Rainfall stable water isotope variability in coastal southwestern Western Australia and its relationship to climate on multiple timescales

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Key Points:

- Precipitation δ^{18}O variability is driven primarily by rainfall intensity in southwestern Western Australia
- Interannual δ^{18}O variability is sensitive to the largest events each year
- Humidity during evaporation drives deuterium excess variability at a daily, but not interannual, timescale

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Abstract

The factors driving variability in rainfall stable water isotopes (the ratios of $H_2^{18}O$ and ^2HHO to H_2O, expressed as $\delta^{18}O$, and deuterium excess, d) were studied in a 13-year dataset of daily rainfall samples from coastal southwestern Western Australia (SWWA). Backwards dispersion modelling, automatic synoptic type classification, and a statistical model were used to establish causes of variability on a daily scale; and predictions from the model were aggregated to longer temporal scales to discover the cause of variability on multiple timescales. Factors differ between $\delta^{18}O$ and d and differ according to temporal scale. Rainfall intensity, both at the observation site and upwind, was most important for determining $\delta^{18}O$ and this relationship was robust across all time scales (daily, seasonal, and interannual) as well as generalizing to a second observation site. The sensitivity of $\delta^{18}O$ to rainfall intensity makes annual mean values particularly sensitive to the year's largest events. Projecting the rainfall intensity relationship back through $\sim$ 100 years of precipitation observations can explain $\sim$ 0.2–0.4‰ shifts in rainfall $\delta^{18}O$. Twentieth century speleothem records from the region exhibit signals of a similar magnitude, indicating that rainfall intensity should be taken into account during the interpretation of regional climate archives. For d, humidity during evaporation from the ocean was the most important driver of variability at the daily scale, as well as explaining the seasonal cycle, but source humidity failed to explain the longer-term interannual variability making d records from this region a poor candidate for reconstructing source humidity.

Plain Language Summary

In cave deposits, as with several other natural systems, the abundance of heavy isotopes of water, oxygen-18 and deuterium, can be used to determine past changes in climate. This is because the isotopic composition of these systems is linked to that of rainfall, while the abundance of heavy isotopes in rainfall is driven by climate parameters such as temperature and rainfall characteristics. For this to be effective, the factors which drive rainfall isotopic variability need to be well known. This study uses a 13-year data set of daily rainfall samples from coastal southwestern Western Australia to better understand isotopic variability for this region. Oxygen-18 variations here are driven mainly by rainfall intensity (the amount of rain each day) both according to measurements at the site and upwind simulations. Deuterium excess, a second order parameter which is often linked to conditions in the evaporation source region, was well-predicted by source region humidity at the daily scale but not when aggregated to annual totals. The relationship between rainfall intensity and oxygen-18 appears to be important over the 20th century, based on a comparison between observed rainfall and a cave record.

1 Introduction

In systems where material is sequestered from the environment, for instance speleothem growth or groundwater infiltration, stable isotope ratios act as one of the markers which record information about environmental change. Speleothems, that is cave decorations such as stalagmites and flowstones, record changes in the oxygen isotopic composition as they grow, and these changes can in turn be linked to changes in rainfall isotopic composition (Lachniet, 2009; Orland et al., 2009; Z. Zhang et al., 2018). Karst regions occur throughout the mid-latitudes (Chen et al., 2017) meaning that cave records can be used to infer past changes regional rainfall (Treble, Chappell, et al., 2005; H. Zhang et al., 2018; Lorrey et al., 2008; Fohlmeister et al., 2012; McCabe-Glynn et al., 2013), in areas where this is not achievable using materials such as coral and ice. Speleothem use is widespread, with the SISALv2 database alone containing 691 records of $\delta^{18}O$ in speleothems (Comas-Bru et al., 2020). Meanwhile, speleothem fluid inclusions (Vonhof et al., 2006;

van Breukelen et al., 2008) and groundwater (Priestley et al., 2020) sample infiltrating rainwater from which hydrogen isotope ratios can be derived.

Interpreting these records is not straightforward, even though the link between water isotope ratios and their drivers is well understood at the laboratory scale. In a closed system, heavier isotopes are concentrated in the more condensed phase according to the temperature-dependent equilibrium fraction factor (Horita & Wesolowski, 1994; Majoube, 1971). When diffusive transport is important, the difference in molecular diffusivity between isotopologues (Merlivat, 1978b) leads to kinetic fractionation. In the climate system, however, precisely which climatic and atmospheric processes emerge with the strongest link to isotopic variations is less clear.

