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Precipitation regime influence on oxygen triple-isotope distributions in Antarctic precipitation and ice cores

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ABSTRACT

The relative abundance of ^{17}O in meteoric precipitation is usually reported in terms of the ^{17}O-excess parameter. Variations of ^{17}O-excess in Antarctic precipitation and ice cores have hitherto been attributed to normalised relative humidity changes at the moisture source region, or to the influence of a temperature-dependent supersaturation-controlled kinetic isotope effect during in-cloud ice formation below −20 °C. Neither mechanism, however, satisfactorily explains the large range of ^{17}O-excess values reported from measurements. A different approach, based on the regression characteristics of $10^3\ln(1+\delta^{17}O)$ versus $10^3\ln(1+\delta^{18}O)$, is applied here to previously published isotopic data sets. The analysis indicates that clear-sky precipitation ('diamond dust'), which occurs widely in inland Antarctica, is characterised by an unusual relative abundance of ^{17}O, distinct from that associated with cloud-derived, synoptic snowfall. Furthermore, this distinction appears to be largely preserved in the ice core record. The respective mass contributions to snowfall accumulation – on both temporal and spatial scales – provides the basis of a simple, first-order explanation for the observed oxygen triple-isotope ratio variations in Antarctic precipitation, surface snow and ice cores. Using this approach, it is shown that precipitation during the last major deglaciation, both in western Antarctica at the West Antarctic Ice Sheet (WAIS) Divide and at Vostok on the eastern Antarctic plateau, consisted essentially of diamond dust only, despite a large temperature differential (and thus different water vapour supersaturation conditions) at the two locations. In contrast, synoptic snowfall events dominate the accumulation record throughout the Holocene at both sites.

1. Introduction

During the past decade, the relative abundance of isotopologues $H_2{}^{16}O$, $H_2{}^{17}O$ and $H_2{}^{18}O$ in meteoric precipitation has been quantified using the dimensionless parameter ^{17}O-excess (Angert et al., 2004), currently defined as $\ln(1+\delta^{17}O) - 0.528\ln(1+\delta^{18}O)$ (Barkan and Luz, 2007).[1] The rationale for this approach is that precipitation has been considered, on a global scale (Meijer and Li, 1998; Barkan and Luz, 2007; Landais et al., 2008; Uemura et al., 2010; Luz and Barkan, 2010), to conform closely to the relationship $\ln(1+\delta^{17}O) = 0.528\ln(1+\delta^{18}O)$. From theoretical considerations, Angert et al. (2004) suggested that the regression line should be slightly offset (positive) from VSMOW; this was subsequently confirmed experimentally, with the exact magnitude of the offset being sample-dependent, e.g. ∼45 ppm for a transect of recent snow in Antarctica (Landais et al., 2008); ∼33 ppm for a collection of 52 diverse meteoric precipitation samples (Luz and Barkan, 2010). Angert et al. (2004) predicted ppm-scale deviations from the regression line, resulting from kinetic fractionation during the diffusive transport of water vapour from the source region (above the ocean surface) into undersaturated air. The same authors also suggested that normalised relative humidity at the vapour source largely controls – and inversely correlates with – the magnitude of ^{17}O-excess, if turbulence in the marine boundary layer is taken into consideration. This was later supported by experimental data (Uemura et al., 2010).

Initial expectation was therefore that ^{17}O-excess measurements of precipitation in the polar regions could be used as a temperature-insensitive proxy for the humidity above the ocean surface at the source region, complementing the long-established 'deuterium excess' parameter (Dansgaard, 1964). Application to the

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[1] $\delta^{17}O$ and $\delta^{18}O$ are defined by $\delta^i O = ({}^iR_{sample}/{}^iR_{VSMOW}) - 1$, where i is 17 or 18. ${}^iR_{sample}$ is the abundance of isotope ${}^iO$ relative to that of ${}^{16}O$ in the sample; ${}^iR_{VSMOW}$ is the abundance of isotope ${}^iO$ relative to that of ${}^{16}O$ in the Vienna Standard Mean Ocean Water international reference material, VSMOW. δ is a dimensionless quantity and is of small magnitude ($-0.07 < \delta^i O < 0.005$ for measurements discussed in this paper), thus commonly reported as 'per mil' (‰).

