# Monsoon reconstruction from radiocarbon dated tropical Indian speleothems

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Abstract: The potential of tropical speleothems as a climate proxy has been investigated. Amplitudes of  $\delta^{18}O$  and  $\delta^{13}C$  variations are found to be large and are likely to be primarily controlled by past rainfall. Contribution from past temperature variations seems to be relatively small. The 'amount effect' in rainfall has been observed and quantified by analysing rainwater samples collected during a monsoon season. A tentative chronology to these speleothems is assigned by the <sup>14</sup>C radiometric dating method. Assuming that the variations in the  $\delta^{18}O$  of cave carbonates are solely due to the past variations in rainfall, a history of the latter has been reconstructed. A high-resolution rainfall reconstruction up to the last ~3400 years is now available from Gupteswar cave, Orissa, subject to validation of dates by the U-Th method. It is observed that in a tropical speleothem  $\delta^{13}C$  is dominantly controlled by rainfall. The study has shown that tropical Indian speleothems faithfully record the annual ( $\approx$  monsoon) rainfall in the cave site.

Key words: Speleothems, radiocarbon, stable isotopes, oxygen isotopes, carbon isotopes, rainfall reconstruction, southwest monsoon, rainfall, India, late Holocene.

#### Introduction

It is important to document past climatic variations: first, to assess the natural variability in climate on different timescales; and, secondly, to provide a data base to serve as boundary conditions for climate models aimed at long-term monsoon prediction. In India, meteorological observations of temperature and rainfall are available for not more than a century, and then only in selected metropolitan cities. For older periods, and other nonurban centres, one has to use natural climate proxies. Even though palaeoclimate data available to date reveal that past monsoon rainfall was significantly different from that in the present, cave deposits occurring in limestone terrains have not been explored so far as potential recorders of past climate. Secondary calcite deposits formed in limestone caves, commonly known as speleothems, are very important proxies for terrestrial palaeoclimate. Their stable isotope compositions (carbon and oxygen) and trace elements hold the potential for the reconstruction of past cave environments (e.g., Hendy, 1971; Gascoyne, 1992; Roberts et al., 1998). During CaCO3 precipitation in the deeper parts of a cave, stable isotope ratios of oxygen ( $^{18}O/^{16}O$ ) and carbon ( $^{13}C/^{12}C$ ) in the  $HCO_3^-$  ions in the dripping water are affected by the cave environment. These effects remain preserved in the CaCO3 laminae, which can be used to decipher the past climate. We have undertaken a study of tropical Indian speleothems to evaluate their paleoclimatic potential.

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# **Methods**

#### Sample locations

Two actively growing speleothems from different caves located  $\sim 30 \, \mathrm{km}$  apart, Gupteswar and Dandak (Figure 1), were collected in 1996 (for details, see Yadava and Ramesh, 1999a). The Gupteswar cave (18°45′N, 82°10′E) is in the Koraput district of Orissa state. A stalactite hanging in the passage at about 100 m distance from the cave entrance was collected from this cave in February 1996 (winter season).

The other cave, Dandak (19°00'N, 82°00'E), in the Kanger Ghati National Park, west of the Gupteswar, is in the Jagdalpur district of the Chhattisgarh state (Figure 1).

Presently, the vegetation is dense and is of C3 type on both caves, which form a part of the western foothills of the northeast—southwest trending ranges. The important rock types in the area are limestones, purple shale and quartzite.

Meteorological data from the Jagdalpur district (nearest station to the cave sites) show that the average (30 year: between 1931 to 1960) *monthly* rainfall during the wet season (June to October) is 270.4 mm, average annual rainfall is 1534.1 mm and the mean annual temperature is 25.5°C (Climatological Tables, 1960).

# Analytical methods

Subsamples of about 5 mg were collected from all distinct layers in both the speleothems for stable isotope study (Figure 2). These subsamples were prepared using stainless steel slow-speed drill bits with 0.8 mm diameter. Each powdered subsample was reacted with 100% H<sub>3</sub>PO<sub>4</sub> at 50°C

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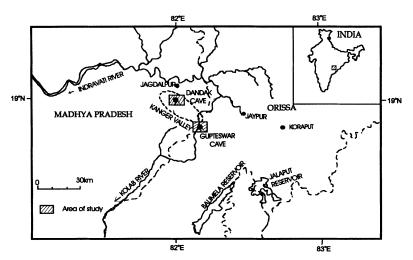


Figure 1 Map showing locations of the two caves Gupteswar (Orissa state) and Dandak (Madhya Pradesh state, now in the new state of Chhattisgarh).

(McCrea, 1950) for 10 minutes to get CO<sub>2</sub> which was analysed by a VG 903 mass spectrometer with an overall precision of  $\pm 0.1\%$  for  $\delta^{13}$ C and  $\pm 0.15\%$  for  $\delta^{18}$ O. A laboratory marble standard (MMB) was run after every 10 measurements to check the reproducibility and accuracy of the measurements. All the isotopic values are reported relative to the V-PDB standard.

Rainwater samples were analysed for  $\delta^{18}$ O by the procedures of Epstein and Mayeda (1953). All values are reported relative to V-SMOW and the analytical uncertainty was  $\pm 0.1\%$ .

About 3-7 g of carbonate were taken from along the growth axis of the speleothems for radiocarbon dating to establish the chronology. Benzene was synthesized from these subsamples which was further assayed for residual activity in the liquid scintillation counter (Quantulus). The overall precision in the dating method is about ±90 yr. Further details of experimental procedures on stable isotope measurements and radiocarbon dating are given in Yadava and Ramesh (1999b).

# Results and discussion

#### Stable isotopes

A total of 233 stable isotope measurements were carried out on the Gupteswar stalactite (shown by dots in Figure 2A). Four laminae representing the sample growth were chosen from the Gupteswar stalactite to test isotopic equilibrium during deposition (Hendy, 1971; Gascoyne, 1992) and the results are presented in Figure 3.  $\delta^{18}O$  and  $\delta^{13}C$  relative to V-PDB of all the subsamples are shown in Figure 4 as a function of depth. The average fluctuation observed from the modern value is about 1.5% for  $\delta^{18}$ O and 3.5% for  $\delta^{13}$ C.

For stable isotope measurement 117 subsamples were prepared from the Dandak stalagmite (shown by dots in Figure 2B). Three representative layers were chosen for testing isotopic equilibrium during deposition, and  $\delta^{13}$ C,  $\delta^{18}$ O values relative to V-PDB are shown in Figure 5.  $\delta^{18}O$  and  $\delta^{13}C$ of all the subsamples as a function of depth are shown in Figure 6. About 2% variations in  $\delta^{18}$ O and up to about 4‰ in  $\delta^{13}$ C relative to the modern value are observed in the stalagmite.

# Radiocarbon dating

The <sup>14</sup>C age of the speleothem is greater than that of organisms deriving carbon from the atmosphere. This is due to contribution of 14C-free carbon from the leached carbonate bedrock (reservoir effect; Mook, 1980).

