# Precipitation trapped in datable rock-forming minerals: estimating Antarctic palaeoelevations - a discussion

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**Abstract:** Meteoric water that interacted with minerals during retrogressive metamorphism and hydrothermalism in the late-stage of mountain building processes contains hydrogen and oxygen isotopes that are potential proxies for palaeoelevation reconstruction in Antarctica. The effects of temperature on meteoric isotopic signatures, meteoric crustal infiltration processes, and the mechanisms of capture and preservation of meteoric  $\delta D$  and  $\delta^{18}O$  values in rock-forming minerals are discussed. Special emphasis is given to Antarctica's geographical high-latitude position and climatic fluctuations over time and to the high-mountain ranges of continental Antarctica, which were tectonically active regions in the past. In this context, a new compilation of recent Antarctic snow and ice  $\delta D$  and  $\delta^{18}O$  data is presented, by which we demonstrate that net elevations versus isotopic depletions are positively correlated for continental Antarctica - a prime requisite when estimating palaeoelevations.

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#### Introduction

For more than 50 years, modern Antarctic exploration and geological discoveries have made a major contribution to our understanding of global tectonics and climate history. Here, we discuss the influence of topography on climate and on the isotope composition of precipitation (e.g. Ruddiman & Kutzbach (1989), Ruddiman et al. (1997) and Poage & Chamberlain (2001)), with special emphasis on Antarctica. At present, there are few methods available that allow quantitative estimates of palaeoelevation. Constraints on the exhumation processes of mountains can be derived from metamorphic cooling ages, petrology and sedimentology; but these constraints yield little insight into elevation history. Classical studies for determining palaeoelevation have concentrated on other methods, such as basalt vesicularity (Sahagian & Maus 1994), cosmogenic nuclides (Brook et al. 1995, Fogwill et al. 2004), palaeobotany (Forest et al. 1999), and composition of authigenic minerals (Chamberlain & Poage 2000). Although these palaeoelevation methods are useful, their application in Antarctica is limited for a variety of reasons, including outcrop weathering and amount of available and accessible material.

Given these limitations, it is important to consider other palaeoelevation methods that are applicable over a wider range of geological areas in Antarctica. Pioneering studies, e.g. Barker *et al.* (2000) and Mulch *et al.* (2004) have shown that isotopic ratios of hydrogen in meteoric water trapped in datable rock-forming minerals from previously tectonically active regions can provide a direct record of palaeoprecipitation compositions (latitudinal/altitudinal derivation). For example, Bennett & Barker (1992) and Barker *et al.* (2000) reconstructed palaeo-elevations using  $\delta D$  of aqueous fluid inclusions in quartz in late-orogenic veins within and near the Caledonian thrusts of north Norway, and Mulch *et al.* (2004) estimated palaeoelevations for a crustal-scale extensional fault in the North American Cordillera. Key assumptions of these studies are that modern isotopic lapse rates can be extrapolated to past times; and that elevation/isotopic depletion in precipitation under palaeoclimatic conditions are comparable to elevation/isotopic depletion in precipitation from modern climates.

In this paper, we review the literature on the principals of  $\delta D$  and  $\delta^{18}O$  determinations from (datable) rock-forming minerals, and evaluate the work of Barker *et al.* (2000) and Mulch *et al.* (2004) in the context of its applicability to south polar environments. We provide an extensive compilation of net-elevation versus isotopic depletion in precipitation data (isotopic lapse) across Antarctica, demonstrating that there is a linear relationship between net elevation and isotopic depletion in continental Antarctica. Based on the concept of isotopic lapse rates, and on the new elevation/isotopic depletion data, we then investigate the use of  $\delta^{18}O$  and  $\delta D$  values as potential palaeoelevation proxies for the Antarctic high-mountain ranges.

#### Background

Many studies have documented systematic variations in the

 $\delta D$  and  $\delta^{18}O$  values of precipitation, which can be related to changes in temperature, elevation, distance from the source of moisture, dominant wind patterns, and, in warm climates, precipitation rate (e.g. Dansgaard 1964, Rozanski *et al.* 1993, Fricke & O'Neil 1999). A general understanding of the inter-relationship between composition of precipitation and environmental factors is important for two reasons:

- 1)  $\delta D$  and  $\delta^{18}O$  of terrestrial formations (e.g. minerals, soils, sediments, hard rocks, ice, water, cellulose) can be used as proxies for the isotope composition of palaeoprecipitation (for a review see Shanley *et al.* 1998) potentially making them quantitative indicators of terrestrial palaeoelevations and palaeoclimates, and
- 2) the incorporation of  $\delta D$  and  $\delta^{18}O$  values of water (at a given time) into general circulation models of the atmosphere (e.g. Hoffmann *et al.* 1998) may place constraints in hydrological cycle parameters in these simulations, enhancing exploration of the complex interactions between atmospheric trace gas concentrations, land surface processes and climate.

Recent analyses of pedogenic minerals have confirmed that they form in oxygen isotope equilibrium with soil-water (Cerling & Quade 1993, Yapp 1993, 2000, Savin & Hsieh 1998, Hsieh *et al.* 1998) and that their  $\delta^{18}$ O is strongly correlated with meteoric precipitation compositions (Lawrence & Taylor 1971, Cerling & Ouade 1993, Amundson et al. 1996, Savin & Hsieh 1998). For example,  $\delta^{18}$ O of pedogenic calcite records the  $\delta^{18}$ O values of soil water during dry seasons (Quade et al. 1989), whereas the  $\delta^{18}$ O of pedogenic phyllosilicates and Fe-oxides records the  $\delta^{18}$ O of soil water during the wetter part of the year (Hsieh & Yapp 1999, Stern et al. 1997). Similarly, Rozanski et al. (1993) and Amundson et al. (1996) demonstrated that changes in atmospheric circulation patterns and air mass rainout histories can be correlated with changes in  $\delta^{18}$ O values of pedogenic minerals. Based on these relationships and  $\delta^{18}$ O values of soil-forming minerals, a reconstruction of palaeotopographies of mountain belts was recently presented by Chamberlain & Poage (2000) and Garzione et al. (2000a) based on the strong correlation between change in temperature and  $\delta^{18}$ O values. For the more distant past, using also  $\delta^{18}$ O values of soil-forming minerals, Tabor & Montañez (2002) showed a shift in late Palaeozoic atmospheric circulation over western equatorial Pangea, and Ufnar et al. (2004) reconstructed palaeoprecipitation rates and compositions for pedogenic horizons in Alaska.

