Tropical South Atlantic influence on Northeastern Brazil precipitation and ITCZ displacement during the past 2300 years

Giselle Utida, Francisco W. Cruz, Johan Etourneau, Ioanna Bouloubassi, Enno Schefuß, Mathias Vuille, Valdir F. Novello, Luciana F. Prado, Abdelfettah Sifeddine, Vincent Klein, André Zular, João C. C. Viana & Bruno Turcq

Recent paleoclimatic studies suggest that changes in the tropical rainbelt across the Atlantic Ocean during the past two millennia are linked to a latitudinal shift of the Intertropical Convergence Zone (ITCZ) driven by the Northern Hemisphere (NH) climate. However, little is known regarding other potential drivers that can affect tropical Atlantic rainfall, mainly due to the scarcity of adequate and high-resolution records. In this study, we fill this gap by reconstructing precipitation changes in Northeastern Brazil during the last 2,300 years from a high-resolution lake record of hydrogen isotope compositions of plant waxes. We find that regional precipitation along the coastal area of South America was not solely governed by north-south displacements of the ITCZ due to changes in NH climate, but also by the contraction and expansion of the tropical rainbelt due to variations in sea surface temperature and southeast trade winds in the tropical South Atlantic Basin.

Northeastern Brazil (NEB), also known as Nordeste, is one of the most vulnerable regions to climate change in South America. During the last decades, the NEB has experienced a drastic reduction in precipitation causing desertification expansion faster than anywhere else on the continent. The causes of such anomalous climatic conditions remain elusive and probably are driven by several processes, which are still not fully understood. A better comprehension of these processes is of particular societal relevance since the NEB is densely populated and currently facing severe problems of water supply.

Despite the dramatic decrease in precipitation over NEB as a whole, there are significant differences between the northern and eastern coastal sectors. Although both are influenced by the Tropical Atlantic Ocean, most of the NEB, especially the northern area, is primarily influenced by the seasonal displacement of the ITCZ reaching its southernmost position during austral autumn (March to May, MAM). The ITCZ is defined as a maximum in tropical precipitation or as a tropical belt of convective clouds – tropical rainbelt, and its mean position varies seasonally from 9°N to 2°N over the Atlantic Ocean. During the seasonal ITCZ shift to the south, higher precipitation in NEB is associated with warmer sea surface temperatures (SST) in the Tropical South Atlantic (TSA) and weaker Southeast (SE) trade winds (Fig. 1A). Indeed, tropical warming in the trade wind convergence zone promotes the ascent of warm and moist air, contributing to deep convective cloud formation. At higher levels, divergence leads to poleward flow and subsidence over subtropical latitudes, where near-surface flow is redirected towards the equator, closing the meridional Hadley cell.

In contrast to austral autumn, the austral winter (June to August, JJA) is characterized by significant cooling in the TSA, stronger SE trade winds that cross the Equator and a northward displacement of the ITCZ (Fig. 1B).
Additionally, in contrast to the northern region, precipitation over the eastern coastal NEB is modulated by the sea breeze circulation and easterly waves disturbances (EWD), which propagate westward over the tropical South Atlantic Ocean and are intensified by the SE trade winds during JJA. This meridional gradient is also known as Atlantic Meridional Mode, where TSA and the tropical North Atlantic (TNA) can vary independently on decadal time scales (e.g.9–13). Other zonal oceanic modes in the tropics may also influence the climate in NEB, such as the Atlantic Equatorial Mode (AEM) or the El Niño Southern Oscillation (ENSO). In contrast to Meridional Mode, the AEM and ENSO records usually do not contain sufficient temporal resolution to allow a discussion of its possible influence. Moreover, the AEM amplitude and impact on an interannual basis are relatively small and hardly detectable in decadal to centennial paleoclimate records.

Northward ITCZ displacements in response to warm NH temperature anomalies during the Holocene are by far the most commonly invoked cause explaining climate variability across the tropical Atlantic coastal regions.
Results and Discussion

Our record documents a predominantly humid period between 500 yrs BCE and 420 yrs CE associated with low δDwax values ranging from −130 to −145‰. These climate conditions are followed by a abrupt aridification and a long dry phase from 500 to 1,300 yrs CE as revealed by higher δDwax values reaching −100‰ (Fig. 2A). Between 1580 and 1900 yrs CE, the δDwax values rapidly declined, reaching values similar to those prior to 420 yrs CE, thus characterizing a long humid period during the Little Ice Age (LIA) in NEB.

