Aerosol forcing of intertropical convergence zone position since AD 1550

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Intertropical convergence zone position is the dominant control on low-latitude precipitation distribution and is largely controlled by hemispheric temperature contrasts[1,2]. Recent modelling[1,3,4] and observational[5,6] studies suggest that anthropogenic aerosols may have contributed to southward inter tropical convergence zone shifts by moderating Northern Hemisphere relative to Southern Hemisphere warming[1,7,8]. Despite this abundant evidence suggesting that recent hemispheric temperature contrasts affected low latitude rain belts, differentiating between anthropogenic forcing and century-scale natural variability is problematic and requires a record with little chronological error and very high temporal resolution. Unfortunately, these types of records are extremely uncommon in tropical regions affected by the intertropical convergence zone. Here, we

use an exceptionally well-dated and monthly-resolved 456 year-long stalagmite record from Belize to demonstrate that unprecedented rainfall decreases coincided with increasing anthropogenic aerosol emission rates. The record also suggests that short-lived drying occurred after large Northern Hemisphere volcanic eruptions since 1550. These results strongly suggest that aerosol injections into the Northern Hemisphere atmosphere result in southward intertropical convergence zone repositioning, and firmly implicate anthropogenic aerosol emissions as driving 20th Century rainfall reductions in the northern tropics. Future predictions of regional susceptibility to severe rainfall shifts should therefore consider geographic changes in aerosol emissions.

Intertropical Convergence Zone (ITCZ) position largely controls low latitude seasonal rainfall distribution. Relative ITCZ position is strongly influenced by hemispheric temperature contrasts and subsequent atmospheric restructuring, which draw the ITCZ toward the warmer hemisphere[1,2,5]. Indeed, considerable proxy evidence links Northern Hemisphere (NH) temperature to low-latitude rainfall throughout the Holocene[9,10,11]. However, since 1900, limited instrumental evidence suggests a southward shift in ITCZ position[3,5], a trend possibly driven by asymmetrical hemispheric warming due to the relative cooling effect of anthropogenic sulphate aerosols in the NH, but that could also arise from undetected natural variability. Climate models have attempted to assess the relative contributions of greenhouse gases (GHG) and aerosols to ITCZ displacement with contradictory results[1,7]. Limited long-term instrumental climate records from low latitudes complicates detecting climate shifts attributable to anthropogenic influences, and consequently future precipitation projections remain ambiguous[12]. Furthermore, chronological uncertainties associated with low latitude rainfall proxy records prevent establishing robust links between low-latitude rainfall amount and atmospheric aerosol distributions at a suitable resolution. Here, we discuss an exceptionally

well-dated, monthly-scale stalagmite rainfall record covering 456 years from 1550 to 2006 C.E., thus covering the critical transition into the Current Warm Period (CWP) with unprecedented detail and providing much needed evidence to support modelling work.

Stalagmite YOK-G was obtained from Yok Balum Cave in southern Belize (16° 12' 30.780" N, 89° 4' 24.420" W; 336 m.a.s.l.) (Supplementary Fig. S7). This site is near the northernmost extent of the ITCZ, a remarkably sensitive location for reconstructing even minor variations in ITCZ position. The cave was undisturbed prior to 2005 and is characterised by a stable low-pCO_2 atmosphere, consistent year round temperatures (22.3°C ± 0.5), and high relative humidity (>95%) (Supplementary Fig. S17 and S18). The cave is remote and located below steep, dense forest that is unsuitable for farming or mechanised logging, minimising potential past human interferences at the site. Outside air temperature only varies between 20°C (December through February) and 24°C (June through August). However, rainfall is distinctly seasonal, ranging from 40-70mm per month in the peak dry season (February through April) to 400-700mm per month during the peak wet season (June through September) due to seasonal ITCZ and associated trade wind migrations that track the thermal equator[13]. Evapotranspiration surpasses precipitation during the dry season[14], reducing effective rainfall and water input to the karst system. Stalagmite YOK-G was collected in 2006 and is 1090mm tall, but only the top 365mm are discussed here. 3648 carbonate samples were collected by milling continuously at 100μm increments along the central growth axis, and carbon and oxygen stable isotope ratios were determined using a Thermo MAT 253 gas source mass spectrometer.