In the case of an actively growing speleothem at the tip, the actual age should be 0 yr. However, measurement shows an apparent age > 0. Assuming that, during the past, the dead carbon contribution from the bedrock remained constant, the actual ages of the speleothem layers are obtained by simply subtracting the tip age from the apparent ages (Tables 1 and 2). A similar assumption was made by Williams et al. (1999). The best test for the application of a dating method is the verification of ages in stratigraphic order. All the ages listed in Tables 1 and 2 increase from the tip to the base. There are some exceptions; for example, in the Gupteswar stalactite (Table 1) ages for PRL-1999 and PRL-1992, and in the Dandak stalagmite (Table 2) PRL-1993 and PRL-1997, are indistinguishable. The reasons could be: (i) small changes in the dilution factor for the associated layers; (ii) the growth rate could have been very high so that the two dated patches are deposited within the period equivalent to the age uncertainty ( $\sim \pm 130$  yrs) and (iii) natural variations in the atmospheric <sup>14</sup>C production. Further details of the chronology of speleothems are discussed below.

#### Gupteswar stalactite

Assuming that during the growth of the stalactite the dead carbon contribution from the bedrock strata was constant, all the radiocarbon ages (Table 1) are corrected by subtracting the surface age (880  $\pm$  100 yr).

By considering a constant deposition rate between two consecutive dating patches (centre of the laminae analysed), successive growth layers were assigned with equivalent ages without considering the associated uncertainties. All the ages are in the stratigraphic order except PRL-1992 (2900  $\pm$  130) and PRL-1999 (2920  $\pm$  130), which are in agreement within errors. From the <sup>14</sup>C calibration curve (Stuiver and Becker, 1998) for 2900  $\pm$  130 yr BP, the associated calendar age range is 2750 to 3230 yr BP. The actual age of PRL-1992 and PRL-1999 could be in the above range. To simplify the calculation of deposition rate, we have assumed all the growth layers between the two dated patches deposited in the duration of 130 yr (error in the apparent ages of PRL-1992 and 1999). Ages to the subsamples taken for the stable isotope study have been assigned accordingly. The average deposition rate is 0.14 mm/yr.

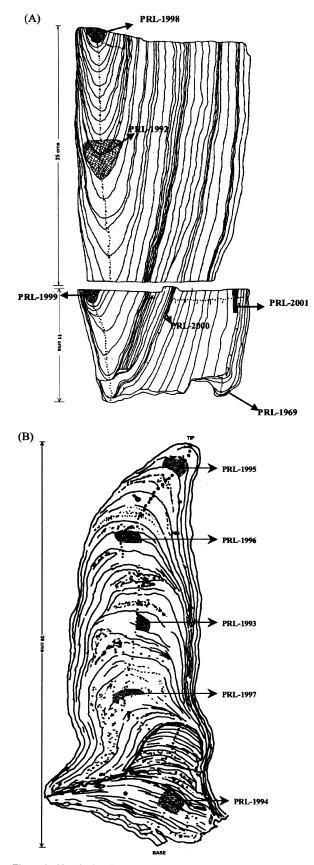


Figure 2 Sketch showing sampling spots for the subsamples for the stable isotopes studies (by dots) and for the radiocarbon dating (by patches) for (A) the stalactite from Gupteswar cave and (B) the stalagmite from Dandak cave.

#### Dandak stalaomite

Considering only the recent (first three) ages, i.e., 1310, 1640 and 1970 yr, an extrapolation gives the surface age to be 1230 yr. As discussed previously, the surface age has been subtracted from other measured ages to give corrected ages for the stalagmite. Radiocarbon ages increase in the older layers except PRL-1993 (740 yr) and PRL-1997 (710 yr), which are the same within the associated uncertainties. This is possible due to a higher growth rate during the period. However, as there is no other date between the hiatus and PRL-1997, ages cannot be assigned assuming uniform growth rate between two dating spots (as was done for the Gupteswar stalactite). The average growth rate estimated based on a best-fit line passing through first three ages (0, 410 and 740 yr) is 0.18 mm/yr. Assuming this to be the average growth rate, the depth of each stable isotope subsample is assigned an age. The texture of the layers observed along the growth direction shows a sudden change in the lower part due to the hiatus (Figure 2B) in the growth of the stalagmite. Also, above the hiatus layer, coarse detrital material is present. As a similar event is not observed in the Gupteswar stalactite, the hiatus in the Dandak stalagmite was probably caused by a change in the flow pattern of the seepage water (a site-specific effect), and not by onset of aridity. All subsampling points for stable isotope measurements beginning from the modern surface (tip) have been assigned ages assuming a uniform growth rate between two dating spots in the sample. Assuming a uniform growth rate (0.18 mm/yr) and using the age of PRL-1994 (3540 yr), the beginning of the hiatus is estimated to be 3200 yr BP and this continued till 1230 yr BP. Hence, the hiatus period prevailed for about 2000 yr. This may not have a climatic significance if it was due to a change in the micro-environment.

A combined plot of  $\delta^{18}O_c$  and  $\delta^{13}C_c$  versus age for Gupteswar and Dandak caves is shown in Figure 7. As the  $\delta^{18}O$  in the two speleothems have concordant variations within the 0-1200 yr BP period, it suggests that the radiocarbon chronology is reasonably correct in this period. However, in the older part of the record (3700-3200 yr BP), this similarity breaks down, indicating that the assumption of constant dead carbon contribution from the parent bedrock is probably not valid during this period.

# <sup>14</sup>C chronology: a tentative estimate

Currently, the U-Th mass spectrometric method is being used to provide precise chronology to speleothems, but besides being expensive it is available only to limited research groups. Although radiocarbon dating is not the most appropriate method to provide chronology due to unknown variations in the dead carbon content, it can provide tentative age estimates if the time duration covered is not very long, e.g.,  $\sim 3700$ years in the present case, under certain simple assumptions. A comparative list of <sup>14</sup>C and U-Th ages available together in speleothems from recently published work (Genty et al., 1999; Williams et al., 1999) is presented in Table 3. Holmgren et al. (1994) have shown that there is discrepancy between <sup>1</sup> and U-Th ages; however, it is small during the late Holocene period (~last 5 kyr). Based on <sup>14</sup>C activity measurements and U-Th (TIMS) ages, Genty et al. (1999) have shown that the dcp (dead carbon proportion) in speleothem had remained reasonably constant for two Holocene speleothems from Belgium and France. An excellent correlation ( $r^2 = 0.98$ ) was found between U/Th ages and the calibrated 14C ages, assuming a constant dead carbon contribution. However, uncertainties in the dcp estimates (~1.5%; Genty and Massault, 1997) contribute to an error in <sup>14</sup>C age, up to 250 years for the last  $\sim 3000$  years. Assuming  $1700 \pm 1000$  as

