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Research Article

SPELEOTHEMS POTENTIAL OF CENTRAL HIMALAYA WITH U-TH DATING UNCERTAINTIES

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ABSTRACT
There are wide evidences that the speleothems especially stalagmite has a great potential to provide the palaeomonsoon variability with decadal to seasonal scale using stable isotopic records. The North-Western Himalaya receives the rain from two different moisture regime e.g., Indian summer monsoon (ISM)/south west monsoon (SW) as well as westerlies. Thus, the present paper focused on the potential and problem related to the speleothems of Central Himalaya, Uttarakhand with possible cause of U-series dating uncertainties. The present study also aims to provide the basic information of 16 caves from different parts of Central Himalaya (Uttarakhand) with a prime objective to identify the reliability of radiocarbon dating of stalagmites of Central Himalaya as it is for the first time being introduced in this part of speleothems research.

Keywords: *Central Himalaya, Speleothems, $\delta^{18}O$, ^{14}C AMS Radiocarbon Dating, U-Th dating, Indian Summer Monsoon*

INTRODUCTION
It is well known fact that the speleothems (especially the stalagmite) have strong potential for preserving a high resolution palaeoclimatic records. They can provide the clues of past precipitation, temperature, humidity and vegetation changes.
Thus, the combined data of stable isotopes and precise dating can provide a continuous high and temporal resolution record of precipitation and temperature changes. The speleothems are calcareous and mostly composed of calcium carbonate in the form of calcite/aragonite. Previous studies have shown that oxygen isotope ratio can be used to determine past temperature changes as the most abundant isotope is ^{16}O with a smaller percentage of ^{18}O, thus, the ^{16}O diffuses more rapidly as being lighter.
Therefore, the heavier $\delta^{18}O$ represents the weakening of the monsoon strength. The major advantage of speleothems research in Central Part of the Himalaya is the different sources of moisture regimes e.g., ISM and mid-latitudinal westerlies.
Therefore, the speleothems can grow even throughout the drought monsoon condition in peninsular India. The previous stalagmites $\delta^{18}O$ based studies pointed out the role of ISM variability as well as behaviour of westerlies over the north-western Himalaya based on $^{230}Th/U$ dating. The stalagmite $\delta^{18}O$ based palaeoclimatic records are available from Lesser Himalaya, e.g., Bølling-Ållerød interstadial between 15.2 to 11.7 ka from Timta cave (Sinha *et al.*, 2005), last ~ 400 years from Chulerasim Cave (Kotlia *et al.*, 2012) and two millennia records ~ 1800 years from Dharamjali cave (Sanwal *et al.*, 2013) and Sahiya cave (Sinha *et al.*, 2015); and more recently mid-Holocene records ~ 4 ka BP from Sainji cave (Kotlia *et al.*, 2015) and 750 years records from Panigarh cave (Liang *et al.*, 2015).
The success rate of speleothems research depends on the shape, size, open and closed system of the cavewith precise U-Th ages due to low analytical error (Wang *et al.*, 2001). The^{230}Th ages determined using inductively coupled plasma mass spectroscopy ICPMS) technique. The three isotopes of uranium (^{238}U, ^{235}U ^{234}U) is bound in silicate and oxide minerals in rocks and is co-precipitated with the calcium-carbonate forming the speleothems. It decays into daughter isotopes (^{230}Th, ^{232}Th) and as thorium is insoluble in water is therefore, absent at the time of deposition however contamination of the daughter isotope ^{230}Th may occur as a result of co-precipitation of ^{232}Th, attached to clay particle present in the cave drip water (Frank *et al.*, 2006). This is one of the main problem in the speleothems studies despite the significant growth patterns.

