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Evidence of Younger Dryas and Neoglacial cooling in a Late Quaternary palaeotemperature record from a speleothem in eastern Victoria, Australia

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ABSTRACT: A calcitic stalagmite collected from a limestone cave in the Buchan area of eastern Victoria has been dated by three mass-spectrometric uranium series analyses. Two growth phases are represented: the older from 13.4 to 10.6 ka and the younger from 3.2 to 2.1 ka. Oxygen isotope analysis reveals that temperatures were below present-day values at all times, but particularly cool conditions are indicated between 12.3 and 11.4 ka, and Neoglacial conditions occurred at about 3 ka. The older cold climate event is clearly synchronous with the Younger Dryas in Europe and this is the first time that strong evidence for this event has been found in Australia. Carbon isotope variations are interpreted as indicating changes in plant productivity on the surface and are most likely controlled by variations in summer rainfall. They indicate particularly high levels of plant productivity from 11.5 to 11.0 ka.

KEYWORDS: speleothem; stable isotope analysis; uranium series dating; Younger Dryas; Australia.

Introduction

The aim of the study is to examine Late Quaternary variations in oxygen and carbon isotope ratios ($^{18}O/^{16}O$ and $^{13}C/^{12}C$) in a Victorian speleothem and to interpret them in terms of environmental change.

The calcite speleothem examined in this paper was collected from Royal Cave, a high-level cave that forms part of the Dukes Cave System, containing over 5 km of passage and developed in the folded Devonian Buchan Caves Limestone in the Buchan River Valley of Eastern Victoria (148°10′E, 37°30′S) at an altitude of between 60 and 100 m above sea-level (Fig.1). The speleothem was apparently broken during the development of Royal Cave as a tourist cave and its exact provenance is not known.

The speleothem was bisected longitudinally and one half preserved for tourist display. From top and bottom of the other half, samples were taken for alpha particle spectrometry uranium series disequilibrium dating. The remaining portion was 250 mm tall and was sampled at 5 mm intervals using a 2 mm drill for oxygen-18 and carbon-13 isotopic analysis (Fig. 2).

In humid temperate cave environments, calcite deposition usually takes place under conditions of high humidity and near constant temperature, approximately equal to the mean annual temperature at the surface. Exceptions to this are likely to be found only in close proximity to cave entrances. Because temperatures in Royal Cave have been strongly affected by tourist traffic, the air temperatures used for this study were those observed by Canning (1985) in Moons Cave, a former tourist cave in close proximity to Royal Cave. She recorded a mean annual temperature of 15°C, with a temporal variation of 1.3°C. Relative humidity measurements at six stations within the cave showed an average relative humidity of 92–93%.

Geochronology

Age determination of the speleothem (RO) was attempted originally by alpha particle spectrometry of two samples taken from the base and top of the speleothem. Dating was unsuc-

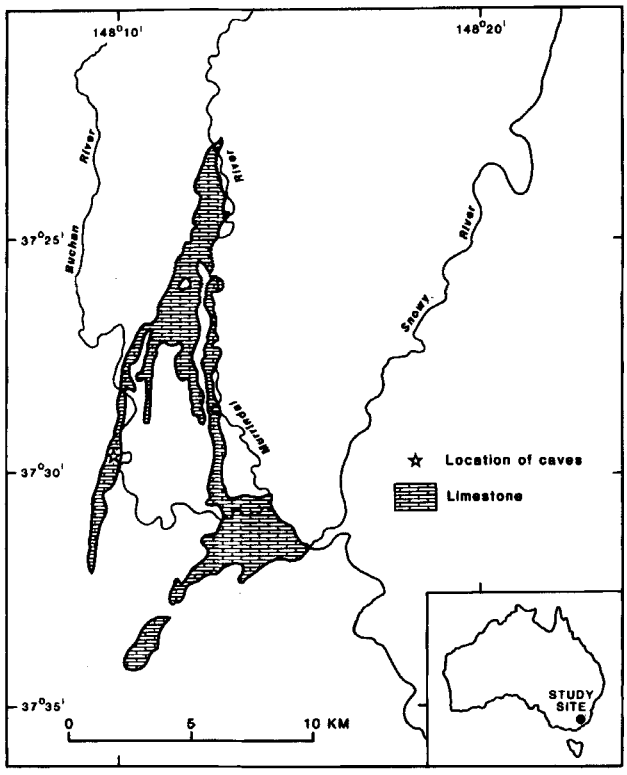

Figure 1 Location map of the study area and the extent of Devonian limestone (after Webb *et al.*, 1992).

