africa
Americas
Australia
Asia
Europe

δ²H mean annual precip (‰, VSMOW)

b)

δ²H *n*-C$_{28}$-acid (‰, VSMOW)

With Tropical Pacific:
m = 0.73 ± 0.03; b = -114 ± 3
R^2 = 0.74; p < 0.0001; n = 242
Without Tropical Pacific:
m = 0.74 ± 0.03; b = -113 ± 3
R^2 = 0.73; p < 0.0001; n = 225

SPCZ
N. America (<25°N)
Switzerland
N. America (25 - 60°N)
N. America (> 60°N)

δ²H mean annual precip (‰, VSMOW)

c)

δ²H *n*-C$_{29}$-alkane (‰, VSMOW)

Lakes
m = 0.902 ± 0.03; b = -114 ± 2
R^2 = 0.82; p < 0.0001; n = 191

Soils
m = 0.976 ± 0.03; b = -115 ± 2
R^2 = 0.67; p < 0.0001; n = 474

δ²H mean annual precip (‰, VSMOW)

d)

δ²H *n*-C$_{28}$-acid (‰, VSMOW)

Lakes
m = 0.758 ± 0.03; b = -113 ± 3
R^2 = 0.75; p < 0.0001; n = 224

Soils
m = 0.860 ± 0.03; b = -102 ± 5
R^2 = 0.98; p < 0.0001; n = 18

δ²H mean annual precip (‰, VSMOW)

Figure 4.

a) *n*-C$_{29}$-alkane

Legend:
- ★ Global (n = 665)
- ● Tropical Pacific (n = 17)
- ■ Africa (n = 117)
- ▲ Asia (n = 383)
- ▼ Europe (n = 22)
- ◆ N. America (n = 105)
- ⬡ S. America (n = 20)

Sampling range:
- ● All data
- ● 100‰
- ● 50‰
- ● 35‰

b) *n*-C$_{28}$-acid

Legend:
- ★ Global (n = 242)
- ● Tropical Pacific (n = 17)
- ▼ Europe (n = 8)
- ◆ North America (n = 217)

Sampling range:
- ● All data
- ● 100‰
- ● 50‰
- ● 35‰

Range of δ^{2}H$_P$ Values (‰)

Figure 5.

	n-C$_{28}$-acid	OIPC	OIPC–extracted	CAM (free)	ECHAM (nudged)	GISS (nudged)	GISS (free)	HadAM (free)	IsoGSM (nudged)	LMDZ (nudged)	MIROC (free)	SWING2 nudged mean
n-C$_{29}$-alkane (n = 665)	74	86	84	64	67	70	69	57	66	65	67	70
n-C$_{28}$-acid (n = 242)		86	82	91	87	84	86	89	86	85	89	88

R

0.5   0.6   0.7   0.8   0.9   1

Figure 6.

Legend:
- Primary forest
- Secondary forest
- Mangroves
- Non-vascular plants
- Dryland herbs
- Wetland herbs
- Aquatic plants
- Ferns

□ △ Different shapes indicate different lakes with same residual values

a) Relative abundance (%) vs n-C_{29}-alkane residual

b) Relative abundance (%) vs n-C_{28}-acid residual