The expansion of techniques based on  $\delta^{18}$ O isotopic compositions from pedogenic minerals to the analysis of  $\delta$ D and  $\delta^{18}$ O in rock-forming minerals from formerly active regions was first suggested in the mid-eighties by e.g. Kerrich & Hyndman (1986), Kerrich & Rehrig (1987), McCaig *et al.* (1990) and Smith *et al.* (1991). They concluded that meteoric water strongly interacts with mylonitized rocks in shear zones during extensional deformation. Actual evidence for the interaction of meteoric fluids with rocks from formerly active regions was recently presented by Fricke *et al.* (1992), Barker *et al.* (2000), Dallai *et al.* (2001) and Mulch *et al.* (2004). However, the connection between crustal infiltration of meteoric water and palaeo-atmospheric conditions (e.g. elevation, Barker *et al.* 2000, Mulch *et al.* 2004) is a complex matter requiring an understanding of the influences of precipitation-hard rock interactions on stable isotopes, and also of the mechanisms of infiltration of meteoric water into the crust. Specifically, it is necessary to understand:

- 1) what parameters affect the isotopic composition of modern and fossil meteoric water,
- 2) how meteoric water infiltrates the Earth's crust during tectonism, and
- 3) how meteoric water is physically stored in rock-forming minerals.

# *Effects on the isotopic composition of modern and fossil meteoric water*

# General aspects

Water is composed of two elements that have stable isotopes with variable ratios, deuterium/hydrogen (D/H) and <sup>18</sup>O/<sup>16</sup>O. These provide two independent tracers of water origin. In rocks, oxygen is the most abundant element, making up about 50% by weight. In contrast, hydrogen is usually a trace element being commonly less than 2000 ppm (e.g. Sheppard 1986). The difference between the hydrogen and oxygen concentration in rocks and waters has important consequences. During water-rock interaction, both H- and O- isotope exchange may occur among the various mineral phases. In systems where the water is of external origin, water is generally the dominant hydrogen reservoir and will therefore control the D/H ratio of the system. The  $\delta^{18}O$ value of the water, however, may be determined by mineraloxygen available for exchange, temperature of exchange and water-rock ratio. For typical water-rock ratios, we may expect rocks to buffer the oxygen isotope values of the fluid, but water to buffer the hydrogen isotope ratio of the fluid. Thus, in spite of mineral H<sub>2</sub>O exchange-reactions, the  $\delta D$  values of the waters in meteoric-hydrothermal or meteoric low- to medium-grade metamorphic systems, are often essentially constant - whereas the O-isotope composition of the water is likely to undergo a considerable  $\delta^{18}$ O shift away from its initial values (e.g. Sheppard 1986). This is best seen in high latitude areas, where the isotopic composition of meteoric water is considerably different from any other water retained in the rocks or minerals (i.e. magmatic, metamorphic), because it has been strongly depleted in the heavier isotopes (<sup>2</sup>H, <sup>18</sup>O) (Taylor 1997).

Because it is less likely to be modified by water-rock interaction, the D/H ratio of the water is likely to be more



Fig. 1a. Schematic high-elevation passage of precipitation across a hypothetical mountain range. Vertical and lateral effects on the  $\delta D$  values, infiltration of water into shallow crust, and interaction between meteoric water and crystals are shown (sketch modified after Alley & Cuffey 2001, Bebout *et al.* 2001, Mulch *et al.* 2004). b. Isotope fractionation factors for evaporation of water as a function of atmospheric temperature variation. The fractionation factor for  $^{18}O(\alpha_{18})$  is defined as the ratio of  $^{18}O/^{16}O$  in the liquid to  $^{18}O/^{16}O$  in water vapour in equilibrium with the liquid. The fractionation factor for D ( $\alpha_D$ ) is similarly defined as the ratio of D/H in the liquid to D/H in the vapour. The graph illustrates the temperature dependence of these isotopic fractionation factors. Plotted from values listed by Dansgaard (1964).

useful than the <sup>18</sup>O/<sup>16</sup>O ratio in determining the water origin. However, analyses of  $\delta^{18}$ O values are still useful because they record both the amount of isotopic exchange in a rock, and the equilibrium temperature for mineral-oxygen exchange in metamorphic systems.

Studies of Precambrian submarine and meteoric hydrothermal systems suggest that the isotopic composition of ancient ocean waters (from 2500 Ma) was comparable to present day values (e.g. Taylor 1974, Costa et al. 1983, Sheppard 1986). This observation is supported by additional studies of fossil formation waters (interstitial waters) (e.g. Fontes 1981, Sheppard 1984) and fossil argillaceous soils and supergene weathering (Sheppard et al. 1969, Savin & Epstein 1970, Lawrence & Taylor 1971). The similarity in isotope composition between the fossil and modern meteoric waters is usually given, since both types follow the trend of the modern Meteoric Water Line (MWL). The MWL is defined as a line, around which most modern meteoric waters plot that have not undergone extensive evaporation. They usually lie within a band up to  $\pm 1$  per mil  $\delta^{18}$ O of this line with the relationship  $\delta D = 8 \cdot \delta^{18} O + 10$  (e.g. Craig 1961, Dansgaard 1964, Chamberlain et al. 1995, Butler et al. 1997, Barker et al. 2000). Given the similarity in isotopic composition between fossil and modern waters, it is therefore assumed that ancient meteoric waters can be treated in a similar way to the modern MWL. This is, of course, a fundamental assumption when reconstructing geomorphologic settings by comparing fossil meteoric water data with modern meteoric water data.

Temperature effects with altitudinal variations

As an air mass is forced aloft during its passage over a mountain belt, decreasing atmospheric pressure leads to adiabatic expansion and cooling. In the cooled air, the



Fig. 2. Example of the change in  $\delta^{18}$ O of precipitation versus net elevation change for four different geographical positions. The best fit regression line shown on each graph is forced through the origin. Also shown on the Central and South America plot is the best fit polynomial curve (after Poage & Chamberlain 2001).