There is a striking resemblance between our δDwax record and the reconstructed Boqueirão Lake level variations for the last 2,000 yrs based on a diatom transfer function in the same sediment core Boqc09/01 (Fig. 2A,B). However, the periods with marked increase (decrease) of ITCZ-related precipitation indicated by lower (higher) δDwax values occur unexpectedly during periods of significant lake level drop (rise) reconstructed by Viana et al.\textsuperscript{16} and Zocatelli et al.\textsuperscript{34}. This inconsistency can be explained by the strong influence of aeolian processes on the Boqueirão Lake level\textsuperscript{13}, which was not considered in previous studies. Boqueirão is a coastal lake that originated from sand-dune damming of a small freshwater river (Fig. S4). Our isotopic data suggest that the lake level high-stands are associated with advancing dunes over the drainage that led to damming during dry periods with reduced river flow. Conversely, during wetter climate periods the increased drainage flow would most likely erode the dune dam and pass through any obstruction, resulting in a lowering of the lake water level.

To investigate latitudinal ITCZ displacements, we compare the data from Boqueirão Lake to the Ti record from Cariaco Basin\textsuperscript{14} (Fig. 2C), which is one of the most commonly used proxies for past ITCZ locations over
the tropical North Atlantic. Because our lake record is located close to the southern limit of the ITCZ domain in South America, we evaluated expansion and contraction of the tropical rain belt by comparing our record with the Cariaco Basin, which is located close to the northern limit of the ITCZ. In the earliest part of our record (i.e. before ~500 yrs CE), relatively humid conditions prevail in both NEB and Cariaco (correlation $< -0.5$, $p < 0.05$, see Supplementary Material), which is not consistent with the traditional pattern of meridional ITCZ displacement regulating precipitation at both sites (Fig. 2). Our new results, combined with the Cariaco record, instead suggest an expansion of the tropical rainbelt that would correspond to a longer rainy season in both hemispheres. This in-phase relationship might be a consequence of a general warming of the Atlantic Equatorial Basin. However, this hypothesis would need to be further supported by new and high-resolution SST reconstructions from the tropical Atlantic for the last millennia.

On average for the whole tropical Atlantic, a warming of the sea surface temperature was documented over the last decades associated with strengthened trade winds without significant long-term changes in the mean position of the ITCZ, although the maximum northern and southern ITCZ displacements occurred during these decades. Alternatively, other mechanisms could also influence the width of the tropical rainbelt, such as cloud-radiative feedbacks, or variations in the tropical moist-static energy budget. Our record is also in agreement with evidence of a tropical rainbelt expansion before ~250 yrs CE from the Pacific ITCZ domain.

NEB faced a very long dry period between 500 and 1,500 yrs CE, which is consistent with a relatively far northern mean position of the ITCZ (Fig. 2A). This climate scenario is consistent with the overall wet conditions in Cariaco relative to the large dry event that characterizes the Little Ice Age (LIA). Humid conditions in Cariaco peaked around 1,000 yrs CE, corresponding to the Medieval Climate Anomaly, and were bounded by two dry periods lasting from approximately 600 to 800 yrs CE and from 1,100 to 1,300 yrs CE (Fig. 2C).

---

**Figure 2.** Record comparison between (A) δD$_{n-C_{28}}$ alkanoic acid from Boqueirão Lake sediment core Boq0901 in Northeastern Brazil (NEB), black line represents a smoothing with a 20-point window (this study) with $^{14}$C AMS ages identified by blue symbols; (B) Boqueirão Lake level reconstruction; (C) Ti record from Cariaco Basin, Venezuela, orange line represents a smoothing with a 20-point window and orange symbols $^{14}$C AMS ages. All smoothed records were calculated according to Savitzky-Golay method performed by the software Origin 8.0. Blue shaded area from 420 BCE to 500 yrs CE indicates a wet period (in-phase) at Boqueirão lake and Cariaco Basin, red shaded area from 500 to 1500 yrs CE indicates a dry period in Boqueirão Lake and wet in Cariaco (antiphased) and gray shaded area from 1500 to 1830 yrs CE indicates wet conditions in Boqueirão and dry in Cariaco (antiphased). The running-mean correlation results are presented in the Supplementary Material.
We associate the northward displacement of the ITCZ with the reinforcement of the SE trade winds, resulting in a shallower equatorial thermocline and TSA cooling. This would promote a long dry phase in NEB (Fig. 2A), as expected for a northward displacement of the ITCZ. We therefore conclude that the large shift in precipitation at ~500 yrs CE in the NEB was strongly influenced by temperature changes in TSA and intensified SE trade winds, as an important, and hitherto overlooked, contributing factor working in conjunction with warmer NH temperature.