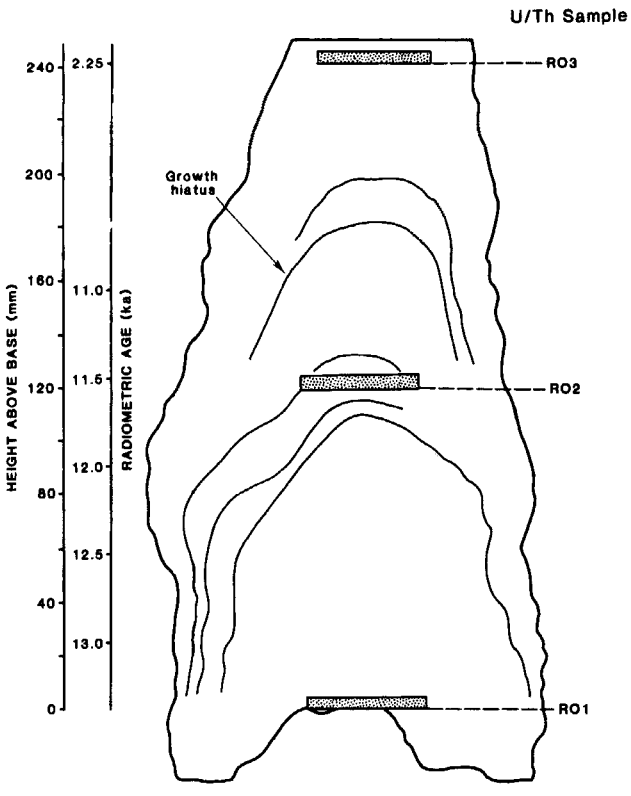

Figure 2 Longitudinal section of RO speleothem showing nature of depositional layering and positions of MS uranium series dates. The time-scale shown is based on these dates.

cessful because of the very low uranium content of between 0.063 and 0.102 μg/g.

Subsequently three samples were taken from the base, middle and top of the speleothem for MS U/Th analysis as shown in Fig. 2. Analytical details of these samples are given in Table 1. The basal and middle samples were analysed twice. The age determinations suggest that the growth hiatus at 180 mm, indicated by an abrupt colour change from milky white to translucent calcite, represents a significant time gap in the depositional record (Fig. 2). A linear growth rate was assumed in order to calculate a time-scale for the lower section of the stalagmite, using sample RO1 together with the average of the two age determinations for RO2. The lower section appears to have been deposited between approximately 13.4 and 10.6 ka indicating a growth rate of some 65 mm ka^{-1}. The upper section is dated by only one analysis (RO3), yielding an age of 2.25 ka at 242.5 mm. On the assumption that the upper section grew at a similar constant rate to the lower section, deposition is found to have occurred from 3.2 to 2.1 ka.

Nature of stable isotopes

The study requires a consideration of the stable isotopes of hydrogen, oxygen and carbon. Isotopic measurements are expressed in per mille (‰) using the delta notation. δD and $\delta^{18}O$ are measured relative to the SMOW (standard mean ocean water) standard where measurements relate to water. $\delta^{18}O$ and $\delta^{13}C$ values relating to calcite are measured relative to the PDB (Peedee Belemnite) standard. The following subscripts are used: p (precipitation), w (seepage water) and c (calcite).