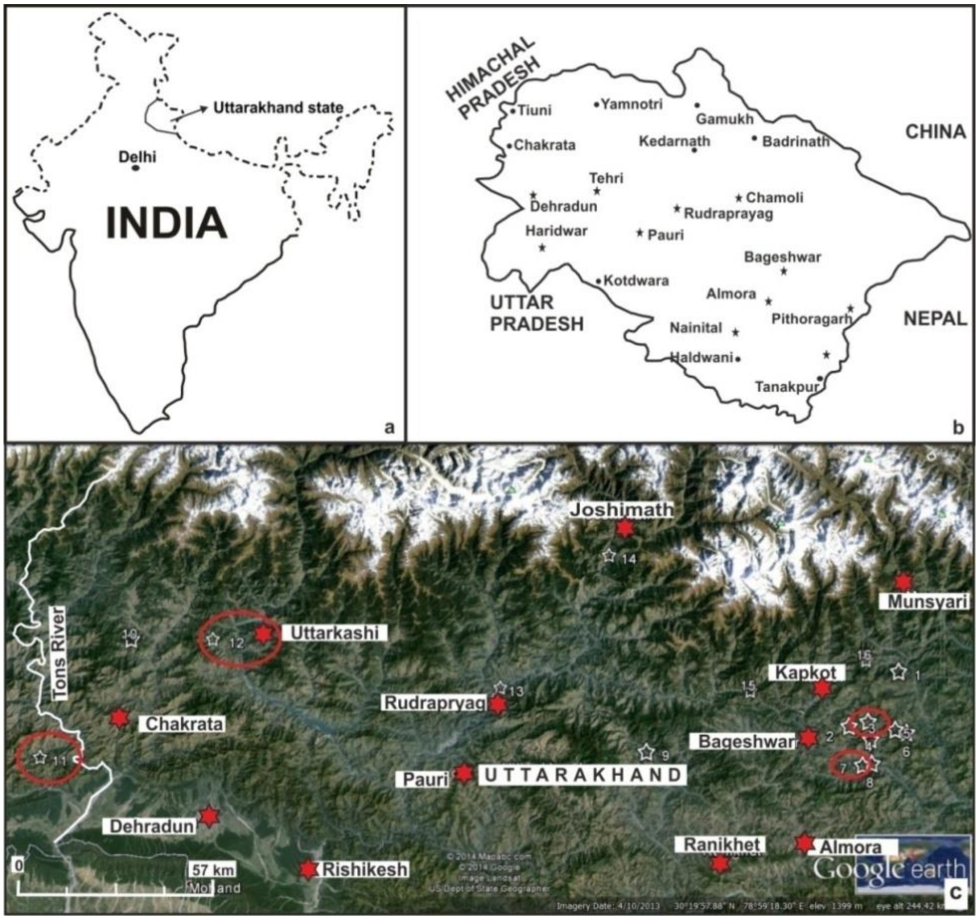

Figure 1: Map of the Uttarakhand State with Locations of Different Caves in Google Map, Denoted by Stars. Encircle Part Shows the Collect Stalagmites for Present Study. The Number for the Different Caves e.g., (1) Jhinya Cave; (2) Kanda Cave; (3) Bhadrakali Cave; (4) Joshi Cave; (5) Suklari Cave; (6) Koteshwar Cave-1; (7) Seraghat Cave; (8) Nayolisera Cave; (9) Mehalchauri Cave; (10) Koti Cave; (11) Tityana Cave; (12) Bhramkhal Cave; (12) Koteshwar Cave-2; (14) Tapod Cave; (15) Mophata Cave; (16) Viyas Cave. For details see Table 1 [Courtesy: Google Earth]

STUDY AREA, GEOLOGY AND CAVES INFORMATION

The present investigation focused on the 16 caves of Central Himalaya (Figure 1) with aim to provide the brief about, the cave locations, cave length, host rock condition, U-Th dating uncertainties and speleothems potential with^{14}C AMS chronology. We had identified mostly the cave formed in Deoban limestone of Tejam Group. Among these caves, only four caves seem suitable due to availability of ideal stalagmites.

The length of most of caves is less than or nearly 10 m and have wide opening with unavailability of the samples (Table 1). Although, the six stalagmites are collected from the eastern, middle and western sectors of the central Himalaya Himalaya e.g., Tityana cave (Near Dehradun, along Tons valley), Joshi cave (Berinag, Pithoragarh), Nayolisea cave (Seraghat, Pithoragarh) and Bhramkhal cave (Uttarkashi) (Figure 1).

The Himalaya is criss-crossed by various thrusts/faults and the caves structure may be controlled by the tectonic activity (Figure 2a). Further, only two stalagmites one from Joshi cave and another from Tityana cave found suitable for dating. The Tityana cave located along the Tones valley and found in Ambota Formation of Deoban Group (Figure 2b). The Joshi cave is located in Deoban limestone (Figure 2c) and it observed the limestone replaced to dolomite and magnesite. The Deoban structural belt appears in the

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Tons valley and then continue SE into the Garhwal Himalaya up to the Yamuna valley. Beyond this, it continues and appears in broken due to the cover of younger sediments from the upper reaches of the Yamuna and Uttarkashi area an altogether different but Homotaxial structural belt of the Tejam-Pithoragarh appears (Valdiya 1980).