Around 1500 yrs CE, NEB climate abruptly transitioned towards wetter conditions, as a consequence of a more southerly position of the ITCZ, affecting latitudes as far south as 5°S. This is consistent with a dry climate recorded in Cariaco from 1600 to 1850 yrs CE during the LIA (Fig. 2, see also S9 in Supplementary Material) and also with the ITCZ stack reconstruction from Lechleitner et al. This period of ITCZ rainfall extending into the southern hemisphere tropics is commonly associated with cooler temperatures in the northern hemisphere. Indeed, a modern-day record of colder TNA and warmer TSA confirms a southern displacement of the ITCZ and stronger precipitation over NEB.

During the LIA, Denniston et al. suggested a contraction of the Indo-Pacific tropical rain belt, however, given the lack of proxy records in the Atlantic margin, it is not possible to accurately constrain the contraction of the ITCZ over the tropical Atlantic domain.

It is well known that ENSO is associated with rainfall variability in NEB. Indeed, there is a delayed anomalous warming of the tropical North Atlantic during El Niño events, which reduces northeast trade winds and favors the ITCZ displacement to the north, while the opposite mechanism occurs during La Niña events. The past variability of ENSO during the last 2 millennia is still not clearly understood due to large inconsistencies among the existing reconstructions (Fig. S5). Such major differences might be attributed to regional features, dating uncertainties, distinct response to ENSO or a non-linear behavior of the proxy-climate relationship.

When comparing the ENSO records from the eastern Pacific with our NEB isotope data over the last 2,300 years, we do not find a clear evidence for an in-phase relationship as expected from the modern climatology (Fig. S5). In addition, the anticipated in phase relationship of long-term precipitation in Nordeste and western Pacific ENSO records or indexes is neither so apparent (Fig. S5D,E).

Our lake record suggests that the ITCZ activity, as represented by the so-called ‘meridional shift of the tropical rain belt’ hypothesis needs to be discussed with caution because it cannot fully explain the long-term precipitation variability over NEB during the last 2,300 years. We show here that the NH climate is not the sole driver of NEB precipitation and the tropical Atlantic needs to be considered as an additional important factor influencing the tropical rainbelt dynamics.

**Methods**

**Lipid extraction, quantification and identification.** Samples were prepared at LOCEAN-UPMC (Laboratoire d’Océanographie et du Climat, Expérimentations et Approches Numériques – Université Pierre et Marie Curie, Paris). Lipids from 89 sediment samples of the Boq09/01 core were extracted by ultrasonication with a DCM/MeOH (3:1) solvent mixture. After saponification of the resulting total lipid extracts with 4 M KOH/MeOH, removal of the neutral fraction with hexane and acidification of the residue, the acid fraction was recovered with hexane/ethyl acetate (9:1), methylated using BF3 and further purified by column chromatography over silica gel and elution with DCM:hexane (2:1). Fatty acid methyl esters (FAME) were analyzed on Agilent 6890 N gas chromatograph (GC) using flame ionization detection (FID). Quantification of compounds was performed by peak area integration in FID chromatograms relative to the internal standard 5α-cholenic acid added prior to extraction. In order to confirm compound identification selected samples were analyzed by GC-MS on an Agilent 7890 GC coupled to an Agilent 5975 mass spectrometer detector. High values of Carbon Preservation Index (CPI) obtained, between 7.7 and 11.1, indicate good plant wax preservation. Concentrations of individual long-chain fatty acids (C24-32) range between 20 and 180 μg/g in the dry sediment samples. Taking into account that the pattern of long-chain fatty acids was very similar to other fatty acids, for instance n-C30, alkanolic acid, we chose to analyze n-C28 alkanoic acids, which is very close to the average carbon length in our samples (27.4) and has major quantities per gram (C-28 average 7.2 mg/g; C-30 average 5 mg/g), resulting in higher confidence, reliability and reproducibility of our results.