The drivers of isotope variability in the climate system are not even consistent between regions. Towards the poles, oxygen isotopes in ice have been used as an indicator of site temperature (Brook & Buizert, 2018); whereas tropical rainfall isotopes have classically been thought of as being controlled by precipitation amount (Dansgaard, 1964). Other important factors include: moisture source (Krklec & Domínguez-Villar, 2014); monsoon activity (Okazaki et al., 2015); the type of precipitation (Aggarwal et al., 2012); the degree of convective organization (Moerman et al., 2013); and trajectories (Deininger et al., 2016). In the mid-latitudes, where rainfall is driven by synoptic-scale weather systems, water isotopes have also been linked to the type of system. This link is reported both in southern Australia (Barras & Simmonds, 2008, 2009; Treble, Budd, et al., 2005), and elsewhere (Lykoudis et al., 2010; Farlin et al., 2013; Tyler et al., 2016; Wang et al., 2017; Schlosser et al., 2017) and arises because of several factors which systematically differ between systems. These factors include: air mass rainfall history, which drives variability because heavy isotopes are preferentially lost from the atmosphere; and the relative contribution of convective versus stratiform precipitation, which fractionate water isotopes differently (Aggarwal et al., 2016; Guan et al., 2013; Webster & Heymsfield, 2003). The location of a synoptic system, relative to the collection site, can also play a role because of isotopic differences between pre- and post-frontal rain (Aemisegger et al., 2015) and, the sensitivity of isotopes to the time which air parcels spend over land (Good et al., 2014).

This study is concerned with southwestern Western Australia (SWWA) in the Southern Hemisphere midlatitudes, where δ^{18}O values in speleothem records (Treble, Chappell, et al., 2005) have low frequency variations that are likely to be linked to climate, but a robust understanding of the mechanism is incomplete. Treble, Chappell, et al. (2005) showed that the stable water isotopes measured in SWWA daily rainfall samples, over a one-year study period, are associated with rainfall intensity, but other drivers may also play a role. It is also unclear whether the intensity dependence holds over longer time periods. An understanding of these drivers is particularly important for this region; winter rainfall here has dropped significantly since the 1970s (Bates et al., 2008) and placing this in the context of the region's long-term natural variability is important for fully understanding the change. This is a challenging task because of the region's strong internal variability, demonstrated in climate models (Cai et al., 2005; England et al., 2006), combined with a short ($\sim$100 yr) instrumental record (Haylock & Nicholls, 2000).

The purpose of this paper, then, is to investigate the factors which influence the abundance of stable water isotopes (^{2}HHO, $H_2^{18}O$) in a modern 13 yr record of SWWA rainfall, taking into account day-to-day variations in synoptic types, upstream conditions, and site parameters. In particular, our goal is to identify factors which are important both at the daily, seasonal, and annual scales. This is most relevant to understanding speleothem records from the region, although we expect the measurements to be more widely useful.

The remainder of this paper is organized as follows: Sect. 2 describes the characteristics of the study region; Sect. 3 introduces the methods used in this study, including a Lagrangian trajectory model and statistical methods; Sect. 4 describes the main results and illustrates links between water isotopes and their drivers; and Sect. 5 compares our results with the literature, tests the ability of our interpretation to generalize to another site, as well as summarizing implications for speleothem record interpretation.

2 Regional setting

Southwestern Western Australia (SWWA, Fig. 1), has an average May–October rainfall of up to 850 mm (Bates et al., 2008, Fig. S1), making it a wet and productive region in comparison to the arid inland. Most rain falls during these cooler months, and the total seasonal rainfall is closely related to the number of fronts which cross the coast, which in turn is coupled to the strength and extent of the Hadley-Walker circulation (Rudeva et al., 2019). Along the coastline south of Perth, about 50% of winter rainfall is associated with fronts, which can be accompanied by thunderstorms (Pepler et al., 2020), 20% with cutoff lows (low pressure systems formed at upper tropospheric levels), and the remainder with warm troughs and other synoptic systems (Pook et al., 2012). Further inland, the proportion of frontal rainfall is lower, and the climate is dryer. Other studies, although differing in how synoptic systems are defined (Hope et al., 2014), have generally classified rainfall-bearing systems into similar synoptic types (Raut et al., 2014; Hope et al., 2006) and agree on the importance of frontal rainfall during the rainy winter season. In summer, when the subtropical ridge lies over the region, monthly rainfall of 20 mm or less is typical and frontal rainfall makes up a smaller proportion of the total. Instead, rainfall comes from a mixture of thunderstorms, extratropical cyclones, (Pepler et al., 2020) and warm troughs (Raut et al., 2014). Also more common during summer are the rare, but potentially extreme, events from ex-tropical cyclones (Foley & Hanstrum, 1994).