Fig. 3a. Schematic map for samples sites in Antarctica from which elevation and  $\delta D$  and/or  $\delta^{18}O$  are known (derivation of raw data: see captions Fig. 3b–e).

saturation vapour pressure of H<sub>2</sub><sup>18</sup>O or HDO is lower than for H<sub>2</sub><sup>16</sup>O (e.g. Dansgaard 1964, Gat 1996). Saturation vapour pressure dictates how much moisture an air mass can hold at a given temperature and increases exponentially with increasing absolute temperature. Reduced saturation vapour pressure of H<sub>2</sub><sup>18</sup>O or HDO relative to H<sub>2</sub><sup>16</sup>O means that an air mass can hold less H<sub>2</sub><sup>18</sup>O or HDO compared to H<sub>2</sub><sup>16</sup>O for a given temperature, and that, during air mass cooling, H<sub>2</sub><sup>18</sup>O or HDO condenses out first. Therefore, by reducing saturation vapour pressure, air-mass cooling is the primary driving force for condensation (Fig. 1). In consequence, during progressive cooling of air masses, as occurs during high-elevation passage, the water that precipitates will be enriched in D or <sup>18</sup>O compared to the water vapour remaining in the air (Alley & Cuffey 2001). Thus, during continued orographic air ascent and cooling, the water vapour in an air mass is progressively depleted in D or <sup>18</sup>O by precipitation ("rain out") (Chamberlain & Poage 2000). The quantitative relationship between elevation change and the hydrogen isotopic depletion of water vapour (hydrogen isotopic lapse rate) and resulting precipitation is usually expressed by the global meteoric



Fig. 3b. Change in δ<sup>18</sup>O values of precipitation versus elevation for Antarctic locations. It is clear to see that locations in the central and northern Antarctic Peninsula (which are exposed to extreme changes in weather) do not show a linear correlation between δ<sup>18</sup>O of precipitation versus net elevation. However, a linear regression line can be fitted for all continental locations. All data (104 samples) are taken from snow samples that were collected in 1m snow pits during the 1990 International Trans-Antarctica Expedition. (Dahe *et al.* 1994). c. change in δ<sup>18</sup>O values of precipitation versus net elevation change for 16 locations across the Antarctic continent, derived in an analogous way as proposed by Poage & Chamberlain (2001). The central and northern Antarctic Peninsula are not contained in these data. Data are taken from: Dansgaard *et al.* 1973 (27 firn samples Victoria Land, 7 firn samples Marie Byrd Land, 7 firn samples East Antarctica); Lorius & Merlivat 1977 (48 snow samples, traverse from Dumont D'Urville to Dome C), Dahe *et al.* 1994 (104 snow pit samples, Trans-Antarctic), Graf *et al.* 2002 (ice core samples), Mulvaney *et al.* 2002 (2 ice core samples, Berkner Island), Stenberg & Hansson unpublished (19 snow pit samples, Dronning Maud Land). The best fit regression line is forced through the origin.



Antarctica

Fig. 3d. Change in δD values of precipitation versus elevation for Antarctic locations. It is clear to see that locations in the central and northern Antarctic Peninsula (which are exposed to extreme changes in cyclonic weather) do not show a linear correlation between δD values of precipitation versus net elevation. However, a linear regression line can be fitted for all continental locations. All data are taken from 1m snow pit samples that were collected during the 1990 International Trans-Antarctic Continent, derived in an analogous way as proposed by Poage & Chamberlain (2001). The central and northern Antarctic Peninsula are not contained in these data (data taken from Dahe *et al.* 1994 (same snow pit samples as referred to in Fig. 3b & c and Lorius & Merlivat 1977). The best fit regression line shown is forced through the origin.

x+504.6

-100

140

4000

water line (MWL) (Craig 1961). Deviations from this general expression, e.g. variations due to geographical position and topographic features (see Sheppard 1986), can be included in empirical expressions for oxygen isotopic depletion of precipitation (oxygen isotopic lapse rate).  $\delta^{18}$ O derived from the Nepal Himalaya (Garzione *et al.* 2000b) for example follows the expression

-150

-8.2

250

81D (%) SMOW

-350

-450

Ō

= - 0.054

West

1000

Antarcti

2= 0.900

0.049y-207

Antarc

- 0.075x-18

2000

altitude (m)

 $= -072x_{c}$ 

Sout

Pole

3000

2= 0.897

$$\delta^{18}O_{\mu} = -(2.61E-7)h^2 - 0.0013h - \delta^{18}O_{el}$$
(1)

where *h* indicates elevation above sea level and  $\delta^{18}O_{sl}$  the isotopic composition of meteoric water corresponding to that at sea level.  $\delta D$  is treated in an analogous way using the MWL relationship, although observational data show less depletion at high elevations than models predict (Dessler & Sherwood 2003). However, it is worth noting that the absolute isotope depletion is predominantly a function of air temperature and that regional correlations between elevation and isotopic depletion are limited to stable, e.g. continental, climates. Figure 2 shows the linear relationship between elevation and depletion in  $\delta^{18}O$  values of surface waters with increasing elevation for isotopic datasets collected throughout the world (after Poage & Chamberlain 2001).

Antarctic snow, firn and ice core data

2000

net elevation (m)

Specific datasets from (predominantly southern) high latitudinal areas related to past and modern water compositions at elevation are available from several sources:

Terre

3000

- 1) Firn samples from Victoria Land, Marie Byrd Land and East Antarctica (Craig 1961, Dansgaard *et al.* 1973),
- 2) snow samples from East Antarctica (Lorius & Merlivat 1977),
- 3) snow pit samples from the 1990 International Trans-Antarctica Expedition (Dahe *et al.* 1994),
- 4) the Vostok core from central East Antarctica (Petit *et al.* 1999),
- 5) snow pit samples from the Millenium expedition (Nikolaev & Valeriy 2000),
- 6) palaeosol analyses by Retallack *et al.* (2001) from the Sirius Group (Transantarctic Mountains),
- 7) the ice core record from Berkner Island (e.g. Mulvaney *et al.* 2002),

- 8) firn core (Graf *et al.* 2002) and ice samples (Stenberg 2000) from Dronning Maud Land,
- 9) samples from the EPICA core from Dome C, East Antarctica (EPICA Community Members 2004) as well as from
- 10)the GRIP and GISP2 ice cores from central Greenland (Grootes & Stuiver 1997, Johnsen *et al.* 1997).