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ice core record could then provide glacial–interglacial comparisons of ocean surface humidity. The first reported high precision oxygen triple-isotope measurements of Antarctic precipitation were of snow sampled from a series (29) of 1 m-depth pits located along a transect from the coast at Terra Nova Bay to the continental interior at Dome C on the plateau of east Antarctica (Landais et al., 2008); the results showed an essentially constant ^{17}O-excess value of about 45 ppm.

Contemporaneous measurements of an ice core section (age 5–150 ka) from Vostok showed a 20 ppm mean ^{17}O-excess decrease between the Early Holocene (11.7–9 ka before present) and Last Glacial Maximum (~25–20 ka age); this was interpreted as indicative of a 20% relative normalised humidity increase in the average ocean moisture source for East Antarctic precipitation during the same interval. Such an inference is unrealistic, however (Jouzel et al., 2013). The interpretation of the Vostok isotope data was questioned at the time (Miller, 2008), on the basis that the δ^{18}O values of almost all the ice core samples were considerably lower than those of the surface snow transect and it could not be assumed that the apparent insensitivity of ^{17}O-excess to temperature demonstrated by the transect results would also apply at the lower temperatures experienced at Vostok. Furthermore, linear least-squares regression of the Vostok $10^3\ln(1+\delta^{17}O)$ versus $10^3\ln(1+\delta^{18}O)$ data gave – surprisingly – a well-constrained slope of 0.5310 ± 0.0004 (Miller, 2008), rather than 0.528. All precision values reported in this paper refer to the 95% confidence level, unless stated otherwise. The scaling factor of 10^3 is adopted here to give similar magnitude to 'per mil' (‰), as is conventional for reporting isotope δ data. A subsequent study (Winkler et al., 2012) concluded that the ^{17}O-excess results from Vostok may be highly sensitive to local effects and that ice core data from coastal or near-coastal Antarctic sites, such as Talos Dome, are more reliable for the reconstruction of normalised oceanic relative humidity.

A potential complication with the interpretation of oxygen isotope data from polar region precipitation and ice cores is that, at temperatures below about −20 °C, an additional kinetic fractionation, of magnitude dependent on the extent of vapour supersaturation, has been postulated to occur during ice formation within the cloud (Jouzel and Merlivat, 1984). To evaluate how this affects ^{17}O-excess of Antarctic precipitation, a simple Rayleigh distillation Mixed Cloud Isotopic Model (MCIM) (Ciais and Jouzel, 1994), adapted to incorporate ^{17}O, has been applied in several studies (Angert et al., 2004; Landais et al., 2008, 2012a, 2012b; Winkler et al., 2012, 2013; Touzeau et al., 2016). For the modelling, supersaturation within the cloud is assumed to be a linear function of condensation temperature T_c, such that $S = p + qT_c$ (Jouzel and Merlivat, 1984). Usually, p is set to unity and q varied between 0.002 and 0.007 (Schoenemann et al., 2014) to obtain the best fit to experimentally-determined variations of ^{17}O-excess with δ^{18}O. It has been suggested (Schoenemann et al., 2014) that this kinetic fractionation mechanism – with sea ice expansion increasing the area over which supersaturating conditions occur, thereby amplifying the effect of colder temperatures – is actually the primary control on ^{17}O-excess in Antarctic precipitation and that variations in moisture source relative humidity have negligible influence. There is not consensus on this point, as it has subsequently been noted (Pang et al., 2015; Schoenemann and Steig, 2016) that the large heterogeneity of ^{17}O-excess in Antarctic precipitation cannot be explained satisfactorily by either mechanism. Furthermore, results of a recent experimental study (Casado et al., 2016) raise doubts about the formulation of kinetic fractionation as currently implemented in the MCIM approach. Attempts to reproduce ^{17}O-excess data by atmospheric General Circulation Models show that these also are not yet able to quantify with confidence the processes controlling ^{17}O-excess (Risi et al., 2013; Winkler et al., 2013; Schoenemann and Steig, 2016).