Annual carbon isotope ratio ($\delta^{13}C$) cycles apparent throughout most of the record provide exceptional chronological control. The uppermost 8mm milled at a 100μm spatial resolution did not reveal $\delta^{13}C$ cycles, which prevented counting cycles back from the date of collection. The

$\delta^{13}C$ cycle chronology is instead anchored to the first evidence of atmospheric 'bomb' radiocarbon in 1955 (Supplementary Information and Fig. S14). Higher resolution (25 μm; weekly-scale) re-milling over the top 8mm also failed to detect $\delta^{13}C$ cycles (Supplementary Fig. S12), strongly suggesting that no $\delta^{13}C$ cycles exist in the most recent part of the stalagmite. If 2006 is used as the cessation of sample growth (due to collection), the calculated growth rate for this interval deviates significantly from the nearly uniform growth rate for the preceding ~500 years. This suggests that either: a) carbonate precipitation slows down at some point since 1984, b) the sample stopped growing earlier than the date of collection, or c) a combination of both a and b. This short interval (from 1984 to 2006) is therefore not included in the discussion due to increased chronological uncertainty. XRD results indicate that YOK-G is entirely aragonitic, which, due to its high capacity for uranium inclusion, permits the construction of a precise ^{230}Th chronology (Fig. 1; Supplementary Table S1). Eighteen high precision MC-ICP-MS ^{230}Th dates confirm that the $\delta^{13}C$ cycle-derived model is robust (Fig.1). Between 1550 and 1983 C.E. YOK-G grew continuously with a mean growth rate of 0.82mm a^{-1}.

Here we utilise the YOK-G $\delta^{13}C$ record as a palaeorainfall proxy. Stalagmite $\delta^{13}C$ in low latitude regions not experiencing temporal shifts in vegetation type (e.g., shifts from C3 to C4 vegetation) largely reflects effective rainfall amount and the hydrology of the drip feeding the stalagmite. Dry intervals promote: a) prior carbonate precipitation (due to lower groundwater flow rates), b) increased bedrock carbon contributions, and c) reduced soil bioproductivity, all contributing to a more positive $\delta^{13}C$. Conversely, wetter conditions result in more negative $\delta^{13}C$ (see Supplementary Information). This interpretation is supported by the remarkable, demonstrably annual $\delta^{13}C$ cycle reflecting seasonal water recharge conditions, as well as by interpretations of other Belizean stalagmite $\delta^{13}C$ records as reflecting rainfall, notably Frap-

pier et al.[15], linking pronounced $\delta^{13}C$ increases to El Niño related rainfall reductions, and Webster et al.[16] linking $\delta^{13}C$ shifts over the last 3,300 years to rainfall. We note that these two studies represent the two published speleothem records from cave sites closest to Yok Balum cave (ATM Cave, ~100km to the north, and Macal Chasm, ~80km to the north), and that both utilised $\delta^{13}C$ as a palaeorainfall proxy (Supplementary Fig. S4). The similarity between the YOK-G $\delta^{13}C$ record and the Cariaco Basin record ITCZ rainfall record[10] also strongly supports this interpretation. We stress that $\delta^{18}O$ is also an extremely useful complementary rainfall proxy (see Supplementary Information), but we believe that under the conditions at our site, $\delta^{13}C$ is more sensitive to subtle shifts in recharge.