Based on the Vostok ice cores, Petit *et al.* (1999) suggested that climate has almost always been in a state of change during the past 420 Ka but within stable bounds; that is, there are maximum and minimum values of properties between which climate oscillates; however, the recent EPICA data from Dome C, which extends the record back to 800 Ka suggests that variations might be greater over longer timescales (EPICA Community Members 2004).

Cross-Antarctic compilation of elevation versus isotopic depletion data

We present here in analogy with Fig. 2 (Poage & Chamberlain 2001), the first cross-Antarctic compilation of elevation versus isotopic depletion data in order to demonstrate that there is linear relationship between (net) elevation and isotopic depletion in Antarctica. Figure 2 hints at this, but with insufficient data to be conclusive. In Fig. 3, we show compiled modern  $\delta^{18}$ O and  $\delta$ D values for many sites across Antarctica. Figure 3a shows a schematic map of the sample sites and Fig. 3b presents  $\delta^{18}$ O values of

precipitation collected by the 1990 International Trans-Antarctica Expedition (Dahe et al. 1994). It is clear that continental sources (West and East Antarctica) show a linear relationship between increase in elevation and  $\delta^{18}O$ depletion. However, locations on the maritime Antarctic Peninsula show distributions that seem random or which are better described by a more complex polynomial fit. Only the more continentally influenced southern end of the peninsula shows a linear relationship. Figure 3c represents net elevations versus change in  $\delta^{18}O(\Delta\delta^{18}O)$  by locality from continental Antarctica, including the samples from the 1990 International Trans-Antarctica Expedition (Dahe et al. 1994), and other continental sample sites (details are given in the figure captions; see for data derivation Poage & Chamberlain 2001). It can again be seen that, depletion in  $\delta^{18}O$  occurs along a linear trend with respect to net elevation. Due to the lack of linear correlation between  $\delta^{18}$ O and elevation in the northern and central Antarctic Peninsula, these data are not included in Fig. 3c. Figure 3d & e are an analogous representation of  $\delta D$  values from Antarctica revealing the same linear relationships between increase in elevation and depletion in  $\delta D$ , as already shown for  $\delta^{18}$ O. In the northern and central Antarctic Peninsula,  $\delta D$ values show a random distribution with respect to elevation (best fit might be polynomials for the central peninsula as shown on Fig. 3b & d). A linear relationship between an increase in elevation (coupled with a decrease in temperature) and  $\delta D$  and  $\delta^{18}O$  depletion is thus demonstrable for modern continental Antarctica.



Fig. 4. The worldwide distribution of  $\delta^{18}$ O and  $\delta$ D (number in parentheses) in precipitation. The observation that the  $\delta^{18}$ O value of precipitation is lower at high latitude, the "Latitude Effect", is apparent (modified after Sheppard 1986). (SMOW - Vienna Standard Mean Ocean Water).

These datasets also show that the determination of elevation change from  $\delta D$  or  $\delta^{18}O$  of minerals is made more complicated by factors such as surface temperature, source of precipitation, and distance from evaporative moisture source (e.g. Antarctic Peninsula data). However, the change in the  $\delta D$  or  $\delta^{18}O$  value of surface waters as a result of the elevation effect is relatively large compared to many of these factors, and thus the change in the  $\delta D$  or  $\delta^{18}O$  value of surface water is presumably a first-order effect that will strongly influence the isotopic composition of fluids involved in deformation and/or low- to medium-grade metamorphic processes (Sheppard 1986). Also, past changes in atmospheric circulation and other climatic parameters (e.g. mean annual temperature) may render the interpretation of palaeo  $\delta^{18}$ O and  $\delta$ D values more difficult (Amundson et al. 1996, Edwards et al. 1996, Boyle 1997, Fricke & O'Neil 1999, Pierrehumbert 1999). However, a variety of theoretical models is available to treat these uncertainties, including sedimentary and fossil records, and Rayleigh distillation models of atmospheric vapour combined with meteorological evolution of air "parcels" for individual precipitation events (e.g. Covey & Haagenson 1984) and locations (Boyle 1997, Pierrehumbert 1999). Similar treatments, coupled to Global Circulation Models (GCM), have produced  $\delta^{18}$ O maps for the modern Earth and provided estimated  $\delta^{18}$ O values for the last Glacial Maximum (e.g. Hoffmann & Heimann 1997). Application of the theoretical models to palaeo  $\delta^{18}$ O proxy data, however is only as good as the knowledge of the relevant meteorological parameters of past climates. A recent twostep regression technique, which needs to be tested for past climates, and deconvolving the effects of altitude and latitude, is now available (Bowen & Wilkinson 2002). This describes  $\delta^{18}$ O values from precipitation for all continents as

# $\delta^{18}O = -0.0051 (|LAT|)^2 + 0.1805 (|LAT|) - 0.002 (ALT) - 5.247 (2)$

However, discrepancies between theoretical expressions and empirical ones still occur. These might reflect variations in specific data collection resolution and regional environment (e.g. moisture transport, variation in seasonal temperatures, latitude (Bowen & Wilkinson 2002)).

In addition to depletion of heavy isotopes (e.g. Dansgaard 1964, Poage & Chamberlain 2001), processes of evaporation, condensation, and rain-out of water from air masses during high-elevation passage also localize extreme lateral gradients in the amounts of precipitation, with high amounts on the windward side and low amounts on the leeward side of topographic highs (Stern & Blisniuk 2002). Principal palaeo wind-directions can be derived from cross-belt  $\delta D$  analyses based on the extreme depletion of <sup>2</sup>H rainfall on the rain-shadow side of a mountain belt compared to its lee side.

# Temperature effects with latitudinal variations

The hydrogen and oxygen isotope signature of meteoric water varies more or less linearly with latitude - this is especially true for high southern latitudes with their relative geographic simplicity (Alley & Cuffey 2001). Figure 4 shows the variations in the isotopic composition of precipitation depending on latitudinal position. This process assumes equilibrium evaporation at the ocean surface (in practice, kinetic isotope effects are involved) followed by continuous condensation from atmospheric water resulting in precipitation that is continuously depleted in  $\delta D$  and  $\delta^{18}O$ (Sheppard 1986). Since the dominant source for water precipitation on land is evaporation from warm low- and mid-latitude oceans, cooling from movement of air poleward away from these sources, or upward movement through convection, frontal activity, or orography, produces progressive depletion of D and <sup>18</sup>O in water in the air.