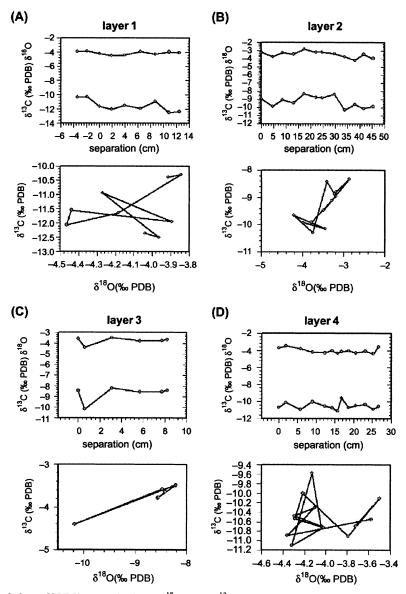


Figure 3  $\delta^{18}$ O and  $\delta^{13}$ C (relative to V-PDB) versus depth and  $\delta^{18}$ O versus  $\delta^{13}$ C for four layers (A-D) selected to test isotopic equilibrium during deposition in the Gupteswar cave.

reservoir age for several speleothems from caves in central and southern Europe and southwest Africa, Goslar et al. (2000) have shown reasonable correlation between <sup>14</sup>C and U-Th ages, at least for the recent ~8 kyr. However, Genty et al. (2001) have reported significant variations in the past values of the dcp (up to 9.5%, within a  $\sim$ 1400-yr period) for the stalagmites from Scotland. The radiocarbon ages we have used are uncalibrated, which may contribute to an uncertainty of  $\sim$  30 to 250 yr. In the light of such evidence, we feel that the chronology in the present work based on <sup>14</sup>C dating is tentative and it should be confirmed by U-Th mass-spectrometric measurements.

#### Oxygen isotopes

The speleothem sample collected has been checked for deposition under isotopic equilibrium, indicated by, along a single lamina, (1) same values of  $\delta^{18}$ O (an enrichment indicates precipitation due to evaporation of water) and (2) that there is no correlation between  $\delta^{18}O$  and  $\delta^{13}C$  (otherwise it is an indication of fast CO<sub>2</sub> degassing and hence kinetic fractionation;

Hendy, 1971; Gascoyne, 1992). For measurements made along some of the distinctly observable lamina, plots of  $\delta^{18}O$  and  $\delta^{13}$ C versus depth and  $\delta^{18}$ O versus  $\delta^{13}$ C show no linear correlation for either Gupteswar (Figure 3) or Dandak cave (Figure 5). This proves that during the growth of calcite layers, humidity was very high, and the degassing rate of CO2 from the dripping water was quite slow leading to isotopic equilibrium deposition. Hence, the Gupteswar stalactite and the Dandak stalagmite can be used to reconstruct past environmental variation.

The  $\delta^{18}O$  of speleothem calcite ( $\delta^{18}O_c$ ) has been interpreted as reflecting the mean isotopic compositions of the precipitation and mean annual temperature (e.g., Dorale et al., 1992; Gascoyne, 1992; Laurtizen and Lundberg, 1999). Any change in the annual temperature will affect the calcite δ<sup>18</sup>O<sub>c</sub> whereby heavier  $\delta^{18}O_c$  values imply lower temperature  $(d(\Delta_{cw})/dT=-0.21\%$  °C<sup>-1</sup> at 25°C, where T is the ambient cave temperature in  ${}^{\circ}K$ , and  $\Delta_{cw} = 10^{3} ln \alpha_{cw}$ , where  $\alpha_{cw}$  is the fractionation factor for the calcite-water system). In such a situation, if temperature inside the cave increases, then less

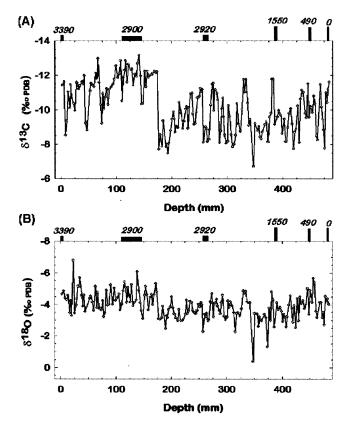


Figure 4  $\delta^{13}C$  (A) and  $\delta^{18}O$  (B) versus depth for measurements along the growth direction for the Gupteswar stalactite. Boxes show corrected radiocarbon ages.

<sup>18</sup>O gets incorporated into the precipitated calcite. Additional accounting for the temperature-dependent changes in the oxygen isotopic composition of meteoric water ( $\delta^{18}O_w$ ) is also required. For example, at subtropical and high-latitude regions  $\delta^{18}O_w = 0.695(t) - 13.6$ , where t is the mean annual surface air temperature in °C (i.e.,  $d(\delta^{18}O_w)/dT = 0.69\%$  °C<sup>-1</sup>; Dansgaard, 1964). There are local variations between 0.5 to 0.9%/°C, within 40°N to 60°N (Rozanski et al., 1993). Usually, the ambient temperature of a cave is approximately equal to the mean annual surface air temperature outside the cave and hence  $\delta^{18}$ O of the speleothem layers is a proxy for the past variations of  $\delta^{18}O$  of meteoric water and of mean annual surface air temperature in high latitudes. The temperature-δ<sup>18</sup>O relationship is empirically known for the high latitudes and can be used in some cases to retrieve the past temperature variations (Dorale et al., 1992).

The temperature sensitivity and therefore the interpretation of speleothem  $\delta^{18}$ O is by no means straightforward and often ambiguous (Lauritzen and Lundberg, 1999). The best way is to compare the speleothem  $\delta^{18}$ O trends with the instrumentally recorded past climate changes and then, assuming the same kind of climate response in the past, one can reconstruct climate history.

# $\delta^{18}O_{\omega}$ in tropical locations

For mid-latitude and semi-arid climate zones  $\delta^{18}O_w$  decreases with increasing rain amount (Dansgaard, 1964; Bar-Matthews et al., 1996; 1997; 2003; Ayalon et al., 1998; Fricke and O'Neil, 1999). In tropical locations any obvious temperature correlation is not observed for the modern rainfall (Dansgaard, 1964; Yurtsever and Gat, 1981; Fricke and O'Neil, 1999).  $\delta^{18}O_{w}$  is rather dependent on the amount of rainfall. More rainfall is associated with less of <sup>18</sup>O content in the precipitation. In most parts of India,  $\sim 90\%$  of the annual rain is received during the summer monsoon between June and September. In the region encompassing both caves, 88% of the rain is received during the summer monsoon. There is only a small contribution during other months whose isotopic signatures have not been investigated here. Hence, in these tropical caves, freshly deposited calcite layers on a growing speleothem should be depleted in <sup>18</sup>O with increasing precipitation and temperature.