The isotopic composition of precipitation normally tends towards isotopically lighter values with increasing latitude, altitude and continentality (Dansgaard, 1964) and shows significant seasonal fluctuations. Seepage water, collected as roof drips in caves shows little isotopic variation throughout the year, and in southeastern Australia its composition tends to approximate to the weighted mean of the isotopic composition of winter precipitation (Goede and Hitchman, 1984).

Isotopic composition of calcitic speleothems, if deposited under conditions of isotopic equilibrium, is determined by the composition of the seepage water from which the calcite is precipitated together with a temperature dependent fractionation effect. Composition may change over time either because of changes in seepage water composition or because of changing cave temperatures. Cave temperatures show little annual variation and in most caves tend to approximate to the mean annual temperature at the surface. If mean annual surface temperatures change over time in response to a changing climate the cave temperature will adjust accordingly.

In the vast majority of the world's karst areas, the oxygen isotope values of speleothems are negatively correlated with mean annual cave temperature — the temperature effect. The Buchan speleothem clearly fits this pattern (Fig. 4), with $\delta^{18}O$ values apparently less negative at times of cooler temperatures. However, at Mole Creek and in the Florentine Valley of Tasmania (Goede and Hitchman, 1984; Goede *et al.*, 1986, 1990) and at Vancouver Island in western Canada (Gascoyne *et al.*, 1980, 1981) the relationship between $\delta^{18}O$ values of speleothems and temperature is positive. The differences between the Tasmanian sites and Buchan are surprising as they are only 500–600 km apart. There are also significant differences between Victorian and Tasmanian sites in terms of the present-day isotopic composition of vadose dripwaters, which are 1 to 1.5‰ heavier in Tasmania than at Buchan.

The contrast is also shown by the different isotopic compositions of rainfall at Cape Grim, an atmospheric baseline

Table 1 Uranium series MS data and radiometric ages

Sample	^{238}U ($\mu g\ g^{-1}$)	$^{234}U/^{238}U$	$^{230}Th/^{232}Th$	^{230}Th ($pg\ g^{-1}$)	$^{230}Th/^{234}U$	Age (yr)
RO1	0.07953 ± 0.00010	1.34131 ± 0.00038	11.25 ± 0.31	0.2017 ± 0.0041	0.1160 ± 0.0023	13,343 + 294-292
RO1 (repeat)	0.07600 ± 0.00010	1.36327 ± 0.00030	Not measured	0.1893 ± 0.0097	0.1121 ± 0.0056	12,863 + 699-694
RO2	0.06308 ± 0.00008	1.3616 ± 0.0019	33.36 ± 0.82	0.1414 ± 0.0032	0.1006 ± 0.0022	11,540 + 293-292
RO2 (repeat)	0.08170 ± 0.00015	1.3253 ± 0.0055	29.11 ± 0.41	0.1768 ± 0.0024	0.1001 ± 0.0013	11,480 + 215-213
RO3	0.03335 ± 0.00010	1.3154 ± 0.0018	4.35 ± 0.051	0.01465 ± 0.00017	0.02041 ± 0.00023	2,253 + 30-30

station in northwestern Tasmania (150–200 km northwest of the Tasmanian cave sites), compared with Melbourne (300 km west of Buchan). These two stations are the closest sites to the Tasmanian cave sites and Buchan, respectively, for which detailed rainfall isotopic data are available. Despite the fact that the Cape Grim station is south of Melbourne and considerably colder (mean annual temperatures of 12.3°C and 15.4°C respectively) it has isotopically heavier rainfall. Arithmetic/weighted means for Cape Grim are −3.8/−4.4‰, compared with −4.6/−5.1‰ for Melbourne (Rozanski *et al.*, 1993).