As well as having a dramatic seasonal cycle, the region's rainfall has changed on interannual to decadal timescales. Since 1970, the water inflow to Perth's dams has decreased by half (Power et al., 2005), due to the combined effect of reduced winter rainfall and increased evaporation, and the rainfall intensity distribution has shifted so that light rainfall contributes to a larger fraction of the total (Philip & Yu, 2020). A number of studies, reviewed by Dey et al. (2019), show the rainfall decrease, in winter, is associated with a change in regional circulation including a poleward shift in westerly winds. The resulting decrease in the frequency of strong fronts (Raut et al., 2014) has been related to a significant warming of the Southern Hemisphere troposphere south of 30°S followed by a decrease in the strength of the jetstream, which, in turn, decreases the instability and makes the formation of synoptic disturbances less likely (Frederiksen & Frederiksen, 2007). This is in agreement with a recent study by Lucas et al. (2021), who described a reduction in the intensity of the upward midlatitude circulation branch in the Southern Hemisphere at 30°S. Climate model projections indicate that the drying trend will continue (Bates et al., 2008; Raut et al., 2016).

There are several approaches for determining climate variability before the instrumental record. Changes in rainfall have been inferred from distant measurements of snow accumulation (Zheng et al., 2020), which is possible because of an anticorrelation between SWWA May–October rainfall and snowfall at Law Dome, Antarctica (van Ommen & Morgan, 2010). Speleothem records, an alternative in situ climate proxy, are found in caves which develop in Tamala Limestone (Geoscience Australia & Australian Stratigraphy Commission, 2017), an eolian carbonate deposited in the Middle to Late Pleistocene, ∼10–250 ka before present (Smith et al., 2012). Tamala Limestone is extensively distributed along several hundred kilometers of the Western Australian coastline (Fig. 1). Meanwhile, groundwater from the confined aquifers of the Perth Basin (Priestley et al., 2020) has been interpreted as a low-resolution record of infiltration. Both speleothem and ground-