A negative correlation between  $\delta^{18}$ O of precipitation and amount of rainfall is observed in the tropics, which is termed the 'amount effect' (Dansgaard, 1964). The effect is a manifestation of gradual saturation of air masses below the cloud base as rainout continues and also due to the preferential removal of <sup>18</sup>O as rainout continues. Initially, the air mass is undersaturated and this leads to evaporation of water droplets, and consequently they are enriched in  $\delta^{18}$ O. As precipitation proceeds, air masses become saturated and in turn the enrichment due to evaporation gradually diminishes (Dansgaard, 1964; Rozanski et al., 1993; Fricke and O'Neil, 1999).

Worldwide modern precipitation data available from island stations in the equatorial belt (collected by IAEA) have shown a linear relationship (Yurtsever and Gat, 1981) between the mean monthly δ<sup>18</sup>O<sub>m</sub> of precipitation and the mean monthly rainfall,

$$\delta^{18}O_{m} = (-0.015 \pm 0.002)P_{m} - (0.47 \pm 0.42) \tag{1}$$

where P<sub>m</sub> is mean monthly rainfall in mm, with a linear correlation coefficient (r) of 0.87 for 14 island stations (each has at least 40 monthly observations). As these are island stations, seasonal temperature variations are small. The average rate of depletion is found to be  $-1.5 \pm 0.2\%$  for a 100 mm increase in the mean monthly rainfall.

Strong amount effect observed in the southwest monsoon A major source of precipitation over India is the southwest monsoon, where the hot, dry air prevailing over India in April and May is replaced by moist, oceanic air coming from the southwest (Pant and Rupa Kumar, 1997). The mean rainfall over the Indian plains in the SW monsoon period is 925 mm as against 145 mm for the rest of the year. In order to verify the presence of amount effect in the monsoon rainwater, water samples from each major rainout spell between July and September (year 1999) were collected at Jharsuguda (22°N, 84°E), about 400 km north-east of Gupteswar. Rainwater was collected in a plastic carboy (20 litres) attached with a plastic funnel of known diameter (20 cm). It had a small neck to minimize evaporation. The amount of water (Si) was measured using a graduated cylinder and part of the water was used to fill completely a 25 ml airtight plastic bottle for isotopic measurement (δ<sup>18</sup>O<sub>i</sub>). The isotopic results are given in Table 4 and plotted in Figure 8. For the amount of water-S<sub>i</sub> and  $\delta^{18}O_i$  the least square fit obtained is:

$$\delta^{18}O_i = -(0.092 \pm 0.011)S_i - (3.4 \pm 0.4)$$
 (2)

where  $S_i$  is the amount (mm) of each rain spell, r=0.87(n = 24), and the average rate of depletion is  $-9.2 \pm 1.1\%$ per 100 mm.

The above data were used to calculate the mean monthly  $\delta^{18}O$  and monthly total amount by simple isotopic mass balance of all the collections in each month. An expression for the best-fit line obtained from three data points for July, August and September (shown in Figure 8) is given below:

$$\begin{split} \delta^{18} O_m &= -(0.022 \pm 0.008) P_m - (2.5 \pm 1.9) \\ r &= 0.94 \ (n = 3) \end{split} \tag{3}$$

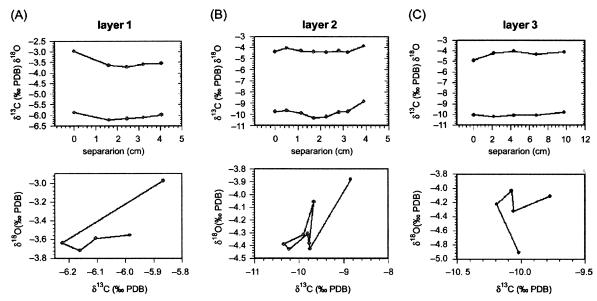


Figure 5  $\delta^{18}$ O and  $\delta^{13}$ C (relative to V-PDB) versus depth and  $\delta^{18}$ O versus  $\delta^{13}$ C for three layers (A-C) selected to test isotopic equilibrium during deposition in the Dandak cave.

This shows that there is  $2.2 \pm 0.8\%$  depletion per 100 mm increase in the monthly rain amount.

The observed value  $(2.2 \pm 0.8\%)$  depletion per 100 mm in monthly rain) agrees very well with the value within errors as shown by the IAEA stations  $(1.5 \pm 0.2\%)$  depletion per 100 mm monthly rain).

#### Past variations in $\delta^{18}$ O and $\delta^{13}$ C in speleothems

For the recent 1200-yr period  $\delta^{18}$ O in both caves (Figure 7) shows concordant variations (any apparent discordance is mainly due to the different sampling resolutions in the two samples). This is expected as the two caves are within 30 km distance and hence changes in surface air temperature or isotopic composition of the precipitation above the caves should have affected both the caves in a similar way.

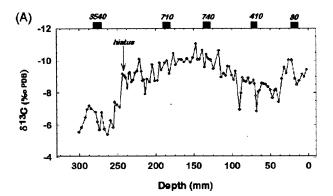
If the changes in  $\delta^{18}O_{c,}$  which are up to 1.5% for Gupteswar and 2‰ for Dandak excluding a few sharp peaks (about 2.5%), are considered to reflect only fluctuations in the past temperature then the estimate for the amplitude of temperature variation would be about 7°C. The  $\delta^{18}O_c$  variations in these speleothems are quite high and are unlike the highlatitude speleothem deposits (Talma and Vogel, 1992; Dorale et al., 1992) where such large changes are not observed. As during the past 3400-yr period temperature fluctuations of more than 1°C are highly unlikely (including the recent historical period, Mann et al., 1998), the varying  $\delta^{18}$ O of past precipitation should have been by and large responsible for the variations in calcite  $\delta^{18} O_c, \, provided \, \, there \, were \, no \, \, significant$ changes in the moisture source. Meteorological observations show that the major source of vapour for rainfall on the cave is the Bay of Bengal; however, a small part of it could also be from the Arabian Sea. Relative contributions from Bay of Bengal and Arabian Sea are governed by the relief features of the vast land area, such as the Himalayas, the Western and Eastern Ghats and the mountains of Myanmar, which are expected to change only in geological timescales. As the speleothems presented here cover ~3700 years, during this period the vapour source can be assumed to have remained

Dandak cave is located at  $\sim 50$  m higher elevation than Gupteswar cave. The higher elevation may not result in a significant difference in the annual rainfall received by them. The difference in the average annual rainfall (for the period 1931 to 1960: Climatological Tables, 1960) recorded between two nearby meteorological observatories with an elevation difference of 360 m, Koraput (1670 mm annual rainfall, 913 m above m.s.l.) and Jagdalpur, ~80 km away (1534 mm annual rainfall, 553 m above m.s.l.), is only 136 mm. Therefore, for both caves, present-day rainfall is assumed to be the same. Spatial separation being about 30 km, the  $\delta^{18}$ O of rainfall should also be the same for both caves.