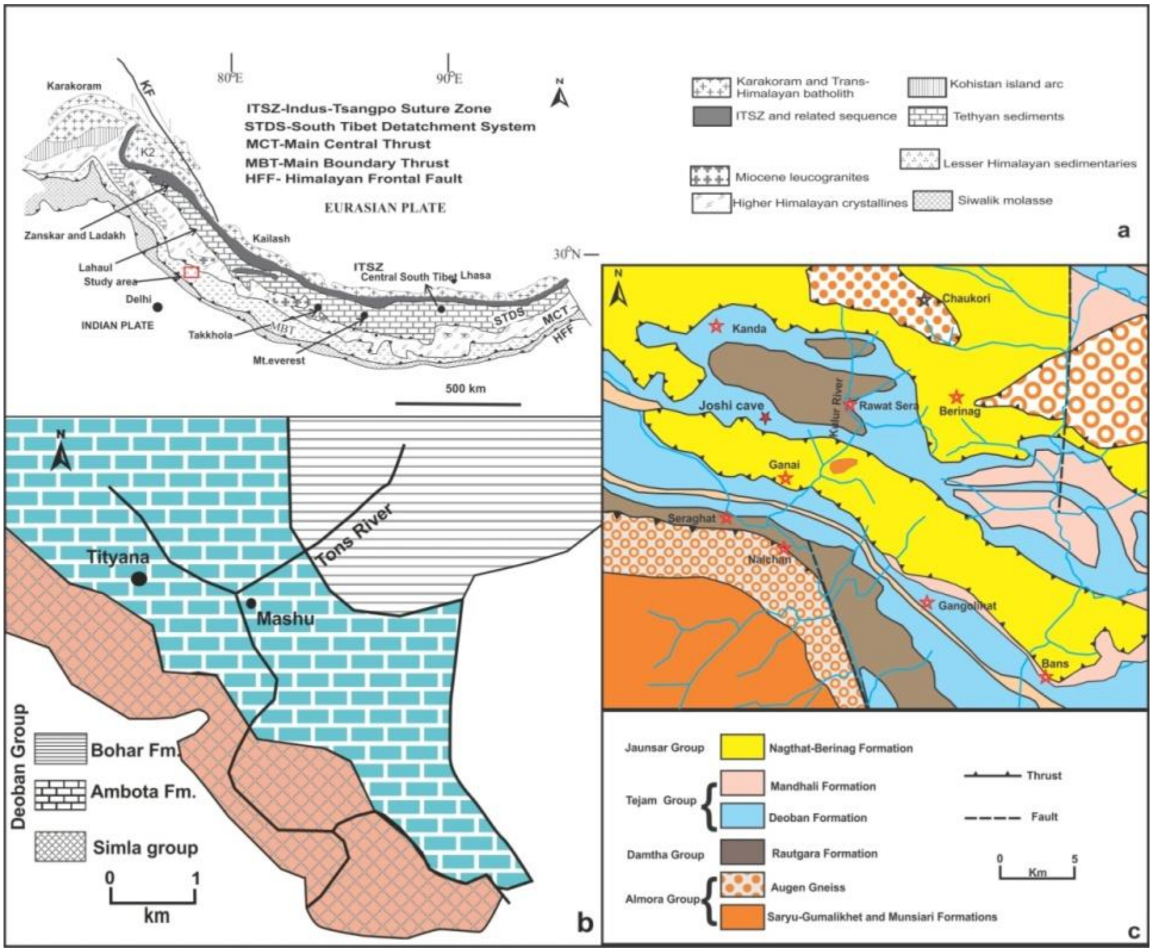

Figure 2: (a) The Himalaya is Criss-Crossed by various Intracrustal Boundary Thrusts/Faults (after Godin 2003); (b) The Tityana Cave Located in Ambota Formation of Deoban Group (After Thakur 1992); (c) The Joshi Cave is Located in Deoban Limestone (after Valdiya 1980) (Figure 2c) and it Observed the Limestone Replaced to Dolomite and Magnesite within the Region

MATERIALS AND METHODS

The collected stalagmites samples were cut into two halves along central growth axis and gently polished using emery paper. Only two samples found ideal for U-Th dating, other seems unsuitable due to contaminated growth rings.