**Isotope analysis.** Compound-specific isotope analyses of n-C28 alkanoic acids were performed at MARUM - Center for Marine Environmental Sciences, Bremen, Germany. Compound-specific δD analyses were performed on a Thermo Trace GC equipped with an Agilent DB-5 column (30 m × 0.25 mm × 0.25 μm) coupled to a Thermo Fisher Scientific MAT 253 IRMS via a pyrolysis interface operated at 1420 °C. Measurements were calibrated against δH reference gas with known isotopic composition and the Hδ+ factor was monitored daily (values varied between 6.7 and 6.9). δD values are reported in permil (%) relative to Vienna Standard Mean Ocean Water (VSMOW). An external standard mixture with known δD values was analyzed repeatedly every six runs, yielding a long-term mean standard deviation of <3% and a mean deviation of <1% from reference values. Stable carbon isotope compositions (δ13C) of the same compounds were measured using the same type of GC and GC column coupled to a Finnigan MAT 252 IRMS via a modified combustion interface at 1000 °C. Calibration of carbon isotopes was achieved by comparison to CO2 reference gas. δ13C values are reported in permil (%) against Vienna Pee Dee Belemnite (VPDB). An external standard mixture was analyzed repeatedly every 6 runs and yielded a long-term mean standard deviation of 0.2‰ with a mean deviation of 0.1‰ from the reference values. Samples were measured in duplicates for δD and δ13C. Mass balance calculations were made for removal of the isotopic contribution of the added methyl group. Mean propagated errors are 3% for δD and 0.2‰ for δ13C. Isotopic results obtained from n-C28 alkanoic acids of Boqueirão Lake core Boq09/01 are reported as δD wax and δ13Cwax.
Correction of δD_{wax} for vegetation-type. To remove fractionation effects based on vegetation type changes, we corrected the isotopic composition of δD_{wax} based on the δ^{13}C_{wax} signal. The δ^{13}C_{wax} varied from −26.1‰ to −32.5‰ (±0.5‰) (Fig. S6) indicating a predominance of the C3 plant signal during the last 2300 years. We tested a mixing model using the epsilon value weighted by different proportions of C3 and C4 vegetation, applying end-members of −35‰ and −22‰ for C3 and C4 plants (Fig. S6), respectively. To correct the δD_{wax}, we weighted fractionation factor according to C3/C4 plants using fractionation factor (ε) of n-alkanes of C3 plants in tropical dry forests and of C4 grasses in semi-arid environments, where the amount of precipitation is the most similar to NEB. Both end-members and fractionation factor were obtained according to compilation of Sachse et al.

The estimated precipitation (δD_{precip-wax}) values were calculated according to the following equation:

\[ \delta D_{\text{precip-wax}} = \frac{[ (\delta D_{\text{wax}} + 1000) (1/1000) + 1]}{1000} \]

The results vary between 32% and −17% for ε weighted by C3/C4 (Fig. S7).

The results using fractionation factors weighted by C3/C4 from n-alkanes are consistent with the modern observed δD_{precip} (Fig. S8). However, there are no significant changes in δD_{precip-wax} patterns (Fig. S7). Therefore, we infer a weak influence of plant physiology and evapotranspiration on isotopic enrichment in soil and leaf waters in highly seasonal environments but discuss our results in terms of trends rather than absolute values.

Data Availability

The dataset generated during the current study will be available in the PANGAEA.