An empirical relation relating the temperature  $(T = {}^{\circ}K)$  and  $\alpha_{c-w}$ , the fractionation factor between calcite and water (expressed as  $\alpha_{c-w} = (\delta^{18}O_{cal} + 1000)/(\delta^{18}O_{water} + 1000)$ , where  $\delta^{18}O_{cal}$  and  $\delta^{18}O_{water}$  are the calcite and water oxygen isotopic compositions respectively, is given as (Friedman and O'Neil, 1977):

1000 ln 
$$\alpha_{c-w} = 2.78(10^6 T^{-2}) - 2.89$$
 (4)

For the Dandak stalagmite, the recent layers near the tip have an average  $\delta^{18}O_{cal\text{-VPDB}}$  value (first three measurements) of -4.4%. Average  $\delta^{18}O_{water\text{-VSMOW}}$  value for Dandak cave, based on seepage water samples collected from several dripping spots during July 1999, September 1999 and June 2000, is  $-1.9 \pm 0.3\%$  (n = 26; Yadava, 2002). The estimated cave temperature, from equation (4), is between 27.5 and 29.2°C. During several visits to the cave, it was observed that the cave temperature varied between 25 and 29°C (during the wet season it was in the range of 25.5 to 25.4°C, whereas during the winter it was between 27 and 29°C). For the Gupteswar cave, the average  $\delta^{18}O_{water-VSMOW}$  is  $-2.8 \pm 0.3\%$  (n = 5). For the recent growth layers in the Gupteswar stalactite, the mean value of  $\delta^{18}$ O is -4.3%. The estimated present-day ambient Gupteswar cave temperature is between 22.6 and 24.2°C. Two cave temperature values measured during sample collections are 20°C (February; winter season) and 26°C (June; beginning of wet season). Considering the magnitude of temporal variations in the measured values of both the cave temperature and  $\delta^{18}O$  of drip water, the above calculation is not inconsistent with the inference that both the speleothems were growing very close to isotopic equilibrium.



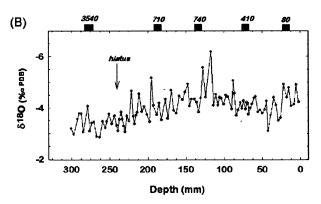


Figure 6  $\delta^{13}$ C (A) and  $\delta^{18}$ O (B) versus depth for measurements along the growth direction for the Dandak stalagmite. Boxes show corrected radiocarbon ages.

#### Rainfall reconstruction

Using speleothem  $\delta^{18}$ O and the present-day relationship between the variations in the average annual rainfall and the variations of  $\delta^{18}$ O in rain for the cave site, both rainfall and temperature have been reconstructed earlier for the Soreq cave (Bar-Matthews et al., 1997). The agreement between  $\delta^{18}O$  depletion in the monthly rainwater observed at the inland station, given by equation (3), and the island stations, equation (1), suggests that during the active monsoon period the vast land area around the Gangetic plains cools down and remains in a narrow range of temperature ( $\sim$ 23 ± 3°C). Therefore, subsequent changes in the  $\delta^{18}$ O of precipitation should be largely due to its amount dependence and temperature-induced changes should be insignificant. As most of the rainfall is received during the monsoon months, we assume that all the variation in the speleothem  $\delta^{18}O$  is due to changes in the  $\delta^{18}$ O of dripping water and the depletion rate observed for the precipitation is also applicable to the

Table 1 Details of the subsamples from Gupteswar stalactite and their  $^{14}\mathrm{C}$  ages

Sample	Depth from oldest part (mm)	Sample weight (g)	Apparent age (years BP)	Corrected age (years BP)
PRL-1969	483.5-480.5	3.8	880 ± 100	0 ± 100
PRL-2001	456.0-458.0	7.0	$1370 \pm 90$	$490 \pm 130$
PRL-2000	390.3-392.8	7.5	$2430 \pm 90$	$1550 \pm 130$
PRL-1999	260.8-270.5	4.0	$3800 \pm 90$	$2920 \pm 130$
PRL-1992	112.8-156.0	7.6	$3780 \pm 90$	$2900 \pm 130$
PRL-1998	2.5 - 10.8	7.7	$4270 \pm 90$	$3390 \pm 130$

Table 2 Details of the subsamples from Dandak Stalagmite and their <sup>14</sup>C ages

Sample code	Depth from tip (mm)	Sample weight (g)	Apparent age (years BP)	Corrected age (years BP)
PRL-1995	12.5-22.3	3.2	1310 ± 80	0 ± 80
PRL-1996	72.3-79.0	4.4	$1640 \pm 80$	$410 \pm 110$
PRL-1993	127.5-135.8	4.6	$1970 \pm 80$	$740 \pm 110$
PRL-1997	176.8-185.3	4.1	$1940 \pm 80$	$710 \pm 110$
PRL-1994	267.3-278.8	5.0	$4770 \pm 100$	$3540 \pm 130$

dripping water. However, these assumptions need to be tested in future. Past rainfall is reconstructed from speleothem  $\delta^{18}O$ , following the approach given below. The ratio of the annual rainfall (1534 mm) to the average monthly rainfall (270 mm) during the monsoon season (June to October) is 5.7, for the nearest meteorological station (Jagdalpur). This ratio can be used to get the annual depletion rate. The depletion rate in the cave dripping water is considered to be 1.5‰ (shown by monthly rain in the IAEA data) for 100 mm increase in the monthly rain in the cave site. We have not used the depletion rate observed in equation (3) as it is based on only three data points and samples collected over three months of the wet season of 1999, whereas equation (1) is based on samples collected over more than 10 years (Yurtsever and Gat. 1981).

The following expression has been used to get a relation between the amount of annual rainfall (Pa, mm) and the  $\delta^{18}O$  of speleothem carbonate:

$$Pa = (100/1.5)(5.7)(\delta^{18}O_{tip} - \delta^{18}O_{i}) + 1534$$
 (5)

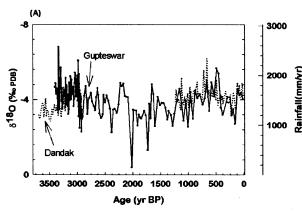
or

$$Pa = 380(\delta^{18}O_{tip} - \delta^{18}O_{i}) + 1534 \tag{6}$$

where  $\delta^{18}O_i$  and  $\delta^{18}O_{tip}$  are the oxygen isotope compositions of the speleothem carbonate with depth and at the tip, respectively.

Using the above equation we have reconstructed annual rainfall (Figure 7A) of the cave locality. Being a continuous record, we discuss here primarily the rainfall reconstruction from the Gupteswar stalactite. An average variation relative to the modern value (1534 mm/yr) of about 500 mm/yr (33%) is observed in the reconstruction during the 3400-yr period. Several periods with sharp changes in precipitation are observed. If the variation in the past temperature for the last 3400 yr is assumed to be within 1°C, then the average rainfall fluctuation will be still be up to 22%. Based on the radiocarbon ages, each layer (subsample) in the Gupteswar stalactite averages about a 14-yr period, with a sampling resolution of about 40 yr between today and about 2900 yr ago. From 3000 yr to the base of the deposit, i.e., c. 3400 yr, layers in the subsamples average about 3 yr, while the sampling resolution is about 11 yr. Subsamples in the Dandak stalagmite have better sampling resolution ( $\sim$ 17 yr) and each subsample averages  $\sim 6$  yr.