Precipitation in the coastal regions of Antarctica is attributed primarily to frontal activity in the circumpolar trough. Further inland, during all seasons except summer, a strong, surface-based temperature inversion persists. Radiative cooling of saturated air or near-saturated air at very low temperatures, in or just above the cold atmospheric boundary layer, causes ice needles (diamond dust) to precipitate from a clear sky. Such events occur almost daily at Vostok, where diamond dust to has been estimated to contribute 75 ± 16% of the annual mass accumulation (Ekaykin et al., 2004). As noted by Stenni et al. (2016), diamond dust formation is not necessarily restricted to clear sky conditions and may also occur below high cloud cover.

At high elevation localities on the Antarctic plateau, the direct influence of coastal synoptic activity is inhibited (Masson-Delmotte et al., 2011). Here, in addition to diamond dust, a variable proportion of the annual snowfall accumulation originates from a relatively small number of synoptic events (Ekaykin et al., 2004; Fujita and Abe, 2006; Schlosser et al., 2010; Dittmann et al., 2016; Schlosser et al., 2016), which result from amplification of Rossby waves in the circumpolar westerlies. At Vostok such events occur, on average, during only ~37 days per year (Ekaykin et al., 2004). Meridional advection of the relatively warm and moist air towards the high altitude interior of the continent results in adiabatic cooling, with precipitation commencing when vapour saturation is attained, resulting in a rapid decrease in the absolute humidity of the transported air.

2. Methods

Isotopic data sets reported in previous studies of Antarctic precipitation, surface snow and ice cores (Landais et al., 2008, 2012a, 2012b; Winkler et al., 2012, 2013; Schoenemann et al., 2014; Pang et al., 2015; Touzeau et al., 2016) are used to provide new insights into the origin of the ^{18}O/^{16}O and ^{17}O/^{16}O distributions. Instead of interpreting the data solely in terms of ^{17}O-excess values, linear least-squares regression (non-weighted, i.e. assuming constant variance in the errors) of $10^3\ln(1+\delta^{17}O)$ with respect to $10^3\ln(1+\delta^{18}O)$ is used to characterise the fractionation behaviour, including the ordinate axis offset of the respective regression lines from VSMOW. Precision of the regression results is reported at the 95% confidence level.

This approach differs from ^{17}O-excess characterisation, which is based on the widely-used definition (Miller, 2002) of the magnitude of any offset ($\Delta^{17}O$) from a reference fractionation line, for when the distinction between $\Delta^{17}O$ and $\ln(1+\Delta^{17}O)$ is smaller than the associated precision of measurement: $\Delta^{17}O = 10^3\ln(1+\delta^{17}O) - \lambda 10^3\ln(1+\delta^{18}O)$. Any value of λ may be assigned, in principle, although if the magnitude differs significantly from that obtained by regression of the sample $10^3\ln(1+\delta^{17}O)$ and $10^3\ln(1+\delta^{18}O)$ measurements, divergence (or convergence) of the respective lines generates a $\Delta^{17}O$ (^{17}O-excess) component as an artefact (Miller et al., 2015). For the ^{17}O-excess definition widely adopted for meteoric waters (after Barkan and Luz, 2007), the reference line was based on the oxygen triple-isotope distribution in meteoric precipitation conforming closely to the relationship $\ln(1+\delta^{17}O) = 0.528\ln(1+\delta^{18}O)$, as first reported by Meijer and Li (1998) and subsequently from high precision measurements of Antarctic shallow snowpit samples (Landais et al., 2008) and from a set of meteoric waters of diverse provenance (Luz and Barkan, 2010).

In some recent publications, δ^{17}O and δ^{18}O values of Antarctic precipitation have been reported to only two decimal places, in conjunction with corresponding ^{17}O-excess data (ppm). In such cases, the published δ^{17}O measurements have been adjusted here so that ^{17}O-excess values calculated in conjunction with the reported δ^{18}O results are identical to the published ^{17}O-excess data.