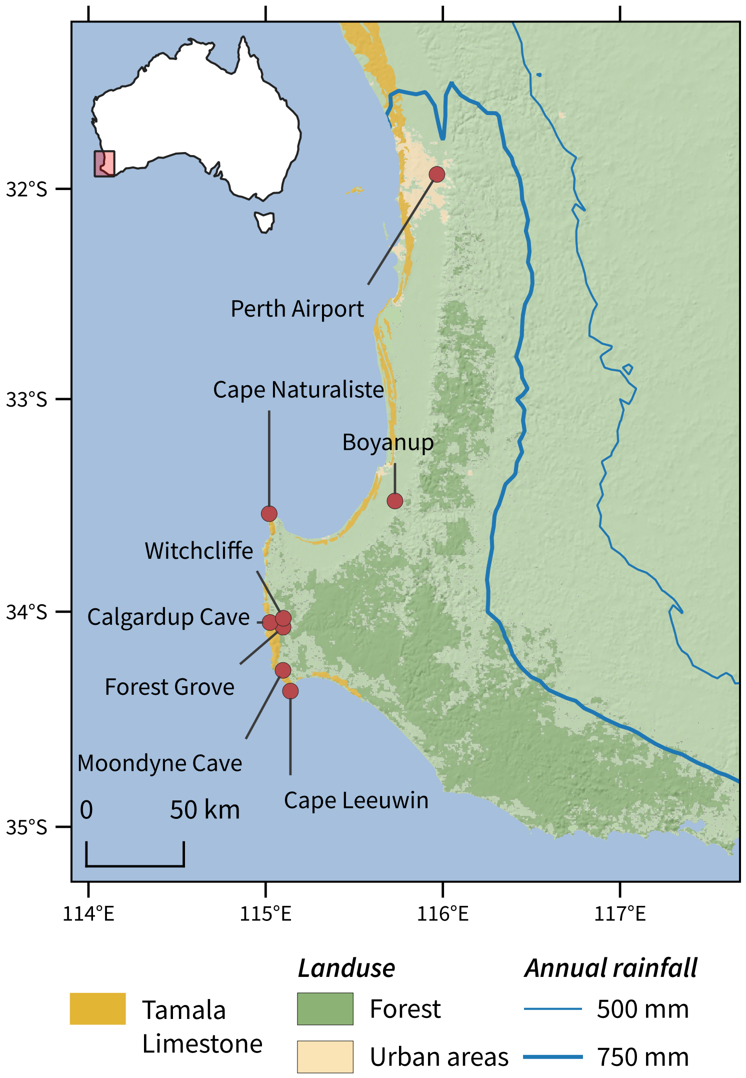

Figure 1. Southwestern Western Australia (SWWA) and locations referenced in the text with: the distribution of Tamala Limestone (a karstic eolianite that occurs along the coast; Geoscience Australia, 2012); land cover (Paget, 2008); and annual mean rainfall (Australian Bureau of Meteorology product IDCJCM004).

water records would benefit from a better understanding of the climate drivers of stable water isotopes.

3 Methods and data

3.1 Rainfall sampling

Rainfall samples were collected from the Calgardup Cave visitors center within a forested nature reserve 23 km from the coast (34.0499°S, 115.0246°E, 70 m ASL, Fig. 1). Samples were collected in a rain gauge consisting of a 203 mm diameter circular funnel draining into a graduated cylinder. The top of the rain gauge was approximately 0.3 m above the ground and within a small clearing; nearby vegetation was kept clear of the gauge. The gauge was checked daily at 0900 local time (0100 UTC) and on days with more than 2 mm of rainfall a sample was collected by filling a 12 ml amber glass bottle completely to the rim. The sample bottle was sealed using a polypropylene lid with Teflon tape placed around the thread to improve the seal. Samples were kept refrigerated until analysis. For this study, measurements were included from the years 2006-18 to avoid including partial years. Occasionally, observers sampled rainfall on days with < 2 mm of rainfall, and these samples were excluded from analysis. In addition, one outlier was excluded. This was recorded on 21 April 2010 with an anomalously high δ^{18}O of -1.2‰ with 52 mm of rainfall, compared to an expected value of about -5‰ for this amount of rainfall. Three rain-days later a sample was anomalously low (-5.0‰ with 4.1 mm of rainfall), so it is possible that samples were mislabeled.

Isotopes are reported in terms of the isotopologue ratios, R, of oxygen–18 ($H_2^{18}O/H_2O$) and deuterium ($^2HHO/H_2O$) relative to Vienna Standard Mean Ocean Water (VSMOW; IAEA, 2006) in rainwater. We use delta notation where $\delta = R/R_{\mathrm{VSMOW}} - 1$, with δ^{18}O and δ^2H representing the two isotopologues. Data up to March 2012 were previously published (Treble et al., 2013). New data reported here were obtained using a Picarro L2120-I cavity ring-down spectroscopy analyzer at ANSTO (reported accuracy of ± 1.0 ‰ for δ^2H and ± 0.15 ‰ for δ^{18}O. All data were calibrated against in-house standards calibrated against VSMOW/VSMOW2 and SLAP/SLAP2.

Because δ^{18}O and δ^2H are strongly correlated, we present δ^{18}O results along with deuterium excess (d), a second-order parameter which characterizes the departure of δ^2H from a linear relationship with δ^{18}O. We follow the most common definition (Dansgaard, 1964) where

$$d = \delta^2\mathrm{H} - 8\,\delta^{18}\mathrm{O}. \tag{1}$$

Defined this way, d is approximately conserved during Rayleigh distillation, provided that the ambient temperature is close to 31°C and that Rayleigh distillation does not proceed too far. Although this is a conventional approach, making our results simple to compare with other studies, it is nevertheless possible for equilibrium processes to change d and other definitions have been proposed, as discussed by Dütsch et al. (2017). At colder temperatures, Rayleigh distillation tends to decrease d, as it proceeds because the equilibrium fraction factors depend on temperature (Horita & Wesolowski, 1994). Since the heavy isotopes are depleted by Rayleigh distillation, the effect is to produce a positive correlation between d and δ^{18}O.