Air masses transported to colder geographic regions are cooled systematically and the amount of vapour remaining in the air decreases systematically as the temperature decreases. Hence, colder high-latitude places, like Antarctica, have precipitation with systematically lower  $\delta D$ and  $\delta^{18}O$  (e.g.  $\delta D$  -440%;  $\delta^{18}O$  -55% (Epstein & Sharp 1967) (see Figs 1b, 3a-e & 4) and these are significantly different from magmatic or metamorphic waters (Taylor 1997). The empirical relationship between elevation and latitude and the effect on today's stable isotope compositions ( $\delta D$ ,  $\delta^{18}O$ ) in precipitation were empirically shown by Poage & Chamberlain (2001) (see Fig. 2) and a theoretical calculation routine including latitudinal effects on isotope fractionation was proposed by Bowen & Wilkinson (2002). These analyses are important for minimizing latitudinal-related temperature bias when performing net elevation versus change in isotope composition studies.

# Latitudinal bias in Antarctica

We discussed above that isotopic depletion of an air mass is a first-order effect of temperature. In this sense, both, latitudinal and altitudinal effects are second-order expression of temperature variations.

It is generally assumed that the Antarctic continent was at a constant position geographical position at high latitudes for ~380 Ma (Scotese & McKerrow, 1990) and that Palmer Land (central and southern Antarctic Peninsula) had a constant position for at least 100 Ma (e.g. Vaughan & Storey 2000, Vaughan *et al.* 2002). However, despite Antarctica's stable geographical position at high latitudes, temperature and other climatic parameters are known to have fluctuated over time (e.g. Barrett 1991, Zachos *et al.* 1994, 1996, 2001, Francis 1996, 1999, 2000, Ziegler *et al.* 1997, Scotese *et al.* 1999, Poole & Francis 2000, Poole *et al.* 2000a, 2000b, Isbell *et al.* 2003, Crame *et al.* 2004, Eklund *et al.* 2004) inducing climatic variations that were similar to modern day latitudinal variations. Therefore, when attempting to calculate palaeoelevations from  $\delta D$  and  $\delta^{18}O$  values derived from crystal lattices or fluid inclusions, we have to constrain the regional Antarctic climate (e.g. temperature, air humidity, precipitation) of the study area at the time when the crystal interacted with the precipitation. We have to ensure that the climate of the tectonically active regions was stable (e.g. continental), so that the modern isotopic lapse (rates) for continental Antarctica can be extrapolated to past times. Then, by comparison with similar modern climates (with known or derivable isotopic lapse rates), Antarctic palaeoelevations can be estimated with the smallest possible "latitudinal" bias.

## Infiltration of meteoric water into the Earth's crust

Fluid flow within the upper brittle crust can be described in terms of topographic and thermal driving forces (e.g. Koons & Craw 1991). In recent years, it has been established that meteoric fluids from precipitation may penetrate to depths of typically 5-12 km within orogenic belts (e.g. Wickham & Taylor 1987, McCaig 1988, Jenkin et al. 1994, Morrison 1994, Miller & Cartwright 1997, Bebout et al. 2001). Infiltration times can vary between several hundreds, to tens of thousands, to millions of years, depending on factors such as permeability, fracture systems and rock rheology at depth (e.g. Aquilina et al. 2004). Also, the amount of infiltration in such systems is variable and depends strongly on the physical rock properties (e.g. fracture network, connectivity of pores). Assuming isotope differences between infiltrating fluid and host-rock, a zero-dimensional water-rock interaction model (Taylor mixing model), widely applied to metamorphic rocks (e.g. Bebout & Carlson 1986, Burkhard & Kerrich 1990, Hoernes et al. 1994), can test fluid-rock ratios for a variety of assumed initial (e.g. meteoric) or measured (e.g. from air trapped in snow, ice or firn for Antarctica)  $\delta^{18}O$  or  $\delta D$  values. Unusually low  $\delta^{18}O$  or  $\delta D$  values recovered from metamorphic rocks might point towards considerable meteoric infiltration (e.g. Sheppard 1986).

Although there remains uncertainty regarding amounts of deep infiltration and the precise mechanisms that permit fluid penetration, it is widely regarded that conditions of sub-lithostatic fluid pressure (P, T conditions at the boundary between greenschist and sub-greenschist facies that are ~0.2 GPa to ~0.3 GPa, ~200°C-~300°C) are most common. However, even deeper circulation of meteoric fluids into the ductile lower crust may be achieved by seismic pumping along active fault zones, and by faultvalve action, where dilatancy lowers the fluid pressure in the fault zone and induces subsequent fluid recharge under hydrostatic pressure (Sibson 1992, McCaig 1988). Also, coupling of deformation and fluid flow and an increase in strain rate during deformation is thought to allow fluid infiltration to great depth (up to 35 km) in compressive tectonic settings (Upton et al. 1995, Connolly & Podlachikov 2004).

Case studies for parts of the Canadian Cordillera suggested topographically driven synkinematic infiltration of meteoric fluids along a thrust-fault system (Nesbitt & Muehlenbach 1995, Price et al. 2001, Cooley et al. 2003). Similarly, topographically driven deep fluid infiltration (~9 km), linked with a hydrothermal system, was shown for parts of the Norwegian Caledonides (Barker et al. 2000). For the Nanga Parbat region (Pakistan Himalayas, Chamberlain et al. 1995, Butler et al. 1997), and the Southern Alps of New Zealand (Koons & Craw 1991), active meteoric-hydrothermal convection to greater depth utilized the linked fracture network of late extensional and pre-existing thrust surfaces. fractures These investigations seem to show that late-orogenic processes allowing fluid infiltration are an inevitable consequence of extensional and collisional tectonics, and should be expected in all uplifted metamorphic belts (e.g. Koons & Craw 1991). This has been demonstrated for various Palaeozoic, Mesozoic and younger orogenic belts across the more temperate regions of the world (e.g. Nesbitt & Muehlenbach 1995, Upton et al. 1995, Barker et al. 2000, Price et al. 2001, Mulch et al. 2004). However, no studies have been undertaken to date of any of the Antarctic orogens.