References

1. Marengo, J. A., Torres, R. R. & Alves, L. M. Drought in Northeast Brazil—past, present, and future. Theor. Appl. Climatol. 129, 1189–1200, https://doi.org/10.1007/s00704-016-1840-9 (2016).
2. Hastenrath, S. Exploring the climate problems of Brazil’s Nordeste: a review. Clim. Change 112, 243–251, https://doi.org/10.1007/s10584-011-0227-1 (2012).
3. Philander, S. G., Gu, D., Lambert, G. & Li, T. Why the ITCZ is mostly north of the equator. J. Climate 9, 2958–2972, https://doi.org/10.1175/1520-0442(1996)009<2958:TWITCZ>2.0.CO;2 (1996).
4. Waliser, D. E. & Gautier, C. A satellite-derived climatology of the ITCZ. J. Climate 6, 2162–2174, https://doi.org/10.1175/1520-0442(1993)006<2162:ASCDCO>2.0.CO;2 (1993).
5. Schneider, T., Bischoff, T. & Haug, G. Migrations and dynamics of the intertropical convergence zone. Nature 513(7516), 45–53, https://doi.org/10.1038/nature13634 (2014).
6. Nobre, P. & Shukla, J. Variations of sea surface temperature, wind stress, and rainfall, over the Tropical Atlantic and South America. J. Climate 9, 2464–2479, https://doi.org/10.1175/1520-0442(1996)009<2464:VOSTSW>2.0.CO;2 (1996).
7. Kousky, V. E. Frontal influences in Northeast Brazil. Mon. Weather Rev. 107, 1101–1113 (1979).
8. Gomes, H. B. et al. Easterly Wave Disturbances over Northeast Brazil: An Observational Analysis. Adv. Meteorol. 2015, 176238, https://doi.org/10.1155/2015/176238 (2015).
9. Enfield, D. B., Mestas-Nuñez, A. M., Mayer, D. A. & Sid-Serrano, L. How ubiquitous is the dipole relationship in tropical Atlantic sea surface temperatures? J. Geophys. Res.: Oceans C4, 7841–7848, https://doi.org/10.1029/1998JC900109 (1999).
10. Wang, C. ENSO, Atlantic climate variability, and the Walker and Hadley circulations. In Diaz, H. F. & Bradley, R. S. eds, The Hadley circulation: present, past and future. Advances in Global Change Research 21. Springer [Dordrecht], 173–202, https://doi.org/10.1007/978-1-4020-2944-8_7 (2004).
11. Mehta, V. M. Variability of the tropical ocean surface temperatures at decadal multimillennial timescales. Part I: The Atlantic Ocean. J. Climate 11, 2331–2375 (1998).
12. Servain, J., Wainer, I., McCreary, J. P. & Dessier, A. Relationship between the equatorial and meridional modes of climatic variability in the tropical Atlantic. Geophys. Res. Lett. 26(4), 485–488, https://doi.org/10.1029/1999GL900114 (1999).
13. Servain, J. et al. Recent climatic trends in the tropical Atlantic. Clim. Dyn. 43, 3071–3089, https://doi.org/10.1007/s00382-014-2168-7 (2014).
14. Haug, G. H. et al. Southward migration of the Intertropical Convergence Zone through the Holocene. Science 293(5533), 1304–1308, https://doi.org/10.1126 science.1080725 (2001).
15. Pessenda, L. C. R. et al. Late Pleistocene and Holocene vegetation changes in northeastern Brazil determined from carbon isotopes and charcoal records in soils. Palaeogeogr. Palaeoclimatol. Palaeoecol. 297, 597–609, https://doi.org/10.1016/j.palaeo.2010.09.008 (2010).
16. Viana, J. C. C. et al. A late Holocene paleoclimate reconstruction from Boqueirão Lake sediments, northeastern Brazil. Palaeogeogr. Palaeoclimatol. Palaeoecol. 415, 117–126, https://doi.org/10.1016/j.palaeo.2014.07.010 (2014).
17. Lechleitner, F. A. et al. Tropical rainfall over the last two millennia: evidence for a low-latitude hydrologic seesaw. Sci. Rep. 7, 45809, https://doi.org/10.1038/srep45809 (2017).
18. Niedermeyer, E. M. et al. The stable hydrogen isotopic composition of sedimentary plant waxes as quantitative proxy for rainfall in the West African Sahel. Geochim. Cosmochim. Ac. 104, 55–70, https://doi.org/10.1016/j.gca.2016.03.034 (2016).
19. Collins, J. F. et al. Estimating the hydrogen isotopic composition of past precipitation using leaf-waxes from western Africa. Quat. Sc. Rev. 65, 88–101, https://doi.org/10.1016/j.quascirev.2013.03.007 (2013).
20. Sachse, D. et al. Molecular paleohydrology: Interpreting the hydrogen-isotopic composition of lipid biomarkers from photosynthesizing organisms. Ann. Rev. Earth Planet. Sci. 40(1), 221–249, https://doi.org/10.1146/annurev-earth-042711-105535 (2012).
21. Zalar, A. et al. The effects of mid-Holocene fluvo–olian interplay and coastal dynamics on the formation of dune–dammed lakes in NE Brazil. J. Quat. Sci. Rev. 196, 137–153, https://doi.org/10.1016/j.quascirev.2018.07.022 (2018).
22. French et al. Millennial soil retention of terrestrial organic matter deposited in the Bengal Fan. Sci. Rep. 8, 11997, https://doi.org/10.1038/s41598-018-30091-8 (2018).
23. Scheufler, E. et al. Hydrologic control of carbon cycling and aged carbon discharge in the Congo River basin. Nat. Geosc. 9(9), 687–690, https://doi.org/10.1038/NGEO2778 (2016).
24. Garreau, R. D., Vuille, M. & Compagnucci, R. & Marengo, J. Present-day South American climate. Palaeogeogr. Palaeoclimatol. Palaeoecol. 281, 180–195, https://doi.org/10.1016/j.palaeo.2007.10.032 (2009).
25. Dansgaard, W. Stable isotopes in precipitation. Tellus XVI, 436–468 (1964).
26. Hastenrath, S. & Keller, L. Dynamics of climatic hazards in northeast Brazil. Quat. J. R. Meteorol. Soc. 103(455), 77–92 (1977).
27. Rao, V. B., Lima, M. C. & Franchito, S. H. Seasonal and interannual rainfall variation in central and eastern Northeast Brazil. J. Climate 6, 1754–1763, https://doi.org/10.1175/1520-0442(1993)006<1754:SAVOR>2.0.CO;2 (1993).
28. Kayano, M. T. & Andreoli, R. V. Climax na região Nordeste do Brasil, In Cavalcanti, I. F. A., Ferreira, N. J., Silva, M. G. A. J. & Dias, M. A. F. eds, Tempo e Clima no Brasil: São Paulo, Oficina de Textos [São Paulo], 213–233 (2009).
60. Steinier, J., Termonia, Y. & Deltour, J. Smoothing and differentiation of data by simplified least square procedure.