Rainfall isotope data are also presented from the Perth Airport Global Network of Isotopes in Precipitation (GNIP) sampling point, 250 km north of Calgardup Cave, where rainfall is accumulated monthly for isotopic analysis (Hollins et al., 2018). Approximately 7 km further inland from Calgardup Cave, there are two automatic weather stations operated by the Australian Bureau of Meteorology (BoM) at sites 9746 (Witchcliffe) and 9547 (Forest Grove). Rainfall measurements are taken from these sites, as well as the more distant sites: 9503 (Boyanup) and 9519 (Cape Naturaliste).

In this paper, the amount of rainfall collected each day is called the 'rainfall intensity', in contrast to 'rainfall total' which is the accumulated rainfall over a longer period. Where averages of δ^{18}O and d are computed, these are weighted by rainfall amount unless noted otherwise.

3.2 Source region diagnostic

Several upstream parameters, chosen because of their potential to affect δ^{18}O and d, were diagnosed using two Lagrangian dispersion models. Models were used to compute a backwards plume, or retroplume, from Calgardup Cave on each day with > 2 mm of rainfall. Backwards plumes are a more realistic generalization of backwards trajectories, with advantages discussed by (Stohl et al., 2002). Lagrangian diagnostics have been widely and successfully used in studies of water isotopes (Pfahl & Wernli, 2008, 2009; Sodemann et al., 2008, e.g.) including the use of backwards dispersion models (Good et al., 2014). Quantities related to the evaporation source region were diagnosed from the source-receptor matrix (Seibert & Frank, 2004) weighted by the instantaneous evaporation rate.

In this study, two sets of backwards plumes were generated. The primary set used FLEXPART version 9.0 (Stohl et al., 2002) with subgrid convective mixing (Forster et al., 2007) and wind fields from the ERA-Interim reanalysis (Dee et al., 2011). A second set of backwards plumes was generated using FLEXPART-WRF version 3.1 (Brioude et al., 2013), forced with a regional atmospheric simulation generated by the Weather Research and Forecasting model version 3.5.1 (WRF Skamarock & Klemp, 2008). The WRF model was forced by the CFSR reanalysis (Saha et al., 2010), and configured with an outer domain which was large enough to contain the backwards plume for approximately 120 h. The second set of plumes was used to verify that the main findings could be replicated and are not discussed further.

Three of the uncertainties in the approach are that: the time of rainfall is only known to within a 24 h sampling window; the appropriate height for beginning the backwards plume has to be estimated; and the error in the plume grows as the model is integrated further back in time. After some experimentation, the beginning time was taken from the time in the WRF simulation with the largest rain rate, and the starting height was taken to be the cloud base in WRF, estimated at the height when relative humidity reaches 80%. Then, to verify that the model indeed produces a useful diagnostic, we checked the correlation between d and humidity relative to saturation at the sea surface temperature, h_s, as a function of back trajectory length. This is a useful diagnostic because d, in vapor, and h_s, at the evaporation site, are strongly correlated (Pfahl & Wernli, 2008) and we assume that d will be approximately conserved during the conversion of water vapor into clouds and then rainfall.

The correlation between d and h_s grows as the backwards plume increases in duration up to about 48 h, but with no further improvement beyond this point (Fig. S2). This indicates that both dispersion models have some skill at determining the evaporation conditions at the moisture source, at least up to 48 h before rainfall.

3.3 Synoptic classifications

On each day, the synoptic type was classified with a Self Organizing Map (SOM), using SOM-PAK (Kohonen et al., 1996), following the approach described by Hope et al. (2006). Synoptic types were derived from the 1200 UTC mean sea level pressure (MSLP) anomaly fields of the ERA-Interim reanalysis on a 0.75° latitude/longitude grid in the region 90–130°E, 50–15°S. The SOM is an unsupervised classification method, producing synoptic types that are arranged in a two-dimensional grid. The arrangement of types into a grid, where similar synoptic types are arranged close to each other, is the main way in which the SOM differs from other statistical classification techniques (Philipp et al., 2016). Synoptic classification was only applied to the rainy months (April–October), due to the presence of seasonally persistent features in the surface pressure field associated with the meridional movement of the subtropical high pressure ridge. Training was performed using data from the years 1979–2018 and grid cells were weighted by area.