Growth rate in the Gupteswar stalactite between 2900 and 3000 yr is higher, as observed from the increased density of data points. Yet a contradiction arises as, during this period, the mean  $\delta^{18}O_c$  has shifted by +1% and  $\delta^{13}C$  simultaneously shows a 3% enrichment (Figure 7), which means a low precipitation regime and hence a slower growth of the stalactite. It is speculated that the low precipitation event recorded both by  $\delta^{18}O_c$  and  $\delta^{13}C_c$  is genuine. The increased residence time of the seepage water because of less rainfall might enhance the



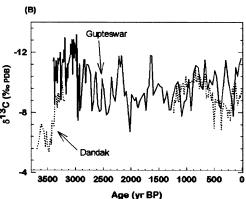


Figure 7 (A) Rainfall reconstruction from the Gupteswar stalactite (solid lines) for the last 3400-yr period.  $\delta^{18}O$  is converted to rainfall amount by using equation (6). Rainfall reconstruction from Dandak (dotted lines) is very similar to that from Gupteswar. For comparison,  $\delta^{13}$ C values are also shown in (B).

exchange of soil CO<sub>2</sub> (relative molar proportion of water being less), which will apparently reduce the dead carbon content in the seepage water before calcite is precipitated. Hence, at this point, because of low precipitation and increased residence time of water, the ages are probably underestimated. The real age of PRL-1992 should have been older.

The period between 3400 and 3000 yr is characterized with rainfall more than or comparable with present-day values. The depletion trend could be due to higher mean drip rate (Dulinski and Rozanski, 1990). Between 2900 and 1200 yr rainfall was comparatively lower with several short dry and wet phases. There are two extremely low rainfall events recorded at 2020 and 1730 yr BP. As subsamples at these points cover about a 14-yr period, these may indicate aridity for a short duration. The associated  $\delta^{13}C$  values are also higher, indicating reduction in the average drip rate. The associated errors in equation (1) and analytical errors in the measurement contribute ~15% uncertainty in the reconstructed rainfall amount.

# Comparison with palaeoclimate data

#### Indian data

Palaeomonsoon variability has been documented from marine cores earlier. A sharp change in vegetation was observed in the west coast of India at c. 3500 BP, indicating less humidity, and it continued till c. 2200 yr BP (Caratini et al., 1994). Weakening of monsoon was also observed based on fluxes of total planktonic foraminiferal shells and Globigerina bulloides

Table 3 Comparison of recently published <sup>14</sup>C and U-Th dates

Sample position	<sup>14</sup> C age Apparent	<sup>14</sup> C age Corrected	U-Th age Alpha
(*mm from tip; **cm from base)	age (yr BP)	age (cal. BP)	•
Max's cave stala			
248.5*	$6086 \pm 67$	2514	-
315	_	_	$3900 \pm 190$
Gardner's Gut st	alagmite GG1 (	Williams et al.	. 1999)
300*	$11853 \pm 61$		_
341	_	_	$10220\pm530$
Courder out of	ala amita CC2	W:11:	1000)
Gardner's Gut sta 300.5*	5301 ± 66	2798	, 1999)
366	3301 ± 66	2198	2770 + 640
703		10 523	$3770 \pm 640$
757		10 323 —	$10110 \pm 490$
131		_	10 110 ± 490
Vil-stm1b stalagr	nite (Genty et	al., 1999)	
80.4**	$1440 \pm 60$	640	_
79.7		_	$680 \pm 30$
71	$2170 \pm 110$	1275	_
70.7			$1370 \pm 40$
59.5	$2320 \pm 60$	1440	$1240 \pm 35$
42.5	$3030 \pm 70$	2200	_
24.6	$3450 \pm 60$	2675	$2590 \pm 85$
5	$3660 \pm 70$	3050	-
1.5	$3800 \pm 70$	3175	$3070 \pm 90$
Han-stmlb stalag	mite (Genty et	al 1999)	
3**	$13020\pm160$		_
12.1	-	-	$10680\pm110$
12.3	$10990\pm160$	10 700	-
32.90	-	-	$10060 \pm 95$
33	$10300\pm110$	9700	-
43.5	$10180 \pm 200$	9650	$10010 \pm 95$
54.3	$9510 \pm 130$	8875	$9070 \pm 105$
66.95	_	_	$8520 \pm 70$
67	$9430 \pm 110$	8700	-
93.7	$8090 \pm 90$	7350	_
93.80	_	_	$7140 \pm 90$
101	$7940 \pm 100$	7225	$7210 \pm 105$
108.80	_	-	$6392 \pm 172$
108.9	$7410 \pm 90$	6725	-
129.4	$6330 \pm 70$	5475	_
129.45	_	<del>-</del>	$5220 \pm 80$
136.10		_	$4780 \pm 35$
137.7	$5460 \pm 60$	4300	

(Naidu, 1996) from the western Arabian sea at c. 5000 BP. which continued till c. 1000 BP. An arid phase was observed in southern India between 6000 and 3500 BP, with a short humid phase around 600 yr ago (Sukumar et al., 1993). More recent work on peat samples (Geeta Rajagopalan et al., 1997) shows an arid phase during 2000-5000 BP. Compared to the above available climate data, time resolution in the present study is higher and hence only coarse trends in the published climate reconstruction changes can be compared.

As our record up to c. 1200 BP shows a good concordance between the isotopic compositions of the two caves, radiocarbon dating and rainfall reconstruction appear to be reasonably valid. Within this period the humid phase starts at c. 1000 BP and attains a maximum between 500 and 750 BP. There is a trend towards aridity reaching the peak value around 125 BP, which, considering the dating uncertainty ( $\pm 130$  yr), coincides with the drought events recorded at the beginning of AD 1900 (Singh et al., 1991).