59. Harris, I., Jones, P. D., Osborn, T. J. & Lister, D. H. Updated high-resolution grids of monthly climatic observations – the CRU

54. Smith, F. A. & Freeman, K. H. Influence of physiology and climate on

52. Schneider, T.

53. Polissar, P. J. & Freeman, K. H. Effects of aridity and vegetation on plant-wax D in modern lake sediments. Geochim. Cosmochim. Ac.

48. Atwood, A. R. & Sachs, J. P. Separating ITCZ- and ENSO-related rainfall changes in the Galápagos over the last 3 kyr using D/H ratios of multiple lipid biomarkers. Earth Planet. Sci. 404, 408–419, https://doi.org/10.1016/j.epsl.2014.07.038 (2014).

45. Li, J.

44. TS3.10 Dataset.

43

74

73

62

34

33

32

31

30

29

28

27

26

25

24

23

22

21

20

19

18

17

16

15

14

13

12

11

10

9

8

7

6

5

4

3

2

1

229

228

227

226

225

224

223

222

221

220

219

218

217

216

215

214

213

212

211

210

209

208

207

206

205

204

203

202

201

200

199

198

197

196

195

194

193

192

191

190

189

188

187

186

185

184

183

182

181

180

179

178

177

176

175

174

173

172

171

170

169

168

167

166

165

164

163

162

161

160

159

158

157

156

155

154

153

152

151

150

149

148

147

146

145

144

143

142

141

140

139

138

137

136

135

134

133

132

131

130

129

128

127

126

125

124

123

122

121

120

119

118

117

116

115

114

113

112

111

110

109

108

107

106

105

104

103

102

101

100

99

98

97

96

95

94

93

92

91

90

89

88

87

86

85

84

83

82

81

80

79

78

77

76

75

74

73

72

71

70

69

68

67

66

65

64

63

62

61

60

59

58

57

56

55

54

53

52

51

50

49

48

47

46

45

44

43

42

41

40

39

38

37

36

35

34

33

32

31

30

29

28

27

26

25

24

23

22

21
Author Contributions
G.U. designed the experiment, performed lipids, δD and δ13C analyses and prepared the manuscript with help from the co-authors; F.W.C. and A.S. directed the project; J.E. helped to designed the experiment and prepared the manuscript; I.B. coordinated the laboratory procedures for lipid extraction and identification; E.S. coordinated the laboratory procedures for lipid quantification, δD and δ13C analyses; V.K. assisted with lipids extraction and identification; M.V. helped with the interpretation and corrections on the manuscript and the production of figures; V.E.N., L.F.P. and A.Z. helped with the paleoclimate interpretations; A.S., J.P.V. and R.T. organized the field work and provided the radiocarbon dates.

Additional Information
Supplementary information accompanies this paper at https://doi.org/10.1038/s41598-018-38003-6.

Competing Interests: The authors declare any competing financial and/or non-financial interests.

Publisher’s note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2019