Both wet and dry season $\delta^{13}C$ values ($\delta^{13}C_{wet}$ and $\delta^{13}C_{dry}$) are clearly distinguishable in the YOK-G record (Fig. 1c), providing a rare opportunity to isolate rainfall amount during specific seasons at a low latitude site. YOK-G $\delta^{13}C_{wet}$ and the NINO3.4 Center of Action (COA) sea surface temperature (SST) reconstruction[17] are anticorrelated ($r = -0.3$, $p < 0.001$ with a nine-year moving average applied) during the preindustrial period (1550-1850), suggesting that eastern equatorial Pacific SST exerted a significant control on Belizean rainfall (Fig. 2a). Additionally, a weak but significant negative relationship ($r = -0.19$, $p < 0.001$) exists between the Esper Northern Hemisphere Temperature (NHT) reconstruction[18] and $\delta^{13}C_{wet}$ during the preindustrial interval of the record (Fig. 2c). This suggests a warmer NH tends to draw the ITCZ to a more northerly position, consistent with the results of numerous previous studies[7,10]. No relationship exists between $\delta^{13}C_{dry}$ and NHT ($r = 0.05$, $p = 0.43$), again consistent with the interpretation of YOK-G $\delta^{13}C_{wet}$ as an ITCZ rainfall proxy. Elevated NHT tended to cause a more seasonal rainfall distribution (greater seasonality) during the preindustrial portion of the YOK-G record ($r = 0.32$, $p < 0.001$ with nine-year moving average applied) (Fig. 2b).

However, post-1850 all the $\delta^{13}C$ data (mean annual, wet season, and dry season) strongly suggest a steady drying trend coinciding with increasing NHT, suggesting a dramatic reversal in the relationship between NHT and ITCZ position (Fig. 3). Additionally, post-1850 YOK-G annual mean $\delta^{13}C$ tracks trends in global GHG concentrations and anthropogenic aerosol emissions (Fig. 4). This indicates a southward ITCZ migration despite increasing NHT.

The timing of this relationship reversal suggests an anthropogenic link. Recent research highlights the competing effects of GHG and anthropogenic aerosols on low latitude rain belts, with GHG increases believed to force the ITCZ to the north, and aerosols to the south[5,7]. Modelling studies suggest that a heterogeneous regional cooling effect induced by NH mid-latitude anthropogenic aerosol emissions drove the southward migration of the ITCZ over recent decades[1,3,4,7], leading to drought in the Sahel[8,19] and parts of monsoonal Asia[20,21]. The rainfall decreases implied by the YOK-G record closely follow patterns of regional industrialisation and aerosol emissions in North America and western Europe since ~1880 (Figs. 4 and Supplementary Fig. S24). Peak US aerosol production during the period 1970-1990 is estimated to have had a direct radiative forcing of -6 Wm^{-2} over the central and eastern US resulting in relative cooling of 0.5-1.0$^{\circ}C$[22]. Cooling over the North Atlantic region modifies atmospheric circulation to accommodate cross equatorial thermal contrasts and subsequently drives the ITCZ southward[23].

The YOK-G record also illustrates that very similar ITCZ repositioning occurred following large NH volcanic eruptions that injected sulphate aerosols into the atmosphere. These affected the ITCZ through a similar mechanism as anthropogenic aerosols, causing preferential NH cooling, southward ITCZ migration, and consequently drying in Belize. Particularly noteworthy is the coincidence of the large and climatologically significant Laki eruption (1783-1784) with the height of the largest preindustrial drought in Belize since 1550 C.E., evident in both

the YOK-G and local historical records. The Laki eruption produced a peak estimated direct radiative forcing in August 1783 of -5.5 Wm^{-2} in the NH[24], similar to the magnitude of the anthropogenic aerosol peak during 1970-1990 (-6 Wm^{-2}), and resulted in comparable drying in Belize. However, we note that the direct climate effects attributable to the Laki eruption were unlikely to have lasted more than three years[24], so the 1783 eruption may have exacerbated or prolonged the 1765-1800 drought but was not the principal driver. Southern Hemisphere (SH) volcanic eruptions, including those at low southerly latitudes, appear to force the ITCZ to the north. Most notable of these is the Tambora eruption in 1815, associated with increased Belizean rainfall the following year (Fig. 4). Of the nine largest NH eruptions identified in the GISP2 ice core sulphate record and the historical record since 1550[25], all are associated with drying in Belize; conversely, all three large SH eruptions are associated with increased rainfall at our site. Specifically, the YOK-G record indicates that NH eruptions result in substantially elevated $\delta^{13}C_{dry}$, and we suggest that this reflects a longer dry season caused by delayed onset of the summer wet season. Our data suggest that NH eruptions shortened the duration of the wet season, and SH eruptions extended wet season duration. The record provides compelling evidence that stratospheric sulphate aerosol injections associated with explosive volcanism resulted in short-lived ITCZ migration (Fig. 4). This result is consistent with recent modelling results suggesting that large volcanic eruptions that inject aerosols into the NH cause the ITCZ to migrate to the south, whereas SH eruptions push the ITCZ to the north[26], and with historical records suggesting reduced Nile discharge following the 1783 Laki eruption[27]. Similarly, continuous NH anthropogenic aerosol emissions during the 20th Century drove sustained southward ITCZ repositioning.