There is a striking difference in the isotopic composition of the two caves in the initial part of the speleothem growth. The

Table 4 Details of samples of rain spells collected at Jharsuguda

Date of collection	Sample no.	Rain (mm)	δ <sup>18</sup> Ο <sub>VSMOV</sub> (‰)
10-7-99	R1	5.73	-3.13
11-7-99	R2	1.91	-3.85
12-7-99	R3	0.32	-0.64
14-7-99	R4	22.92	-3.34
15-7-99	R5	9.23	-2.84
29-7-99	R6	6.37	-2.47
3-8-99	R7	35.01	-8.25
4-8-99	R8	81.17	-9.91
21-8-99	R9	11.14	-3.83
24-8-99	R10	24.83	-5.23
29-8-99	R11	4.77	-2.96
1-9-99	R12/13	71.94	-10.62
2-9-99	R14	37.24	-8.19
3-9-99	R15	4.14	-4.51
5-9-99	R16	8.28	-2.90
7-9-99	R17	10.82	-4.09
9-9-99	R18	2.71	-5.64
13-9-99	R19/20	49.34	-10.64
15-9-99	R21/22	37.24	-7.76
16-9-99	R23	8.28	-5.09
17-9-99	R24	6.37	-5.19
18-9-99	R25	2.01	-4.99
23-9-99	R26/27	98.36	-10.12
28-9-99	R28/29	59.39	-9.70
Monthly cumulative values	R1-R6 (July)	46.47	-3.10
	R7-R13 (Aug)	228.86	-8.93
	R14-R29 (Sept)	324.16	-8.88

Dandak sample shows an arid phase between c. 3700 and 3200 BP, which agrees with the other palaeoclimatic results, whereas the Gupteswar sample shows higher rainfall between c. 3400 and 3000 BP in contrast to other observations. It appears that the assumption about addition of dead carbon being constant is invalid in this part of the speleothem. The concordance of

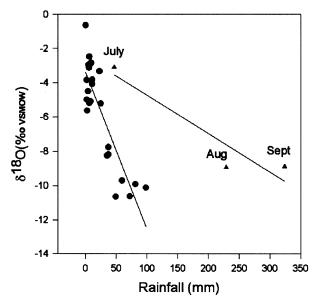


Figure 8  $\delta^{18}$ O versus rainfall plotted for the samples (solid circles) collected from Jharsuguda. The rainfall for each month is added to get the monthly rainfall for July, August and September (solid triangles). A best-fit line shows depletion rate agreeing with global values (see text).

variations in the stable isotopic compositions from both caves breaks down beyond 3200 BP.

#### Varved sediment data from off Karachi

A high-resolution (comparable with the speleothems; i.e.,  $\sim 1$ yr to ~15 yr) climate record from the Indian land mass spanning the last 3400 years has not been available so far. Recently, a high-resolution (~7 yr) record has been reconstructed by von Rad et al. (1999) using thickness variation in the varved sediments collected from the northeastern Arabian Sea, off Pakistan. Monsoon reconstruction from varved sediments and Gupteswar stalactite has been compared below. According to von Rad et al. (1999), precipitation and, hence, river runoff are assumed to control varve thickness. The deposition at the sampling site (von Rad et al., 1999) occurs both during the summer monsoon (June-September: from southwest monsoonal currents) and the winter monsoon (November-March: linked to storms in the Mediterranean sea). These derive alternatively dark and light-coloured sediment sequences which form an annual couplet. The precipitation may have fluctuated due to variations in the extreme positions of the ITCZ (intertropical convergence zone) and, hence, the variability in varve thickness was interpreted as a proxy for past rainfall variations (Lückge et al., 2001). Stable isotope analysis of Gupteswar stalactite has a different time resolution in different periods (Table 5). The varve thickness data shown by von Rad et al. (1999) is a 7-point moving average of annual thickness data and hence our data values were smoothed out by using appropriate running mean during different time periods (Table 5). Figure 9 shows comparison of varve thickness data and rainfall reconstructed from the Gupteswar stalactite. The observations are as follows.

(1) From 0 to 1000 yr BP, the initial trends are quite similar. Later, during 500 to 1000 yr BP, there is an opposite trend in them. The stalactite shows high rainfall around 600 yr BP, whereas thickness data shows low rainfall. Pollen records from different lakes in Rajasthan (Singh et al., 1974; Bryson and Swain, 1981; Swain et al., 1983) show a good monsoon rainfall similar to our observations. Nitrogen content which is an indicator of marine productivity is strongly related to the upwelling of the surface water. If it can be used as a proxy of past rainfall (as higher-intensity monsoonal winds will drive intense coastal upwelling and also increase monsoon rainfall on land), the recently reconstructed data from the Indian coastal area in the Arabian Sea for the last 1 kyr from a ocean sediment core (Agnihotri et al., 2002), shows a reduced level of productivity and hence a low level of rainfall around 600 yr BP. These observations suggest that the Arabian branch of the SW monsoon (source of rain for coastal India and Pakistan) was subdued whereas the Bay branch was active during 600 yr BP. A possible mechanism can be envisaged whereby the extreme ITCZ positions either moves towards the northeast or the northwest. This may cause weakening of the Arabian Sea branch and strengthening of the Bay of Bengal branch and vice

- (2) From 1000 to 2000 yr BP, speleothem records aridity, with two highly arid durations around 1700 and 2000 yr BP. The varve data show an increasing trend of rainfall starting at 1000 yr BP and then after attaining high values again decrease with sharp low-rainfall events around 1900 and 2000 yr BP. The low-thickness event during 2000 yr BP is also shown by our rainfall reconstruction. Lückge et al. (2001) have recently reported lowest Ti/Al ratio (terrigenous) in the same core during this period.
- (3) From 2000 to 3400 yr BP, the trends of increasing rainfall are very similar in both the records.

# Carbon isotopes

The carbon in the speleothem CaCO<sub>3</sub> is incorporated from two sources: (1) soil CO<sub>2</sub> which is isotopically lighter than the atmospheric CO<sub>2</sub> and (2) bedrock carbon. The carbon isotopic evolution of speleothems is a complex process that depends upon several factors (Hendy, 1971; Wigley et al., 1978; Lauritzen and Lundberg, 1999), mainly the following.

- (1) Photosynthetic pathways: climatic change may alter the abundance of C3 and C4 type of plants on the top of the cave and hence the relative contribution to the speleothem  $\delta^{13}$ C (Clark and Fritz, 1997; Lauritzen, 1995; Gascoyne, 1992).
- (2) Biological activity: carbon dioxide in the soil is derived from two sources: microbial decomposition of the soil organic matter and root respiration of the plants (this has relatively more negative  $\delta^{13}$ C). During wet periods intense respiration process might result in high pCO<sub>2</sub> and depleted δ<sup>13</sup>C (Hesterberg and Siegenthaler, 1991).
- (3) Bedrock proportion: limestone dissolution may proceed (Hendy, 1971; Dulinski and Rozanski, 1990; Clark and Fritz, 1997) along two extreme routes, i.e., open system and closed system. Temporal variation in the dissolution mechanism will modify speleothem  $\delta^{13}$ C.
- (4) Drip rate in the cave: there is progressive enrichment in the successive (time variation) CaCO<sub>3</sub> deposits (Hendy, 1971; Wigley et al., 1978; Dulinski and Rozanski, 1990; Hellstrom et al., 1998) even when oxygen isotope equilibrium is observed. Due to this effect, higher drip rate results in depleted speleothem  $\delta^{13}$ C. As the drip rate is directly proportional to the rainfall in a crude way, this may result in dependence of speleothem  $\delta^{13}$ C on the amount of rainfall.

The interpretation of  $\delta^{13}C_c$  depends upon the settings of the cave environment where one of these processes may be dominant and others may be assumed invariant (Lauritzen and Lundberg, 1999).