In addition to the SOM classifications, fronts were detected in the reanalysis fields and used as an aid to interpret the SOM classifications. The position of fronts was found using the wind shift method (Simmonds et al., 2011) based on ERA-Interim 3-hourly 10 m wind fields. This is a straightforward method which is applicable to SWWA (Hope et al., 2014). It does not produce spurious fronts along the coastline, that are often found by a more commonly used methods based on the temperature gradients (Pepler et al., 2020). The wind-based method works well to define meridionally elongated fronts, that are mainly cold fronts, and is particularly well suited for the Southern Hemisphere (Schemm et al., 2015).

3.4 Generalized additive models

To combine information from site measurements, backwards plume, and synoptic type we used Generalized Additive Models (GAMs; Wood, 2017). Separate models were constructed to predict δ^{18}O and d in daily rainfall samples. GAMs, a generalization of linear regression models, allow the relationships between predictor variables and the response variable to be modelled as smooth curves rather than straight lines. In contrast to many nonlinear machine learning techniques, a benefit of using GAMs is that the relationship between predictor and response variables is simple to visualize, making the models readily interpretable.

The GAM implementation was provided by mgcv, a package for R (R Core Team, 2014). Relationships between predictor and response variables are modelled with penalized regression splines in which the smoothness is estimated during the fitting process using restricted maximum likelihood (REML; Wood, 2011), and models used the identity link function. In this implementation, predictors which can be modelled with a linear response are modelled that way, and predictors with insufficient explanatory power are dropped from the model. The mgcv models can also incorporate categorical variables, allowing the synoptic classification to be included within the same framework.

In this study, we also assessed the importance of terms for explaining the observations on different timescales. As well as allowing the models to drop unimportant terms (using REML) we followed a procedure where models were constructed term-by-term. Beginning with an empty model, each candidate term was tested, and the term resulting in the best performing model retained. The search for the best term was then repeated by adding a second term to the model, and so on.

The metric for assessing model performance was the 13-fold cross-validated mean-square error (MSE) applied to daily predictions of δ^{18}O or d. To score a model, one year is held out, and the other years are used to train the model, then the MSE computed on the held-out year, defined as

$$\mathrm{MSE} = \frac{1}{N}\sum_{i=1}^{N}\left(\hat{y}_i - y_i\right)^2 \tag{2}$$

where N is the number of observations, y_i is the ith day's observation and $\hat{y}_i$ is the ith day's model prediction. This is repeated for all years in the data set, and the MSE is taken as the average from all the hold-out sets. During model building, terms are added in the order of the greatest reduction in daily cross-validated MSE.

Once the set of models has been obtained, the cross-validated MSE is then recorded for three groupings: 1. the original, daily, data 2. the mean seasonal cycle during the rainy months (April–October); and 3. the annual precipitation-weighted means.

3.5 Modelled precipitation isotopes

In addition to the diagnostic and statistical models described above, we also use output from a prognostic model: a 40 year simulation of IsoGSM (Yoshimura et al., 2008). This is one of several atmosphere general circulation models with water isotope tracers (Risi et al., 2010; Sturm et al., 2005; Schmidt et al., 2007; Lee et al., 2007, e.g.). IsoGSM is forced with the NCEP/DOE Reanalysis and output from the model is available with a horizontal resolution of 2.5°.

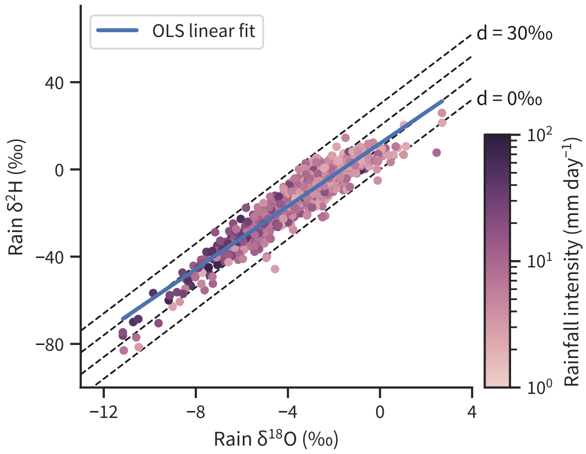

Figure 2. Rainfall δ^{18}O and δ^{2}H measured in daily samples from Calgardup Cave visitors centre coloured by the daily rainfall intensity. For comparison, the global average d in precipitation is about 10‰.