Conservation of  $\delta D$  versus  $\delta^{18}O$  of meteoric fluids during crustal infiltration

So far, we have discussed the fractionation behaviour of  $\delta D$ and  $\delta^{18}$ O with respect to altitude and latitude. Natural waters found in the crust have different stable isotope characteristics, according to their sources and the nature of subsequent fluid-rock interaction (e.g. Sheppard 1986). For infiltrating fluids, the original  $\delta D$  characteristics of the fluid interacting with a metamorphic host rock are largely retained, in contrast with the  $\delta^{18}$ O characteristics, because in general hydrogen is consumed and not liberated in hydration reactions (for example the formation of chlorite from the breakdown of garnet). However, in some cases  $\delta^{18}$ O is also conserved, and probably the most spectacular example of meteoric water alteration in a metamorphic rock conserving meteoric  $\delta^{18}$ O values comes from the work of Yui et al. (1995) on the ultrahigh pressure rocks of the Dabie Shan and Sulu terranes of eastern China. The extremely light  $\delta^{18}$ O values (-10.4‰ to -9.0‰) that they derived from all minerals were in equilibrium for the expected metamorphic temperatures (confirming equilibrium isotope fractionation), suggesting latitudinal/ altitudinal alteration with meteoric water before their burial to great depth (> 100 km).

In general  $\delta D$  values are more stable because during hydration reactions in a retrogressive metamorphic regime, hydrogen is not liberated, and fractionation factors between water and hydroxyl minerals (for example biotite) have consistently shown deuterium depletion in the mineral

relative to water (e.g. Taylor 1974, Kyser & Kerrich 1990). Therefore, deuterium-depleted retrogressive fluids can only be realistically sourced from infiltration of originally deuterium-depleted meteoric water. Consequently, a regional spread in  $\delta D$  can very likely be attributed to the original variability of  $\delta D$  of the infiltrating fluids (e.g. variable elevation source) (e.g. Ambach *et al.* 1968, Chamberlain & Poage 2000, Mulch *et al.* 2004). Given that meteoric  $\delta D$  (and/or  $\delta^{18}O$ ) values appear to be conserved during retrogressive metamorphism and hydrothermalism, it should be in principal possible (for known latitudinal positions, e.g. temperatures) to estimate palaeoelevations based on the rain-out effect (Figs 1–3).

Recovery of  $\delta D$  values reflecting the infiltration of meteoric fluids to greater depths during the later stages of an orogeny has been successful from rock-forming minerals (muscovite, biotite, quartz, feldspar, hornblende) from a variety of geodynamic settings. These include extensional detachment zones in the Ruby Mountains-East Humboldt Range core complex (Nevada) (Fricke et al. 1992) and the Shuswap Metamorphic Complex (North American Cordillera) (Mulch et al. 2004); from the compressional transcurrent Alpine Fault in New Zealand (Jenkin et al. 1994), and from late-orogenic quartz veins within and near the Caledonian thrusts of north Norway (Bennett & Barker 1992, Barker et al. 2000). SD values were also recovered from hydrothermal systems related to rifting and pluton emplacement in Victoria Land (East Antarctica) (Dallai et al. 2001).

## Antarctic orogens

Antarctic mountain building processes involve large-scale compressional and extensional tectonics including extensive retrogressive metamorphism and hydrothermalism (e.g. Goodge et al. 1993, Peacock & Goodge 1995, Vaughan & Storey 2000, Curtis 2001, Dallai et al. 2001, Vaughan et al. 2002, Wendt et al. 2003a, 2003b). However, investigations of  $\delta D$  and  $\delta^{18}O$  values in rock-forming minerals focussed so far on exhumed igneous rocks (Willan et al. 2000) and hydrothermal systems in granitoids (Dallai et al. 2001). Willan et al. (2000) presented  $\delta D$  and  $\delta^{18}O$  measurements from Jurassic and Cretaceous granitoids at different localities in Antarctica. They reported unusually low and variable values for both isotopes, which may be attributed to different origins of the water (primary depleted melt, degassing magma, an exchange between the plutons and depleted meteoric water (Willan et al. 2000)) or interaction between all of them. In any case, magmas are difficult candidates for isotope studies of the type required to determine palaeoelevations due to a variety of reasons, including stable isotope exchange between partial melts and hydrothermally depleted country rocks (e.g. Larson & Taylor 1986) and differential pressures between melt and country rock (pressure increase of factor of 2.5 to 3 in the melt) (Taylor & Sheppard 1986). Work by Dallai *et al.* (2001) suggests that late- and post-magmatic processes are more promising. They measured  $\delta D$  and  $\delta^{18}O$  values from hydrothermally altered granitoids from northern Victoria Land (Dallai *et al.* 2001), revealing a consistent dataset of relative  $\delta D$  values that excluded altitude effects, but suggested a cooling episode in the Ross Sea region during late Eocene time.

Based on this discussion, we suggest that continental Antarctic locations with active tectonics, e.g. shearing during late orogenic processes, constitute the best environment for undertaking a study in which  $\delta D$  (and  $\delta^{18}O$ constrain values should palaeoelevations. Accordingly, we propose that studies with the highest likelihood of success should be focussed on uplifted highelevation regions along continental suture zones in Antarctica where extensional/compressional tectonism facilitated fluid infiltration to mid-crustal levels, and which still present today a high relief. The Antarctic Peninsula (AP), the Ellsworth Mountains (EWM) and the Transantarctic Mountains (TAM) form modern high elevation mountain ranges reaching ~2000 m in the AP, ~5000 m in the EWM and ~4000 m in the TAM providing extensive snow and ice free rock outcrops. During the Mesozoic and the Cenozoic, faulting and/or shearing accompanied uplift of these three mountainous regions, making them potential very-high altitude areas propitious for the study of  $\delta D$  values from meteoric water trapped in rock-forming minerals for palaeoelevation reconstructions (large-scale shearing is known to be a prominent driving force for rapid mountain building and uplift to high elevations in all parts of the world and in all geological times, (England & Thomson 1984, England & Searl 1986, Molnar et al. 1993, Wittlinger et al. 1998, Cowgill et al. 2000, Dokka & Macaluso 2001)).