Cape Grim oxygen isotope data also show a strong seasonal variation with more negative values in the winter months, a typical pattern for the mid-latitudes (Rozanski *et al.*,1993; Desmarchelier, 1994). There is a statistically significant positive correlation between monthly $\delta^{18}O$ values and mean monthly temperatures. In contrast to Cape Grim, the Melbourne data display an anomalous pattern. The $\delta^{18}O$ values are lighter than might be expected for that latitude. They display a poorly defined winter minimum and a pronounced maximum in October–December. There is no significant statistical correlation between monthly values and mean monthly temperatures. The causes of these features are related to the origin of the air masses that precipitate Melbourne's rainfall (Wright, 1988) and will be discussed in more detail elsewhere (Webb, in preparation). Although no isotopic record of precipitation is available for the Buchan area, isotopic data on cave dripwaters, to be presented in this paper, indicate similarities between Melbourne and Buchan.

Interpretation of the carbon isotope variations within speleothems deposited under conditions of isotopic equilibrium have long been a contentious issue. Much relevant research has been done in recent years and three explanations can now be advanced to account for changes within a single speleothem.

1. Changes in the relative abundance of two groups of plants that follow different photosynthetic pathways: C3 and C4 (Brooke *et al.*, 1990; Dorale *et al.*, 1992; Talma and Vogel, 1992). Because C4 plants are predominantly tropical grasses (Teeri and Stowe, 1976; Tieszen *et al.*, 1979) this explanation is not relevant to the Buchan area.
2. Changes in vegetation productivity, which control the amount of isotopically light organic matter supplied to the soil, which is broken down to provide CO_2 to the soil atmosphere, where it mixes with CO_2 derived directly from the atmosphere. Isotopic variations in soil carbonates (Quade *et al.*, 1989) and subaqueous wallcrust in a karst spring (Coplen *et al.*, 1994) have also been attributed to this cause.
3. Changes in the isotopic composition of carbon in atmospheric CO_2, with glacial periods being characterised by less negative values. The most reliable estimate so far suggests a change of 0.7‰ (Marino *et al.*, 1992). If this estimate is confirmed, the effect of changes in the isotopic composition of atmospheric CO_2 on that of the soil is likely to be quite small. More significant are changes due to the burning of fossil fuels, estimated at −2.2‰ since 1900 (Baskaran and Krishnamurthy, 1993), and instrumental records show a change of −0.23‰ from 1982 to 1993 (Francey *et al.*, 1995).

Isotopic sampling

Before palaeotemperature work could be done on any speleothems from the Buchan Caves area it was necessary to sample modern seepage waters and actively growing straw stalactite tips in order to determine the isotopic composition of both. As Royal Cave is an active tourist cave it was not feasible to collect samples there, so they were collected in nearby Moons Cave.

Water sampling

It was feasible to visit the area only once, in January 1992, to collect samples. Bottles to collect drip water had been set up a fortnight earlier to ensure representative samples for analysis. This was regarded as adequate because it has been shown elsewhere that there is little seasonal variation in the composition of dripwaters in humid temperate environments (Goede *et al.*, 1982; Yonge *et al.*, 1985). Five water samples were collected.

Sampling of modern calcite

In order to estimate the oxygen and carbon isotope composition of modern calcium carbonate being deposited under conditions of oxygen isotope equilbrium, actively growing straw stalactite tips were collected from the same area of the cave as the drip waters. Because of the rapid growth rate of straw stalactites the assumption can be made that the material has been deposited during the last few years (Harmon *et al.*, 1978).

Speleothem sampling

The speleothem (RO) had been cut in two along its length to provide a longitudinal section, as shown in Figure 2.

To determine whether deposition of the stalagmite had taken place under conditions of oxygen isotope equilibrium, three sets of eight 2-mm-diameter samples for isotopic analysis were drilled from each of three growth layers.

Samples for isotopic analysis were also obtained by drilling along the core at 5 mm intervals using a 2 mm drill. This provided a set of 49 samples that could be used to construct a dated longitudinal profile of secular change in the values of $\delta^{18}O_c$ and $\delta^{13}C_c$.