Hendy test used in present study taken from the one layer of Tityana cave sample (Joshi *et al.*, Pers. commun., 2016). The ^{230}Th/U dating were done in the Mass Spectrometry and Environment Change Laboratory (HISPEC), Department of Geosciences, National Taiwan University using Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS), following the method developed by Shen *et al.*, (2003, 2012).

The samples could not be precisely dated by the Th/U method due to low Uranium concentration (Table 2). Simultaneously, the ^{14}C AMS chronology (see table 2 for depth) was done at Poznań Radiocarbon Laboratory, Poland using the technique of Accelerator Mass Spectrometry (AMS).

Table 1: Location of Identified Caves in Uttarakhand, Himalaya. Star Denotes (*) the Caves used for U-Th and AMS Dating

S. No.	Name of the Cave	Location	Length	Conditions
1	Jhinya	N 29^0 53' 30.7"; E80^0 07' 02"	5m	Wide entrance lot of immature samples
2.	Kanda cave	N:29º 49´13.03"; E: 79º 54’ 30.59”	3.5m	Contaminated samples
3.	Bhadrakali cave	N 29^0 48' 33.9"; E79^0 58' 01.1"	5m	River dominated cave
4	Joshi cave*	N 29^0 45' 50.1"; E 79^0 57' 01.	55 m	Two sample collected
5.	Suklari cave	N 29^0 45' 32"; E80^0 01' 52.6"	10m	No growth of speleothems
6.	Koteshwar cave- 1	N 29^0 44' 20.3"; E80^0 03' 29.8"	10 m	Some flow stone and columns
7.	Seraghat cave	N 29^0 42' 49"; E 79^0 53' 41.8"	3 m	No growth of speleothems
8.	Nayolisera cave	N 29^0 42' 22.9"; E 79^0 55' 20.6"	3.5 m	Few speleothems/two collected
9.	Mehalchauri cave	N 30^0 00' 50.3"; E79^0 19' 11.8"	14 m	No growth of speleothems
10.	Koti cave	N 30^0 49' 18.34"; E78^0 03' 04.91"	10m	Sample not found
11.	Titiana cave*	N: 30^{0}38’30.7’’; E77^{0}39’07.4’’	13 m	Few stalagmite/two collected
12.	Bhramkhal cave	N 30^0 43' 55.4"; E78^0 16' 55.2"	7 m	Flow stone/few stalagmite/one collected
13.	Koteshwar cave-2	N 30^0 18' 06.3"; E79^0 00' 27.1"	5 m	River dominated cave
14	Tapod cave	N 30^0 29' 30.5"; E79^0 28' 22.2"	5m	Looks river cave/only one thick stalagmite
15	Mophata cave	N 30^0 00' 58.0"; E79^0 40' 57.4"	10 m	No growth of speleothems /debris
16.	Viyas cave	N 29^0 57' 24.4"; E80^0 02' 21.1"	50m	No growth of speleothems

Table 2: U-Th and ^{14}C AMS Chronology of Tityana and Joshi Caves, *Tityana Cave Dates used from Joshi *et al.*, (pers. commun., 2016)

Depth (mm)	^{238}U ppb^a	^{232}Th ppt	d^{234}U Measured	Corrected (U-Th) Ages	Uncorrected ^{14}C AMS Dates
Joshi Cave (JC)					
5	33.901 ± 0.048	3247 ± 10	205.2 ± 3.6	4,664 ± 1098	6110 ± 40 BP
230	64.962 ± 0.062	63560 ± 640	199.3 ± 1.8	5,042± 12092	6890 ± 40 BP
Tityana Cave (JC)*					
1	97.00± 10	94916± 795	39.5 ± 2.0	-846± 16432	2985 ± 30 BP
128	171.00± 0.24	46516± 272	22.2± 1.8	9,149± 3792	4940 ± 35 BP

RESULTS AND DISCUSSION

U-Th Dating and its Uncertainties

The precision of the ^{230}Th/U dating strongly depends on the concentrations of uranium and detrital thorium (Goslar *et al.*, 2000). In presented stalagmites the ^{230}Th/U method have proved unsuccessful due to multiple source of non- authigenic ^{232}Th and low uranium concentration (< 10 ppb). In the speleothems the activity of detrital ^{230}Th do not produced from the decay of ^{234}U. The authigenic input due to recrystallization of rock or may be allogenic transported from the nearby area through water and wind activity. Thus, the uncertainty of the ^{230}Th/^{232}Th activity ratio affects the accuracy of the ^{230}Th/U age, especially when the activity of ^{232}Th is high for the sample (see Table 2). Thus, the Radiocarbon chronology was introduced for the collected sample.