During Mesozoic times, the compressive Eastern Palmer Land Shear Zone (EPLSZ, e.g. Vaughan & Storey 2000) is thought to have generated a high mountain range along the southern part of the Antarctic Peninsula. The region around the EPLSZ is interpreted to form a palaeo-plate boundary (Vaughan & Storey 2000) that was uplifted to highelevation due to active shearing in a convergent setting in Cretaceous time (Vaughan & Storey 2000, Vaughan et al. 2002. Ferraccioli et al. 2003). The large-scale compressional shear movement and successive uplift of the shear zone were accompanied by active faulting permitting meteoric infiltration to mid-crustal depth recorded by greenschist-facies retrogressive metamorphism and latestage hydrothermalism that followed mineral deformation under earlier amphibolite-facies and granulite-facies metamorphic conditions (Wendt et al. 2003a, 2003b, Wendt unpublished data). Glacial erosion or later crustal extension reduced the palaeoelevation to that observed at the presentday.

The present-day TAM, together with the EWM, is dominantly a manifestation of Cenozoic rift tectonics (e.g.



Fig. 5. Oxygen fraction factors: a. Experimental and calculated oxygen isotopic fractionations between quartz and water as a function of temperature (modified after Richter & Hoernes 1988), b. Experimental and calculated oxygen isotopic fractionations between micas and water as a function of temperature (modified after Zheng 1993), c. Experimental hydrogen isotopic fractionation between micas and water as a function of temperature (modified after Suzuoki & Epstein 1976).

Stern & ten Brink 1989, Behrendt & Cooper 1991, Fitzgerald 1994). Variations of the high modern topography in the TAM range and between the TAM and the rifted-off EWM are attributed to differential uplift along the largescale Cenozoic transverse-faults associated with these rift movements (Behrendt & Cooper 1991). However, it has been argued that the fundamental underlying tectonic architecture of the TAM is largely a result of the Ross orogeny during the late Proterozoic to early Palaeozoic (e.g. Stern & ten Brink 1989, Behrendt & Cooper 1991, Fitzgerald 1994). Therefore, in the northern TAM, for example,  $\delta D$  analyses in retrogressive greenschist facies metamorphic rocks, developed along sinistral strike-slip shear zones (360 Ma, Adams & Kreuzer 1984) from the



Fig. 6. Schematic illustration of various mass transfer and diffusion processes that control the rates of isotopic partitioning between a fluid (or gas) and a solid (modified after Manning 1974, Cole & Chacraborty 2001).

Lanterman Fault Zone, might reveal palaeoelevations reached during docking between the Wilson and the Bowers terranes (e.g. Capponi *et al.* 1999). Similarly,  $\delta D$  values from quartz veins related to dextral strike-slip structures (e.g. Capponi *et al.* 1999) would reveal elevations reached during the Cenozoic.

#### Storage of water in minerals

#### Crystalline bound oxygen and hydrogen.

The most fundamental prerequisite for the understanding of storage of water in minerals is to determine (1) mineralfluid fractionations and (2) microscopic processes that enhance isotopic exchange in mineral-fluid and mineralmineral systems.

Isotopic fractionation describes in general the distribution of isotopes between two phases and is defined as the isotopic fractionation factor

$$\alpha_{\rm A-B} = R_{\rm A} / R_{\rm B} \tag{3}$$

where *R* denotes the concentration ratio of the heavy and light isotope (e.g. D/H, or  ${}^{18}O/{}^{16}O$ ) in the phases A and B.

For a variety of mineral phases, the equilibrium fractionations of D/H and <sup>18</sup>O/<sup>16</sup>O have been determined with reasonable success among fluid and mineral phases at hydrothermal conditions (~100°C–300°C), and at higher temperatures ( $\geq 400$ °C), both from experimental exchange studies and theoretical calculations (e.g. O'Neil 1986, Kyser 1987, Criss 1999, Chacko *et al.* 1996). The main minerals studied include quartz (e.g. Matsuhisa *et al.* 1979, Clayton *et al.* 1989a), muscovite (e.g. Suzuoki & Epstein 1976,

fluid inclusion trail in quartz composition: aqueous + vapour bubble



semi-brittle deformation in muscovite (kink-bands) I semi-brittle deformation



ductile deformation in muscovite (elongation, shearing) T<sub>ductile deformation</sub>

semi-brittle deformation in biotite (kink-bands)

T<sub>semi</sub>-brittle deformation





**Fig. 7.** Natural examples of recrystallized and deformed minerals. Material recovered from metamorphic rocks along the EPLSZ on the Antarctic Peninsula. Matsuhisa *et al.* 1979), albite, anorthite (Clayton *et al.* 1989a), phlogopite (e.g. Fortier *et al.* 1994, Chacko *et al.* 1996), calcite (Clayton *et al.* 1989b, Chacko *et al.* 1996) and rutile (Chacko *et al.* 1996). The experiments and calculations were calibrated using mineral-water or mineral-mineral isotope fractionation systems and any of their various combinations (Chacko *et al.* 1996). In Fig. 5, examples of mineral-fluid isotope fractionations are presented as a function of temperature for the minerals quartz, muscovite and biotite. These minerals will be most important for the study  $\delta D$  and  $\delta^{18}$ O values of metamorphic rocks from high southern latitudes.

Important microscopic processes of isotope exchange between minerals and/or minerals and fluids, are recrystallization, solution/precipitation, diffusion (on surfaces, along grain boundaries, through crystal lattices) and replacement of minerals. These processes can be basically grouped into two major categories: diffusion and chemical reaction (Fig. 6). When minerals and fluids are at or very near chemical equilibrium, isotopic exchange can occur commonly through a diffusional mechanism (Cole et al. 1983, Eiler et al. 1992, 1993, Farver 1994, Giletti 1985, 1986, Kronenberg et al. 1984). In contrast, isotope exchange in mineral-fluid systems that are initially far from chemical equilibrium is largely controlled by chemical reactions resulting in noticeable changes in the nature of the solid. These include dissolution-precipitation (Dubinina & Lakshtanov 1997) and recrystallization (Cole et al. 1983, Cole & Ohmoto 1986, Stoffregen 1996, Cole & Chakraborty 2001). Field studies involving the recovery of  $\delta D$  values of hydrous minerals from the high pressure metamorphic rocks of the Monte Rosa nappe in Switzerland showed that they were similar over a very large area compared to the  $\delta^{18}$ O values (Frey *et al.* 1976). They concluded that a fluid phase induced hydrogen isotope equilibration, whereas the lack of oxygen isotope equilibration in the same rocks might be explained by mass balance and/or exchange kinetics.