Isotopic analysis

Water samples

Water samples were analysed to determine both deuterium–hydrogen and oxygen isotope ratios. Samples for δD_w analysis were prepared by converting water to hydrogen by reaction with zinc shot at 450°C, using the technique as modified by Kendall and Coplen (1985). Gas samples were then measured on a VG Micromass 602D mass spectrometer with an analytical precision of 2 per mille. For $\delta^{18}O_w$ determination water samples were equilibrated with CO_2 gas at 25°C for 5 h in a VG Isoprep 18. The gas samples were then dried and analysed on a VG SIRA Series II mass spectrometer with an analytical precision of 0.05 per mille. The results of both sets of analyses are shown in Table 2.

As can be seen from Table 2, δD_w values range from −40 to −45‰, with an average value of −43‰, and $\delta^{18}O_w$ values range from −6.20 to −6.74‰, with an average value of −6.44‰. The general relationship between δD and $\delta^{18}O$ values is well known. According to Dansgaard (1964) the so-called meteoric water line is best expressed as

$$\delta D = 8\delta^{18}O + 10‰ \quad (1)$$

Substituting the average value of $\delta^{18}O_w$ (−6.44‰) in this expression yields a δD_w value of −41.5‰, which falls well within the range of measured values.

When these isotopic values are compared with the oxygen isotope record of Melbourne precipitation, we find that the values fall within the lower part of the range for monthly means during the winter months. When compared with the 10 yr (1982–1991) record at the Cape Grim baseline station, which has a similar altitude to the Buchan site, it is clear that the Buchan waters are much lighter isotopically despite the lower latitude. Isotopic values at two inland Tasmanian karst sites (Mole Creek and the Florentine Valley) are also significantly heavier despite their higher latitude and elevation (Goede *et al.*, 1986–1990). The reasons for these differences will be explored in detail elsewhere (Webb, in preparation).

Modern calcite

For isotopic analysis of all calcite samples, CO_2 was prepared by reacting samples in a vacuum with anhydrous H_3PO_4. Both $^{18}C/^{16}C$ and $^{13}O/^{12}O$ ratios were determined as ‰ PDB using a VG SIRA Series II mass spectrometer with an average measurement precision of approximately 0.01‰. The oxygen-18 and carbon-13 values of nine modern calcite samples determined are shown in Table 3.

Correlation of the two data sets gives $r = 0.836$, a clear indication that some of the samples have been fractionated. The two samples least likely to have been affected are MS4 and MS5 because they have the lightest values for both isotopes. Oxygen isotope equilibrium deposition in Moons Cave under present-day conditions can be estimated by a $\delta^{18}O_c$ value of approximately −5.8‰ PDB, corresponding to a $\delta^{13}C_c$ value of approximately −9.6‰ PDB.

Cave temperature calculation

In order to see if the estimates for $\delta^{18}O_w$ for cave dripwaters and the $\delta^{18}O_c$ of modern calcites are approximately correct, an independent estimate of present-day cave temperature can be made using these values by applying the expression of Craig (1965)

$$t = 16.9 - 4.2(\delta_c - \delta_w) + 0.13(\delta_c - \delta_w)^2 \quad (2)$$

This requires the conversion of $\delta^{18}O_w$ values to the PDB standard by adding 0.22‰ (Craig, 1961).

Using the mean value of dripwater, −6.44‰, SMOW (−6.22‰ PDB) and a value of −5.80‰ PDB for modern calcite and substituting these in expression (2) we obtain $t = 15.16$°C. This value is well within the range of temperatures actually measured in Moons Cave by Canning (1985).

Speleothem calcite

Analyses of the three sets of eight growth layer samples are presented in Fig. 3. In all three sets there is an absence of correlation at the 5% level of significance between the pairs $\delta^{18}O_c/\delta^{13}C_c$, $\delta^{18}O_c/D$ and $\delta^{13}C_c/D$, where D is the distance along the growth line from the centre of the stalagmite core. It indicates that the speleothem has been deposited under conditions of isotopic equilibrium. Another test of such equilibrium is made by correlating the $\delta^{18}O_c$ and $\delta^{13}C_c$ values of the 49 samples taken along the length of the core. Correlation analysis yields $r^2 = 0.181$. The lack of strong correlation provides further evidence of isotopic equilibrium deposition.