At other sites, IsoGSM reproduces daily, monthly, and seasonal variability in water isotope ratios, with more skill at simulating δ^{18}O than d (Yoshimura et al., 2008). At the daily scale, the low accuracy of the model-produced precipitation (that is, the model may not necessarily produce rain on a rainy day) limits the accuracy of predicted water isotopes.

4 Results

Our results include a description of the stable water isotopes in Sect. 4.1–4.3 before moving onto the more interpretive results from statistical and dispersion models in the later sections.

4.1 Daily δ^{18}O, δ^{2}H, and precipitation

Over the 13 year monitoring period (2006-18 inclusive, days with $\geq$2 mm day^{-1} of rainfall) the precipitation weighted mean δ^{18}O was -4.45‰, d was 15.4‰, and $\delta^2 H$ was -20.2‰. More than 2 mm of rain fell on an average of 90 days each year, and the mean annual precipitation from these events was 839 mm. The daily isotope samples, when plotted in $\delta^{18}\mathrm{O} \sim \delta^{2}\mathrm{H}$ space, are strongly correlated and lie about the so-called local meteoric water line (LMWL; Fig. 2), with fit parameters given in Tab. S1.

There is a tendency for intense rainfall to have lower δ^{2}H and δ^{18}O and for low intensity rainfall to both have high δ^{18}O and, above -2‰, depart from the straight line trend. Deuterium excess for these high δ^{18}O samples tends towards the $d = 0$ ‰ line, contrasting to the overall mean d. For comparison, the global meteoric water line (GMWL) of Craig (1961) lies on the $d = 10$‰ line.

As a result of this tendency for low δ^{18}O samples to have lower d, the slope of the LMWL is lower than the GMWL. The parameters for straight-line fits to the daily rainfall samples are shown in Tab. S1, with both ordinary least-squares (OLS) and precipitation weighted least squares (WLS, Hughes & Crawford, 2012). Taking uncertainty into account, the slope of the LMWL at Calgardup Cave is indistinguishable from the Perth

Airport LMWL, but the there is an offset between the two sites since the intercept differs by about two standard deviations.

As noted in Sect. 3.1, the temperature-dependence of equilibrium fractionation would lead to an increase in d with δ^{18}O. Here we see the opposite trend, which is indicative of non-equilibrium processes becoming relatively more important during light rainfall.

4.2 Seasonal cycle

The composite seasonal cycle of δ^{18}O, d, and P has been published previously for Perth (Hollins et al., 2018; Liu et al., 2010) and the seasonal cycle at Calgardup is broadly similar (shown later in Fig. 8, but also Fig. S1). The similarity is consistent with isotopes at the two sites being driven by similar factors.

The δ^{18}O minimum occurs in May or June, earlier than the July peak in rainfall. January stands out as an exception with anomalously low–and variable–rainfall δ^{18}O when compared with the surrounding months, likely because of the occurrence of rare, but intense, rainfall events. Deuterium excess, d, also has a strong seasonal cycle which mirrors δ^{18}O, peaking in the rainy months. Unlike δ^{18}O, summer variability is not especially pronounced.

4.3 Annual mean time series

Rainfall δ^{18}O, aggregated to annual precipitation-weighted averages, follows an overall decreasing trend, which is present at both Calgardup Cave and Perth as well as in IsoGSM model output (Fig. 3). From 2009 onwards, however, there is no statistically significant trend. Comparison with longer term model output, and earlier data from Perth, (Hollins et al., 2018) indicates that 2006-08 were anomalously high, compared to the long-term average. The annual-mean d (Fig. 3b) shows similar trends at Perth and Calgardup Cave, but the IsoGSM simulations are unable to reproduce the observed trends. There is no consistent trend in d if the first three years are excluded.

On average, annual δ^{18}O values are 0.61‰ higher at Perth implying a meridional gradient in δ^{18}O of 0.29 ‰ per degree of latitude. This agrees with a persistent feature of isotope enabled GCMs which simulate a δ^{18}O maximum over the Indian Ocean north of Perth, near 30°S and under the descending branch of the Hadley Cell, with decreasing values towards the pole (Werner et al., 2011; Lee et al., 2007; Noone & Simmonds, 2002; Risi et al., 2012). The offset between mean values for Perth and Calgardup Cave shows no trend through time, implying that the meridional gradient has remained consistent over the monitoring period.