Isotope exchange controlled by diffusion is typically several orders of magnitude slower than that influenced by chemical reactions (e.g. Cole *et al.* 1983, Stoffregen *et al.* 1994). Despite significant differences observed in equilibration times between these two major pathways, both are dependent to varying degrees on a number of other factors that include temperature, pressure (or water fugacity) (e.g. Cole *et al.* 1983, Stoffregen *et al.* 1994), solution to solid ratio, grain size, surface area (e.g. Cole *et al.* 1983, Stoffregen *et al.* 1994), solution composition, mineral composition and structure, and defects (e.g. Cole & Chakraborty 2001). Figure 7 shows some natural examples of recrystallized, deformed minerals from Antarctica containing crystalline bond H and/or water in fluid inclusions.

Thus, assuming subsolidus equilibrium fractionation between mineral and fluid phase, those parts of a crystal that were transformed by diffusional reactions or recrystallized, reprecipitated or were replaced, will have the  $\delta D$  isotopic signature of the fluid (e.g. Satish-Kumar *et al.* 1998). Under this assumption, the analysis of the interactions between minerals and fluids is a powerful tool for elucidating temperatures, material fluxes, rock and fluid origins associated with ancient and active fluid-rock interaction process in the Earth's crust (Valley 1986).

Fluid inclusions. Water can also be trapped as fluid inclusions in minerals (Fig. 7). Here, growth irregularities during crystallization, or recrystallization, of a mineral in contact with a fluid result in the trapping of small portions of fluid in the solid crystal. Inclusions are categorized as several types: if original irregularities are sealed off during further growth of the host crystal, this yields primary inclusions; healing of fractures formed at some later time yields secondary inclusions. In some minerals, several fluid inclusion generations exist, reflecting the complicated (re)crystallization history of the mineral through time. In such a case, it is easily possible that the fluids were trapped millions of years apart, and that they may show distinct isotope compositions (e.g. Roedder 1984). These distinct isotope compositions can be recovered using different decrepitation temperatures. Best practice in analysing  $\delta^{18}$ O and  $\delta D$ , however, is the use of samples dominated by one generation of fluid inclusions. Traditionally, fluid inclusions in calcite speleothems are used for oxygen and hydrogen isotope analyses because they are unique in containing a high resolution record of both palaeotemperature and isotope composition of palaeogroundwater: They may contain up to 0.1wt% of relict drip water in the form of fluid inclusions that were trapped in the host calcite at the time of growth (Kendall & Broughton 1978, Dennis et al. 2001).

When concentrating on rock-forming anhydrous minerals, quartz is probably one of the main mineral phases that incorporates hydrogen and oxygen. In quartz, hydrogen and oxygen can be found in H<sub>2</sub>O fluid inclusions and in molecular H<sub>2</sub>O or OH bonded to the mineral surfaces (e.g. Aines et al. 1984, Rovetta & Holloway 1986, Cordier & Doukhan 1989). Fractionation between quartz and waterbound oxygen is well documented (see Fig. 5a). However, the incorporation and fractionation behaviour of hydrogen into quartz is not well understood and interpretation remains vague (Simon 2001). Hydrogen isotope fractionation factors for crystalline Si(OH), and water are not available in the literature. Only Kuroda et al. (1982) have presented D/H fractionation analyses between quartz melt and water at high pressure and high temperature (2 GPa, 1300°C). Their experiments revealed that  $\delta D$  depleted from -76% to -101‰ (-99.8‰) in structural water and to -89.1‰ (-87.4‰) in fluid inclusion-like features producing a fractionation factor of  $\alpha = 0.967$ . From these experiments, and based on the consideration that other hydrogen isotope fractionation factors for OH-bearing Si- and Al-rich minerals (like muscovite) are always negative and temperature-dependent, it is reasonable to assume that D/H fractionation can occur between quartz and water (e.g. Simon 2001).

Temperature-related factors may complicate the situation, however. Simon (2001) recently assessed whether structural water and water trapped in fluid inclusions may interact. He argued that at temperatures higher than 200°C, the diffusivity of O and H through the crystal lattice of quartz becomes fast enough that the composition of fluid inclusions do not any longer reflect the original  $\delta D$ . He suggested, therefore, that hydrogen isotope compositions in fluid inclusions can only be considered to reflect the composition of the environmental fluid if the formation temperature was equal to, or below, 200°C.

# Conclusions

In this paper, we have discussed the boundary conditions that control the storage of hydrogen and oxygen in minerals, provided examples of their recovery from various tectonic settings, established the relationship between meteoric isotopic depletion and elevation in modern Antarctica, and presented geodynamic Antarctic environments that allowed fluid infiltration to crustal depth in the past. In detail, we showed that

- 1)  $\delta D$  and  $\delta^{18}O$  values of rock-forming minerals both can reflect meteoric signatures. However,  $\delta D$  values are more likely to be constant during mineral H<sub>2</sub>O exchange compared to O-isotopes,
- ancient and modern meteoric water appears to follow a similar trend in isotopic composition for various geographical settings,
- 3) hydrogen and oxygen depletion of an air mass are mainly temperature dependent (latitude/altitude),
- 4) a latitudinal (temperature) bias in Antarctica is induced by fluctuations in climatic conditions over time despite Antarctica's constant geographical position,
- 5) modern hydrogen and oxygen depletion during highelevation passage of an air-mass correlates positively with increasing elevation in continental Antarctica,
- 6) meteoric  $\delta D$  (and  $\delta^{18}O$ ) can be stored in the crystal lattice of (hydrous) minerals and as fluid inclusions,
- 7) compressional and extensional tectonic settings are best for fluid infiltration, so that meteoric  $\delta D$  and  $\delta^{18}O$  values can be recovered from many orogenic settings, and
- 8) the high-altitude mountain ranges in continental Antarctica are geodynamic environments in which minerals have interacted with meteoric water during late-stage orogenic activity.

What we have shown here, is that the principal application for estimating palaeoelevation using  $\delta D$  and  $\delta^{18}O$  values from datable rock-forming minerals (e.g. Fricke *et al.* 1992, Barker *et al.* 2000, Dallai *et al.* 2001, Mulch *et al.* 2004) is also possible in south polar regions. We have shown that the main prerequisites for such a palaeoelevation proxy study in Antarctica are satisfied assuming that the modern isotopic lapse rates for continental Antarctica can be extrapolated in time due to the nearly constant continental landmass of Antarctica.

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