The results of stable isotope analysis of samples taken along the length of the core are provided in Fig. 4, where five-point

Table 2 Deuterium–hydrogen and oxygen-18 determinations of five dripwater samples from Moons cave, Buchan area

Sample	δD_w (‰ SMOW)	$\delta^{18}O_w$ (‰ SMOW)
MN1	−44	−6.45
MN2	−43	−6.47
MN3	−40	−6.32
MN4	−45	−6.74
MN5	−42	−6.20
Average values	−43	−6.44

Table 3 Oxygen-18 and carbon-13 values determined for nine straw stalactite tips from Moons Cave, Buchan area

Sample	$\delta^{18}O_c$ (‰ PDB)	$\delta^{13}C_c$ (‰ PDB)
MS1	−5.74	−9.31
MS2	−5.59	−7.50
MS3	−5.76	−8.86
MS4	−5.85	−9.53
MS5	−5.82	−9.61
MS6	−5.68	−8.26
MS7	−5.55	−8.42
MS8	−5.35	−7.03
MS9	−5.62	−9.40

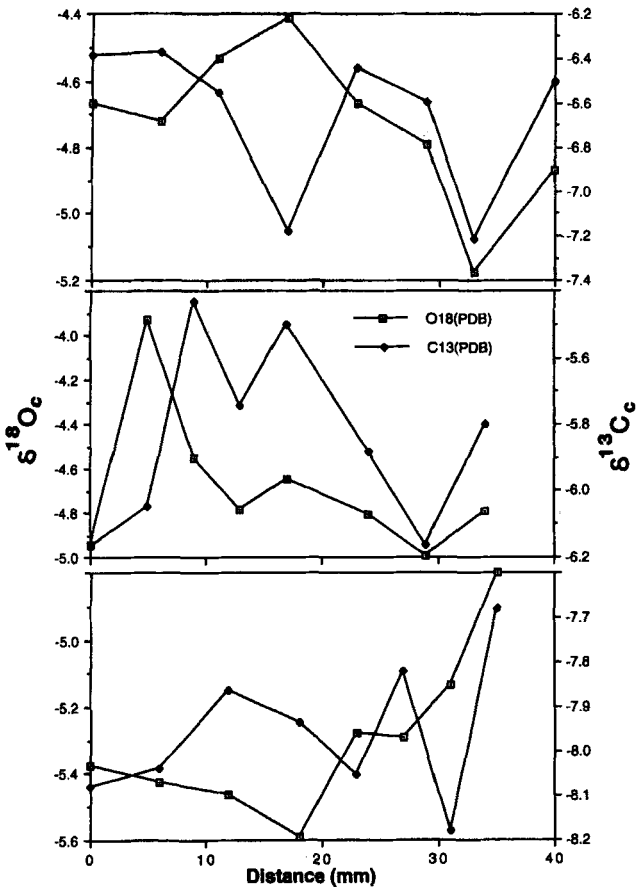

Figure 3 Patterns of $\delta^{18}O_c$ and $\delta^{13}C_c$ values with distance from the centre along each of three growth layers to test for deposition under conditions of isotopic equilibrium.