The monthly-resolved YOK-G $\delta^{13}C$ rainfall record provides the strongest proxy evidence currently available that recent droughts in the northern tropics are attributable to extra-tropical anthropogenic forcing. Rather than being a cyclic natural phenomenon, sustained rainfall re-

ductions in Belize only occurred after atmospheric aerosols increased following regional industrialisation in the NH. The record also indicates that similar (albeit shorter lived) ITCZ repositioning occurred in response to sulphate aerosol forcing associated with large NH volcanic eruptions. Future modelling should focus on determining how shifts in regional aerosol emission rates might affect ITCZ position. This is particularly relevant to currently industrialising regions where large populations are dependent on seasonal rainfall.

Analytical Methods

Stable isotope analysis

Stalagmite YOK-G was continuously milled at 0.1mm steps along the central growth axis using a computer-controlled ESI/New Wave Micromill with a standard 0.8mm tungsten carbide drill bit. Samples were milled to specifications of 0.1mm width, 10mm length and 1mm depth, with the long axis parallel to growth layers. 220µg - 250µg of powder were used for stable isotope analysis, conducted using a Thermo-Finnigan (Bremen, Germany, now Thermo Fisher Scientific) MAT 253 Isotope-Ratio Mass Spectrometer with a Gasbench II (external precision of ca. 0.05-0.10‰). About fifty speleothem carbonate samples were loaded for each run. Carbonate samples were dissolved in 10 drops of orthophosphoric acid (H_3PO_4) under helium (grade 5) atmosphere. The solution was left to digest at 50˚C for two hours. The resultant gas mixture (CO_2 and He) was introduced to a gas chromatographic column and the CO_2 separated from the gas mixture. After passing the second water trap the analyte was admitted to the mass spectrometer. Each sample run included 14 reference standards of IAEA international reference materials NBS18 (carbonatite), NBS19 (limestone) and LS VEC (lith-

ium carbonate) and an additional internal laboratory standard. Normalisations and corrections were made to NBS19 and LS VEC. $^{13}C/^{12}C$ ratios are reported relative to the VPDB standard.

Chronology

Uranium-series (^{230}Th) dating of powders was performed at the University of New Mexico Radiogenic Isotope Laboratory. Milled powder samples (20-120 mg) and a drip water sample (13.3 g) were spiked with a solution containing ^{229}Th, ^{233}U, and ^{236}U. Uranium and thorium were separated using anion chemistry and analysed on a Thermo Neptune multi-collector inductively coupled plasma mass spectrometer. Improved half-life values for ^{230}Th and ^{234}U were applied to these age results (See SOI). Drip water and YOK-G carbonate powders extracted at 0.5 and 1.0 mm from the stalagmite top (years AD 2006 and 2004) were used to establish that stalagmite YOK-G detrital thorium has a high initial $^{230}Th/^{232}Th$. This high initial value was found to be inversely correlated to ^{232}Th concentration such that initial $^{230}Th/^{232}Th_{atomic}$ (ppm) = 323.67(X) - 0.269, where X = concentration of ^{232}Th in pg/g, and the calculated initial $^{230}Th/^{232}Th_{atomic}$ (ppm) has an assumed ± 10% absolute error. These high initial values result in higher absolute errors on ^{230}Th ages but do not significantly affect age results older than AD 1800.

