시스템 정보

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Regional Setting

Northwest Sumatra is located within the equatorial rainfall regime, which is characterized by high amounts of rainfall throughout the year and no dry season (1). Depending on the seasonal position of the Intertropical Convergence Zone (ITCZ), rainfall delivered by the maritime trade winds originates from the converging branches of either the northern or the southern Hadley cell (Fig. S1), resulting in two rainfall maxima during spring and fall. As the contribution of each season to the total amount of rainfall does not differ significantly, Northwest Sumatra is not attributed to the realms of the Australian–Indonesian or Asian monsoons (2, 3). With increasing distance from the equator, however, seasonal changes in the amount of rainfall received at the Maritime Continent become more pronounced.

Rainfall received across central West Sumatra derives from the Indian Ocean, as both the northern and southern monsoonal trade winds get deflected eastward near equator through Coriolis force. The leeward East of Sumatra, however, is blocked from the humid trade winds by the Barisan mountain range (4) and the Malay Peninsula (Fig. 1A). As a result, precipitation is highest in areas West of the mountain range (>3,000 mm/y), and decreases to <2,500 mm/y in the eastern areas (1) (Fig. S2). It further decreases toward the South of Sumatra due to its proximity to the Australian continent.

Lipid Analysis

Lipids were extracted from freeze-dried and finely ground sediment samples (>8 g) using the Maxis Xpress microwave extraction system from CEM GmbH. Sediments were suspended in 50 mL of 9:1 dichloromethane (DCM) and methanol (MeOH) and heated to 100 °C for 15 min with stirring. The solvent extract was filtered through a precombusted GF/F filter, then dried at 36 °C under a stream of nitrogen. Total lipid extracts were saponified in 1 M NaOH (aq) at 80 °C for 3 h. Neutral lipids were extracted with hexane, the pH was reduced to ~2 with HCl, and alcanoic (fatty) acids were then extracted into methyl t-butyl ether.

To enable analysis by gas chromatography (GC), fatty acids were methylated using BF₃-MeOH (10% wt/wt, Supelco) in DCM (60 °C, 10 min). Fatty acid methyl esters (FAMEs) were then extracted into hexane, dried over Na₂SO₄, and further purified through silica gel column chromatography (hexane/toluene 3:1) and AgNO₃/silica gel column chromatography (hexane/DCM 2:1).

The δD values of individual FAMEs were measured at least in duplicate using a Thermo Trace GC Ultra equipped with a Rxi-5ms column (30 m × 0.25 mm, film thickness 1 μm) coupled to a Delta V+ IRMS via a combustion furnace operated at 1030 °C. A CO₂ standard gas, together with a co-injected n-C₃₃ alkane standard, were used for calibration. An external standard of eight fatty acid alkyl esters (the same as used for δD analyses) with known δC values was analyzed periodically to monitor analytical performance. We report an average SD of sample n-alkanoic acids of about 0.6‰.

The δD and δ13C values were corrected for the isotopic composition of the attached methyl group, which was determined through repeated methylation of a phthalic acid standard of known isotopic composition. We report isotope ratios for surface n-C₃₀ alkanoic acid, which was the most cleanly separated compound of adequate abundance. Additionally, we show δD values for the n-C₃₂ alkanoic acid for the Holocene period. This restriction derives from poor data quality for the last glacial which is due to both low abundance and coelution of an unknown compound. All δD (δ13C) values are reported as permil [‰] variations relative to the Vienna standard mean ocean water (Vermont Pee Dee Belemnite) standard.

Corrections to the n-C₃₀ Alkanoic Acid δD Record

The δD record of the n-C₃₀ alkanoic acid was corrected for glacial–interglacial changes of ice volume and temperature. Correction for changes in ice volume was performed using the δD(H₂O)seawater reconstruction of Shackleton et al. (6). Using the modern Global Meteoric Waterline, changes of δD(H₂O)seawater were multiplied by 8 to approximate corresponding changes of δD(seawater) values, which were subtracted from the n-C₃₀ alkanoic acid δD record.

Temperature correction (Fig. S3) used a minimum estimate of an LGM to Holocene temperature difference (ΔT) of 3 °C [sea surface temperature of West Sumatra (7, 8)] and a maximum ΔT of 7 °C [West Sumatran highlands (9)]. The effect of ΔT on equilibrium fractionation was approximated as +0.8‰/°C D per °C [0.1‰/°C δ18O (10)] and was added to the ice-volume corrected n-C₃₀ alkanoic acid δD record.

We emphasize that temperature effects other than those on equilibrium fractionation at the evaporating surface, e.g., changes of the atmospheric temperature gradient and rainfall formation temperature, cannot be included at this point. Therefore, the temperature correction suggested here can only be a minimum estimate. Moreover, changes in relative humidity near the evaporative surface and associated kinetic isotope effects cannot be assessed. Given the tropical marine environment of our study site, however, we surmise the impact of kinetic isotope effects during evaporation on our integrated δD record to be small.

Provenance of Sedimentary Long-Chain Alkanoic Acids

Long-chain (>n-C₃₀) alkanoic acids recovered from marine sediments have been found to derive almost exclusively from higher terrestrial plants (11). Together with long chain n-alkanes and certain alkanols, they are integral parts of higher terrestrial plant leaf waxes (12). Depending on climatic and geomorphic conditions, they may accumulate in the ground (e.g., soils) or be carried by fluvial erosion.