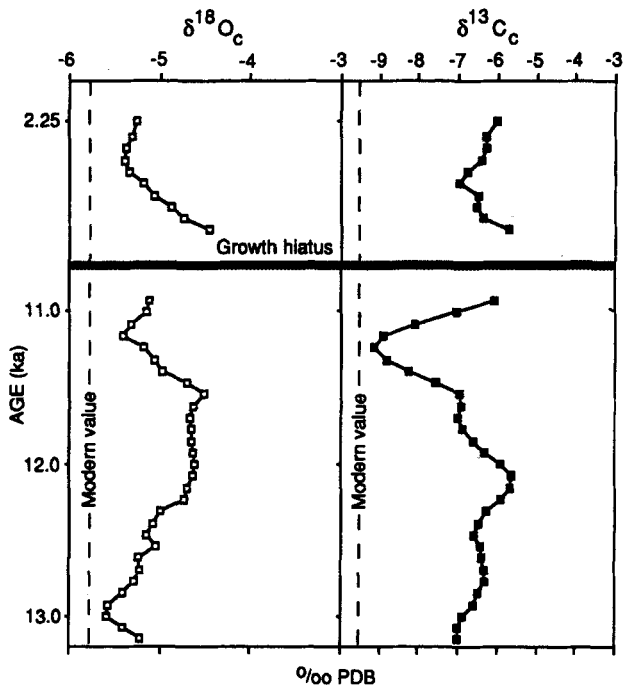

Figure 4 Five-point running means of temporal variations in the values of $\delta^{18}O_c$ and $\delta^{13}C_c$ measured at 5-mm intervals along the longitudinal axis of the stalagmite (RO) from Royal Cave, Buchan. The modern values shown on the diagram have been determined from straw stalactite tips at nearby Moons Cave. The $\delta^{18}O_c$ values bear a negative relationship to mean annual temperature.

smoothed values are plotted against estimated age on the assumption of a uniform growth rate. The $\delta^{18}O$ values are consistently heavier isotopically than the modern value of −5.8‰. Because world temperatures are known to have been lower than today during much of the time span covered by the stalagmite, it confirms a negative relationship between $\delta^{18}O$ values and mean annual temperature. This is the relationship found in most of the world's stalagmites examined so far but is opposite to that found in the nearby island of Tasmania (Goede *et al.*, 1986, 1990).

Temperatures thus appear to have been lower than today throughout the two growth periods represented in the depositional record, although it is not possible to make a precise estimate of how much lower they may have been.

Two particularly cool periods are indicated. The older one from 12.3 to 11.4 ka clearly corresponds to the Younger Dryas stadial in Europe. The younger cold climate event is indicated at about 3 ka and coincides with the resumption of speleothem deposition following a major growth hiatus between 10.7 and 3.0 ka.

Running mean isotopic values for $\delta^{13}C_c$ vary from approximately −5.8 to −9.2‰. As discussed earlier, likely causes have been suggested to account for temporal carbon isotope variations in a single speleothem but only two are relevant to the Buchan area.

The atmospheric explanation is unlikely to account for any changes in isotopic values in excess of 1 per mille nor is it capable of explaining the rapid variations that occurred between 11.5 and 11.0 ka. The changes observed are believed to reflect predominantly changes in vegetation productivity, which in turn are likely to be related to changes in moisture availability during the growing season.

Discussion

Many speleothems from caves in southeastern Australia, including the Buchan area, are characterised by low uranium contents and this makes them difficult to date with any degree of precision by uranium series disequilibrium techniques using conventional alpha spectrometry. The introduction of thermal ionisation mass spectrometry (TIMS) has enabled precise dating of such speleothems. High precision dating makes it possible to compare isotopic records of speleothems with well-dated climatic proxy records from elsewhere in the world.

In the RO stalagmite from Buchan, the period of time between 12.3 and 11.4 ka characterised by high values of $\delta^{18}O_c$ can be correlated confidently with a cold climate event (Younger Dryas) between 12.9 and 11.7 ka in the very precisely dated ice-core record (GISP 2) from Greenland (Taylor *et al.*, 1993). The Younger Dryas event was identified originally in pollen sequences from western Europe, and its age was determined by radiocarbon dating. These ages are now known to be underestimates (Bard *et al.*, 1990). The time discrepancies in the boundary values between the ice-core and speleothem records are within the limits of accuracy of the U/Th MS dates. This is further confirmation of the negative relationship between the oxygen isotope ratio of speleothems and mean annual temperature at Buchan. Until 1994 most evidence for the Younger Dryas event in the Southern Hemisphere has come from pollen profiles in South America and interpretation of the evidence has been a source of considerable debate (Heusser, 1993; Markgraf, 1993). Denton and Hendy (1994) have now presented conclusive evidence that

the Waiho Loop terminal moraine related to an advance of the Franz Josef Glacier on the western flank of the Southern Alps in New Zealand is synchronous with the Younger Dryas in the North Atlantic region.