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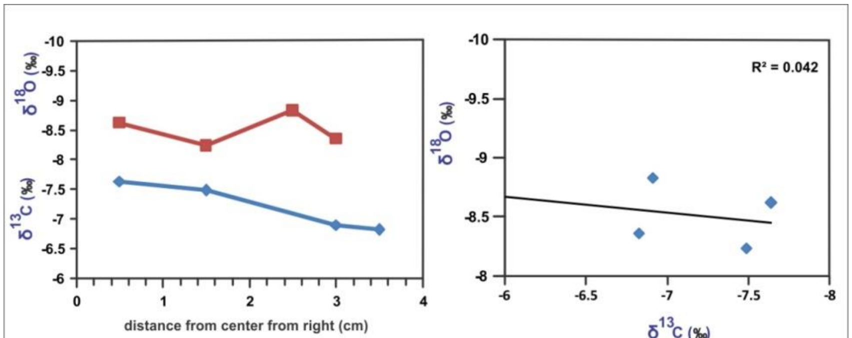
^{14}C AMS Dating

Low Uranium with high ^{232}Th contents in the stalagmites is unsuitable to apply ^{230}Th/U dating. Thus, the ^{14}C AMS dating can solved the propose and used as alternative choice (Zhao *et al.*, 2015). Although, for speleothem dating, it is necessary to know the initial activity of ^{14}C (or reservoir effect; Goslar *et al.*, 2000) of the carbonate material. In general, the radiocarbon dating could not be employed due to dead carbon fraction during the carbonate precipitation. The dissolved CO_2 in the seepage water exchange well with atmospheric CO_2 in an open system of carbonate reservoir, the dead carbon influence becomes minimal (Hendy 1971). Although, the AMS method has successfully been applied to help to build chronologies for young speleothems (Yadava and Ramesh, 2005; Laskar *et al.*, 2013). The reliability of ^{14}C age need to be know as it introduced first time in Central Himalaya for speleothems research. The ^{14}C AMS dates for Joshi cave at bottom and top (230mm and 5 mm) is slightly higher than ^{230}Th/U dates, seems partly affected by dead carbon. Although, the ^{14}C AMS dates are uncalibrated due to unknown source of dead carbon. Moreover, the ^{14}C AMS dates of Tityana cave at bottom (128mm) is younger than the ^{230}Th/U dates (see Table 2). Thus, the younger ^{14}C AMS dates than ^{230}Th/U dates, reflects negligible dead carbon influence in the ^{14}C AMS dates and strong influence of initial ^{230}Th in ^{230}Th/U ages (Zhao *et al.*, 2015).

Hendy Test

Hendy (1971) suggested that ^{18}O values remain constant along a single growth layers and there is no relationship between ^{13}C and ^{18}O. Although, recent study shows that the Hendy test is not the only criterion to judge the speleothems (Fleitmann *et al.*, 2004; Dorale and Liu, 2009; Kotlia *et al.*, 2012). Hendy (1971) further suggested that the degassing of CO_2 was sufficiently slow to maintain the isotopic equilibrium during carbonate precipitation. In our case, a small fluctuation of δ^{18}O and δ^{13}C along layer could be due to a little shift in layer as the low value (0.04) of correlation coefficient (R^2) as well as P-value (<1) between δ^{18}O and δ^{13}C for both layers suggest that the kinetic fractionation was negligible and thus the δ^{18}O can reflect the climate signals (Figure 3).



	Coefficients	Standard Error	t Stat	P-value	Lower 95%	Upper 95%	Significance F
Intercept	-9.483453276	3.257907602	-2.9109	0.100534	-23.501098	4.53419176	
X Variable 1	-0.134444976	0.451036419	-0.29808	0.793757	-2.0750981	1.80620811	0.793757028

Figure 3: Shows the Variations between δ^{18}O and δ^{13}C with Distance from the Central to Right of the Sample to Determine the Isotopic Equilibrium during Carbonate Precipitation. The Low Correlation Coefficient and Significance of P Values Less than One with 95% Confidence Level Suggests the Slow Degassing of CO_2 for Tityana Sample