The mineralogy and radiogenic isotope composition of clay minerals in Sumatran Shelf sediments indicate that rocks of North and central Sumatra are the dominant source of terrestrial material (13, 14). According to Ehlert et al. (14), characteristics of local sediment radiogenic isotopes are sharply separated from sediments offshore central Sumatra, which mix with clays from sources farther south. Our study site is located near the North Sumatran catchment as defined by Ehlert et al. (14) and has been geochemically isolated from the southwestern coastline during the
last sea-level lowstand of the LGM through shelf exposure (for a detailed reconstruction of the glacial shift of coastline position, see refs. 15 and 16). This natural barrier vanished during the Holocene, potentially allowing for mixing of local sediment with laterally advected plant waxes from the Sunda Shelf during sea level rise (15, 16) through concurrent strengthening of the Indonesian Throughflow (17). However, values of alkanolic acid 2δC values remain fairly stable throughout the Holocene. If consider- able mixing with sediment from southern sources had occurred, we would expect this to appear as a 13C enrichment, particularly for plant waxes transported from the Australian continent (dom- inantly C3 plants). In summary, we therefore infer that higher plant waxes deposited at the study site derive predominantly from the adjacent hinterland of Northwest Sumatra.

**Controls of the 6D Value of Terrestrial Plant Waxes**

**Atmospheric Controls.** The 6D values of higher plant leaf waxes are linked to the isotopic composition of the plant’s growth water, i.e., precipitation (18–22). In mid- and low-latitude coastal areas, dominant control on the isotopic composition of rainfall is exerted by the amount of rainfall (23, 24). Here, the degree of reevaporation and condensation during precipitation determines the 6D value of rainfall received at the surface (25). This leads to stronger D depletion in areas of high precipitation (and, as a consequence, of high relative humidity) and D enrichment when precipitation and relative humidity are low.

However, whereas 6D values of higher plant waxes isolated from marine and lacustrine environments in tropical and sub- tropical Africa are being interpreted as qualitative estimates of past rainfall amounts (26–29), recent studies indicate that the amount effect alone does not sufficiently explain 6D values measured in rainfall across the Maritime Continent (30–33). These studies indicate that 6D values of rainfall in equatorial Indonesia are a function of both local rainfall amounts and the rainfall history of monsoonally advected moisture from adja- cent tropical regions (30, 31). This includes latitudinal transport of air parcels across the Indian Ocean and the Pacific through Walker circulation, which may result in progressive isotope depletion of rainfall with increasing distance from the primary site of evaporation (G. J. Bowen, Gridded maps of the isotopic composition of meteoric precipitation, 2009, available at www.waterisotopes.org). As a result, rainwater 6D at a given site may reflect both the amount of local rainfall (“amount eff- ect”) as well as the rainfall history of the air parcel (30, 31, 33) (see also Bowen online maps, cited above). However, changes of moisture sources resulting from the seasonal migration of the ITZ do not correspond to a seasonal cycle in rainwater 6D collected in NW Sumatra (31), indicating that the amount effect exerts dominant control on the 6D value of rainfall in the study area. We therefore interpret changes in alkanolic acid 6D to primarily reflect changes in the amount of rainfall across the catchment area of our study site.

**Influence of Vegetation.** Since there is debate about whether photosynthetic pathway and plant morphology have an influence on the 6D value of plant waxes (34), we also measured their 6D values during periods of high 6D variability to control for changes in photosynthetic pathway (35). Values of 6D for the n-C30 alkanolic acid from the study site (Fig. 2C) show little variation between about −30‰ and −33‰ with slightly more 13C depletion during the LGM. Analyses of air bubbles trapped in an Antarctic ice core (36) indicate that levels of atmospheric 13C during the last glacial were between 0.3‰ and 1‰ lower than during the Holocene. The observed range of 3‰ therefore must partly be attributed to changes in atmospheric 6D. Pal- yological studies from the highlands of NW [Lake Toba (37) and central Sumatra [Danau Padang and Danau di Atas (38, 39)] provide evidence for persistent dominance of tropical trees (C3 plants) throughout the last 24 ka (summarized in ref. 40). We therefore consider the remaining change of about 2‰ in the n-C30 alkanolic acid 6D record as too minor to reflect signifi- cant changes in the ratio of C3/C4 vegetation, the more so as the physiological range in the 6D value of tropical tree leaf waxes comprises at least 4‰ (41). We further note that changes other than the ratio of C3/C4 plants such as changes of canopy closure may account for considerable variability of 6D [about 6‰, canopy effect (42)] among C3 plants. Our primary conclusion from the carbon isotopic evidence, however, is that we can rule out wholesale ecosystem shifts, and thus hydrogen isotopic varia- bility is likely to reflect hydrological changes. This conclusion is further supported by the rather weak correlation of 6D and 6C of r = 0.16.

11. Kusch S, Rethemeyer J, Schefuß E, Mollenhauer G (2010) Controls on the age of plant waxes transported from the Australian continent (dom- inantly C3 plants). In summary, we therefore infer that higher plant waxes deposited at the study site derive predominantly from the adjacent hinterland of Northwest Sumatra.

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Fig. S3. Temperature functions applied for the correction of the $n$-$C_{30}$ alkanoic acid δD record. Blue (orange) line depicts a deglacial warming of 3 °C (7 °C).

Fig. S4. Residual isotope records from Borneo (1) and Sulawesi (2) after detrending (3) to reveal precipitation changes on central Indo-Pacific Warm Pool rainfall other than those induced by sea level rise.