A Holocene cold climate event is indicated at about 3 ka at the resumption of growth of the speleothem following a major depositional hiatus. This is in good agreement with evidence from two stalagmites from different karst regions in Tasmania with records indicating lower mean annual temperatures (2°–3°C) between 4 and 3 ka (Goede and Hitchman, 1984; Goede *et al.*, 1990). It also confirms evidence from the Snowy Mountains in southern New South Wales by Costin (1972), who presented ^{14}C dated evidence for increased periglacial activity and identified a cold phase from 3000 to 1500 radiocarbon years BP. Williams (1978) has summarised evidence of environmental change from the Southern Tablelands of New South Wales. He found that hillslopes were unstable and streams were aggrading between 4000 and 1500 radiocarbon years BP. He suggested lower temperatures, drier and windier conditions and changes in rainfall seasonality as possible causes.

Although temperature differences cannot be quantified, it is possible to calculate a minimum amount of temperature lowering from the RO oxygen isotope record using formula (2). As stated earlier the speleothem oxygen isotope record is affected by two opposing influences. If the cave temperature, which is assumed to be equal to the mean annual temperature at the surface, drops in response to climatic cooling then the oxygen isotope value of any calcite being deposited tends to become isotopically heavier—the temperature effect. This clearly is the dominant factor at Buchan.

This effect may be counteracted to some extent by changes in the isotopic composition of precipitation, which are controlled by the temperature conditions in the moisture source area and in the local atmosphere where moisture is condensing—the precipitation effect. Precipitation will, therefore, tend to become isotopically lighter as the climate cools and this will produce isotopically lighter seepage waters.

However, if we make the assumption that there was no precipitation effect we can use equation (2) to calculate a theoretical mean annual temperature of 12.9°C for conditions of maximum cold during the two cold periods. This provides a minimum estimate for the amount of temperature lowering of 2.25°C relative to the present.

The time sequence of $\delta^{13}C_c$ values in Fig. 4 is believed to represent a record of varying conditions of moisture availability with more negative values indicating more available moisture during the growth period. The values cannot be compared directly with the modern value of –9.6% obtained from straw stalactite tips in Moons Cave. The reasons are firstly that Moons Cave is some distance away from Royal Cave where the RO speleothem was collected and secondly that burning of fossil fuels has significantly changed the $\delta^{13}C$ value of atmospheric CO_2 in recent decades (Baskaran and Krishnamurthy, 1993).

Conclusions

The stalagmite provides a detailed environmental record of two time periods—the older from 13.4 to 10.6 ka and the younger from 3.2 to 2.1 ka. Oxygen isotope values reveal cooler conditions than today, with a stadial between 12.3 and 11.4 ka, contemporaneous with the Younger Dryas in Europe and a glacial advance in the Southern Alps of New Zealand.

Neoglacial conditions occurred at approximately 3 ka, followed by a gradual warming. There is considerable evidence for colder conditions at this time from both Tasmania and southeastern Australia.

Temperatures during both cold climate events were at least 2.25°C lower than at present and the temperature drop may well have been greater. It is evident that the Younger Dryas in the Southern Hemisphere was nowhere near as severe as it was in Europe. This supports the notion that the factors leading to its occurrence originated in the northern Atlantic. The large extent of the Southern Ocean is likely to have been responsible for reducing the amplitude of temperature variations quite significantly.

Three hypotheses have been proposed to account for secular variations in speleothem carbon isotope values and they have been examined in the light of the evidence available. It is concluded that at Buchan these changes can be interpreted predominantly in terms of variations in moisture availability during the growing season. Isotopic changes in atmospheric CO_2 may have made a minor contribution.

The speleothem data offer a challenge to palynologists to find evidence from high-resolution pollen cores in order to document the impact on the Australian vegetation cover.

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