# $\delta^{13}$ C values of Gupteswar and Dandak

 $\delta^{13}C$  of the representative leaves collected from the Gupteswar and Dandak caves have  $\delta^{13}C$  values close to -28.0%. Considering the average  $\delta^{13}$ C of the soil CO<sub>2</sub> to be -28.0%, enrichment due to diffusion (by -4.4%; Cerling et al., 1991) will result in soil CO<sub>2</sub> environment labelled as  $\delta^{13}$ C = -23.6%.

At 25.5°C temperature, calcite precipitated in isotopic equilibrium with the CO<sub>2</sub>(aq), HCO<sub>3</sub>, CO<sub>3</sub><sup>2</sup> in the seepage water is enriched by 10.4% (Clark and Fritz, 1997). Hence, at the tip,  $\delta^{13}$ C should be -13.2%. However, this is the value for the first calcite precipitated from the drip water and successive calcite is further enriched as shown by the values of the tip layers,

Table 5 Time resolution in the rainfall record from Gupteswar stalactite and varve thickness record from von Rad et al. (1999). Appropriate running means have been calculated to make the time resolutions to same level

Period	Rainfall record from Gupteswar		Varve thickness record from von Rad et al. (1999)	
	Time resolution (yr)	Running mean applied	Time resolution (yr)	Running mean applied
0-2920 2920-2980 2980-3400	13.36 0.45 3	none 16 pt 2 pt	7 7 7	2 pt none none

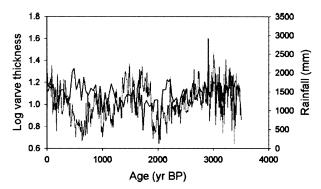


Figure 9 Comparison of rainfall record from the Gupteswar stalactite (black line) and the varve sediments (grey line) from the east Arabian Sea (von Rad et al., 1999).

 $\delta^{13}C = -11.4\%$  and -9.4% for the Gupteswar and Dandak caves, respectively. Soil thickness above both caves varies between 10 and 30 cm.  $\delta^{13}$ C values at the tip are different for Gupteswar and Dandak. This can be explained based on the average drip rate in the cave, which depends on the roof size. Roof thickness of the Gupteswar cave (~20 m) is comparatively less than that of the Dandak cave (~40 m). Due to this the average drip rate should be more in the Gupteswar cave compared with the Dandak cave and, hence, the Gupteswar values are probably relatively depleted (-11.4%).

The  $\delta^{13}$ C matches well only between 600 and  $\sim$  1200 yr BP and then the similarity breaks down between 600 and 0 yr BP (Figure 7). Between 600 and 0 yr BP  $\delta^{13}$ C values of the Gupteswar sample are generally higher than those of the Dandak sample. A possible reason for this may be the internal changes in the flow pathways in either of the caves due to which the mean drip rate might have changed. Between 1250 and 600 yr BP the mean drip rates of the two caves were probably almost the same, hence there is a good similarity in the  $\delta^{13}$ C profile.

# $\delta^{13}$ C versus $\delta^{18}$ O in speleothems

 $\delta^{13}C$  versus  $\delta^{18}O$  for the two speleothems are plotted in Figure 10. For the regression equation  $\delta^{13}C = m\delta^{18}O + c$ , m and c values are listed in Table 6. Significant correlations of  $\delta^{13}C$  with  $\delta^{18}O$  (r = 0.59 to 0.72) show that  $\delta^{13}C$  is also dominantly controlled by rainfall variations. It is possible

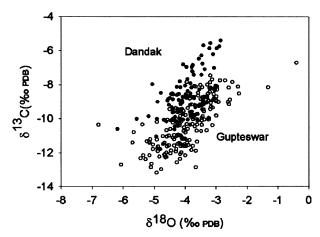


Figure 10  $\delta^{13}$ C versus  $\delta^{18}$ O measured along the growth direction for the Gupteswar stalactite (open circle) and the Dandak stalagmite (black circle).

**Table 6** Regression equations relating  $\delta^{13}$ C and  $\delta^{18}$ O

Cave	Equation	Correlation coefficient and no. of data points
Gupteswar	$\delta^{13}C = (1.28 \pm 0.09)$	0.72
_	$\times \delta^{18}O - (5.21 \pm 0.35)$	, 198
Dandak	$\delta^{13}C = (1.31 \pm 0.17)$	0.58
	$\times \delta^{18}$ O – (3.45 ± 0.72)	, 103

that varying drip rate is dominantly controlling the  $\delta^{13}$ C changes. However, changes in the respiration generated partial pressure of CO<sub>2</sub> which is proportional to rainfall may also be involved in the  $\delta^{13}$ C-rainfall dependence. There are other processes which are not rainfall dependent: such as proportion of bedrock carbon contributing dead carbon to the seepage water, random fluctuations in the seepage pathways, changes in the biological activity, etc. These may contribute to the natural variability and may be responsible for the scatter in the data.

### Conclusions

Speleothem  $\delta^{18}$ O is found to be primarily controlled by the δ<sup>18</sup>O of the local precipitation. The observed depletion rate in precipitation ( $-2.2 \pm 0.8\%/100$  mm), agrees very well with the globally observed depletion rate ( $-1.5 \pm 0.2\%/100$  mm). A significant correlation between  $\delta^{18}O$  and  $\delta^{13}C$  (along growth direction) indicates that rainfall controls the  $\delta^{13}$ C value of the speleothem. Assuming a constant dead carbon contribution from parent bedrock for all past, ages have been assigned to each speleothem. The longest duration, 0-3400 yr BP, is spanned by the Gupteswar stalactite. Growth of the Dandak stalagmite started at ~3700 yr BP and continued up to 3200 yr BP. After a hiatus of ~2000 yr the growth started from  $\sim 1200$  yr BP and continued till the present. δ<sup>18</sup>O variations from 0-1200 yr BP duration are concordant for the two speleothems, which shows that the radiocarbon chronology is reasonably correct, and also common environmental changes in  $\delta^{18}\text{O}$  are recorded by the speleothems. In the initial duration of these speleothems (3700 to 3200 yr BP) the assumption of constant dead carbon contribution from parent bedrock is probably not valid. Rainfall reconstruction from speleothems agrees well with the earlier observations using other proxies. Before  $\sim 1200$  yr BP, an arid phase is indicated by the Gupteswar stalactite, which is observed by the sediment cores in the western Arabian sea. Sharp (~14 yr timespan) arid events observed around 2000 yr BP may have been due to large-scale weakening of the southwest monsoon system as similar events are observed in the varved sediments collected from the northeastern Arabian Sea. At  $\sim 600$  yr BP, higher rainfall compared to the present day is shown by both the speleothems. This study also demonstrates the potential of the technique, that speleothems in the Indian tropical area can be used to reconstruct past rainfall variations. However, the chronology needs to be strengthened by the use of the U-Th method of dating.

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