Caves Entrance, Contaminated Growth Rings and Possible Causes

The entrance of Jhiniya and Viyas caves is wider and the Kanda cave (4m x 3.5m), Tityana cave (3mx1m), Bhramkhal cave (1m x1.5m), Koteshwar cave (2.5m x 3m), Nayolisera cave (2mx1.5m), Mophata cave and Tapod Cave (2x3m) have moderately wider entrance. On other hand, the Bhadrakali cave (2' X 1'), Koteshwar-1, Joshi cave (1.5'x2') have smaller entrance. The stalagmite collected from the Joshi cave which has highest length and smallest entrance among all the caves shows that the contaminated growth ring perhaps the silica precipitation through the cracks as the contamination through wind borne dust is not possible (Figure 4a). The cave is located along the Berinag thrust and it observed that the limestone is replaced to dolomite. The another 23 cm high stalagmite collected from same cave(Figure 4b), further used for ^{230}Th/U and ^{14}C AMS dating and seems the higher growth of the sample as ^{14}C AMS age is 780 yrs BP (See Table 2). Further, the two stalagmite collected from the Nayolisera cave found unsuitable (Figure 4c-d) due to the fast degassing of CO_2 as open cave system with wider entrance and smaller length (2mx1.5m and 3.5 m length). The rock is highly shattered within the cave region due to proximity of Raintoli fault/Saryu River Fault (Joshi *et al.*, 2016). Consequently, the rings are shifted and possibly useful to determine the past seismic activity (Figure4c-d). The recent study by Rajendran *et al.,* (2015) also suggested that the stalagmite growth perturbations become potential earthquake indicators. The Tityana cave sample seems well define growth rings (Figure 4e). The other speleothems collected from the Bharamkhal cave made up with the calcite and appeared the rapidly deposition as ring are not developed or dissolved due to fast degassing of atmospheric CO_2 (Figure 4f). Moreover, the slightly thicker top of some samples distinguished to be formed due to detrital input and also suggested that the evaporation and fast carbon dioxide escape during carbonate precipitation.

Figure 4: Cavestalagmites from Four Different Caves (a and b) Stalagmites Collected from Joshi shows the Contaminated as well as Prominent Growth Rings, mostly Formed by Calcite; (C and d) The Stalagmites of Nayolisera Cave Shows the Shifted Growth Pattern Possible due to Seismic Activity, the Arrow Indicates the Pattern of Shifting of Growth Rings; (e) Tityana Cave Sample Shows the Prominent Growth Ring; (f) Stalagmite of Bhramkhal Cave Demonstrate the Rapid Growth of the Sample

Isotopic Measurement

The oxygen isotope ratios of stalagmite calcite primarily show variations in the amount of rainfall with more negative $\delta^{18}O$ indicating higher monsoon rainfall. While, $\delta^{18}C$ can provide the information of vegetation above the cave in form of C_3 and C_4 plants. In preliminary study of Tityana cave, Joshi cave, and Nayolisea cave made up with the calcite and primarily shows the light and darker layers. Moreover, the Bhramkhal cave does not reflects the growth patterns. Although, the other speleothems from Bhramkhal cave studied by Tewari (2008) suggested that the variation in $\delta^{18}O$ is -5.37 ‰ and -7.65 ‰. The $\delta^{18}O$ fluctuation for Tityana cave is between -7.5 ‰ to -10‰. It seems that the more negative $\delta^{18}O$ of

Himalayan caves are related to amount effect, means more negative values associated with higher rainfall due to two different moisture regimes.

Conclusion

It is well established that the speleothems have potential to provide the paleoclimate information on multi decadal to decadal scales. Moreover, the fast growing stalagmites could be useful to understand the high resolution and seasonal climatic changes. There is a need to study the soil and host rock conditions and human influence to know the cave environment and potential. Thus, we can conclude that the mostly the caves in Central Himalayan are much smaller in length due to immature terrain. The detrital input in the caves as a result of deformation of rock due to proximity of thrust and faults. The contamination of growth rings suggesting that the precipitation of soil/silica during the higher rainfall events. Most of the cave stalagmite suggests the wetter or semiarid environment as suggested by contaminated or fast growth rings. The higher growth rate may be due to good contribution of westerlies over the part of Himalaya. Despite the ^{230}Th/U dating uncertainties, the caves are reliable for ^{14}C dating due to younger ages. Although, the correction of dead carbon fraction is needed. Moreover, the speleothems have a potential to provide the palaeoseismic activity other than the climate study as most of the caves studied so far are located within the zone of active thrusts/faults. The human presence should be taken into consideration for recent speleothems as it can increase the cave temperature. We also suggest that the caves should be mapped and developed as geo-tourism sites as most of the caves are unknown.

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