Incremental AMS ^{14}C on the upper 48mm of YOK-G was performed to help constrain growth rates over the last 50 years. Samples were milled along visible growth increments offset from the stable isotope milling track. AMS ^{14}C analyses were made on a modified National Electronics Corporation compact spectrometer and all ^{14}C ages were corrected for mass dependent fractionation. Identification of the initial part of the atmospheric bomb spike at AD 1955 provided both an independent confirmation of the accuracy of the top ^{230}Th dates and a date (1955) with which to anchor the annual $\delta^{13}C$ cycle chronology. Carbon cycles were

counted from this marker. Within the carbon cycles, higher $\delta^{13}C$ values correspond with the end of the dry season (April) when the karst is most extensively drained and lower $\delta^{13}C$ values represent the peak of the wet season (July). The carbon cycle chronology is consistent with the ^{230}Th chronology and is never outside the errors associated with any particular ^{230}Th date. The $\delta^{13}C$ cycles therefore provide an accurate sub-seasonally resolved chronology.

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Author contributions

HER produced the stable isotope record with assistance from CGM and JLP. YA, VP and VVA were responsible for developing the uranium-series chronology. DJK and BJC performed radiocarbon analyses. HER, KMP, and JULB contributed significantly to fieldwork. MZ and TX performed GHCN analyses. FAL performed high resolution stable isotope reanalyses. HER and JULB wrote the manuscript. JA contributed to logistics associated with fieldwork. All named co-authors contributed to the project, discussed manuscript ideas, and approved the final manuscript.

Figure captions

Figure 1. YOK-G $\delta^{13}C$ record and chronology. a, ^{230}Th dates with errors (black line) and $\delta^{13}C$ cycle chronology (red line) fitted with cubic splines. Shaded grey boxes indicate intervals where the $\delta^{13}C$ cycles are present but less clear. The shaded pink box indicates the interval (1983-2006) where $\delta^{13}C$ cycles are absent. ^{230}Th dates verified the $\delta^{13}C$ cycle count chronology, but are independent of the final chronological model. **b,** The entire YOK-G $\delta^{13}C$ record against depth. **c,** An expanded view of 40 mm of growth illustrating $\delta^{13}C$ annual cycles with peak wet ('W') and dry ('D') season $\delta^{13}C$ values identified.

Figure 2. YOK-G $\delta^{13}C$ record. a, YOK-G $\delta^{13}C_{wet}$ record and the Niño 3.4 COA reconstruction[17] for the period 1550 to 1850. **b,** Seasonality defined by the amplitude of each annual $\delta^{13}C$ from peak wet season to peak dry season and Esper NHT[18] for the period 1550-1850. **c,** YOK-G $\delta^{13}C_{wet}$ against NHT for the period 1550-1850. **d,** as in **a** but for the industrial interval of the record, 1851-1983. **e,** as in **b** but for the period 1851-1983. **f,** as in **c** but for the period 1851-1983.

Figure 3. Scatterplot of YOK G $\delta^{13}C_{wet}$ versus Esper NHT[18]. During the preindustrial period (1550-1849) (unfilled circles), showing weak significant negative correlation ($r = -0.19$, $p < 0.005$), and during the CWP (1850-1983) (black filled circles), which exhibits a switch to a significant positive correlation ($r = 0.43$, $p < 0.001$).

Figure 4. Annual mean YOK-G $\delta^{13}C$ and links to sulfate aerosols. a, Annual mean $\delta^{13}C$ (black) and GISP2 total sulphate record (blue)[25]. Aerosol production estimated using CO_2 emissions relative to 1992 levels[28,29] for Europe (green) and North America (yellow). Major (VEI≥5) NH eruptions (red) and SH eruptions (blue) are marked. Dashed lines designate conventionally accepted eruption date. (*) highlights VEI≥6 eruptions. **b,** Climate response to NH and SH eruptions implied by YOK-G $\delta^{13}C$ values (normalised to monthly means in the year preceding the eruption). Shading denotes the one sigma standard deviation from the monthly mean values over the entire record. Thick lines represent mean $\delta^{13}C$ response to NH and SH eruptions.