Annual mean departures from the trend are not consistent between sites (Fig. 3a), suggesting that δ^{18}O anomalies are related to local processes. At least in part, the low correlation between sites is because annual mean δ^{18}O is particularly sensitive to the heaviest events of the year, shown by plotting four similar time series in which the heaviest 1–4 rainfall events from each year are excluded. Excluding the heavy events shifts the mean δ^{18}O higher and, in years like 2015 and 2018, can change annual means from anomalously low to high. As with any rainfall event, these events will be sampled differently by the two monitoring sites (Good et al., 2014), so stochastic variability is a major contributor to the annual precipitation-weighted mean δ^{18}O. In contrast to δ^{18}O, the interannual variability in d is not as strongly affected by these intense rainfall events (Fig. 3b), so the annual-mean difference between Perth and Calgardup Cave time-series are not as sensitive to stochastic variability.

To examine the factors which drive these long-term changes, and the seasonal cycle, we analyze the conditions on each rainy day in the following sections.

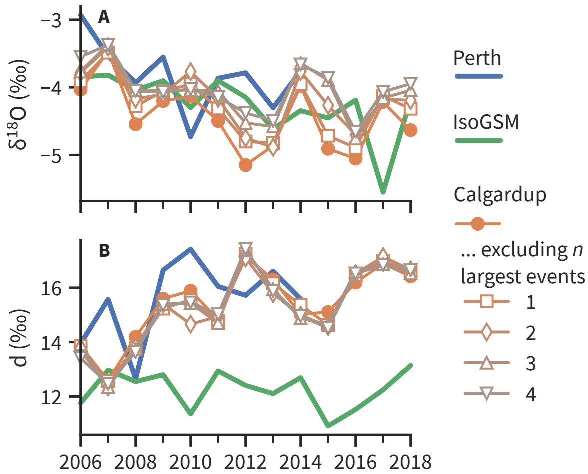

Figure 3. Annual precipitation-weighted mean δ^{18}O and d from Calgardup Cave and Perth. As well as showing the entire dataset, the annual mean values for Calgardup are also computed after incrementally leaving out the four largest daily rainfall accumulations, illustrating the sensitivity of interannual δ^{18}O variations to a few events. Results from the IsoGSM isotope-enabled general circulation model are also shown.

4.4 Synoptic systems

Self organizing maps (SOMs) were used to classify synoptic regimes. We identified 35 synoptic types using MSLP fields from ERA-Interim, and each day was associated with one of types shown in Fig. 4. Supplementary interpretation is provided by the frontal density and 500 hPa height fields in Fig. S3, and Fig 5 summarizes several observations according to synoptic type.

Although the SOM is not derived directly from frontal information, the location of fronts is related to the surface pressure field and the synoptic types are therefore associated with front positions. The top two rows in the SOM are most strongly associated with the presence of rain-bearing cold fronts directly over SWWA, while the sequence around the outside edge of the SOM, A4$\cdots$A1$\cdots$E1, tracks the progress of cold fronts beginning offshore to the west and moving east across the region. This is a common occurrence, and appears as a path with high transition probabilities in Fig. 5A. Types in the top left are more pre-frontal rainfall, while types in the top right are post-frontal.

Synoptic types away from the top rows are not as strongly associated with frontal rainfall (Fig. S3); although fronts are detected they are generally away from SWWA. Notably, the pressure pattern for classes A5, A6 resembles a trough, associated with moisture transport from the northwest, and A7 is a blend between a trough and cutoff low. These three classes, in general, are related to upper-tropospheric processes with fronts being detected too far to the west to be responsible for rainfall.

As shown in Fig. 5 synoptic types are a reasonable predictor of rainfall properties, several of which show a strong dependence on SOM classification. In particular, the wettest class (A1) has a rainfall probability of 66%, much higher than the driest class with 5% probability of rain (Fig. 5d). Rainfall intensity (Fig. 5c) is also sensitive to synoptic type, with column A showing the most intense rainfall, especially for classes A5 and A6. Although these non-frontal classes are associated with heavy rain, and A6 accounts for the highest total precipitation, frontal events are responsible for more rainfall overall as they

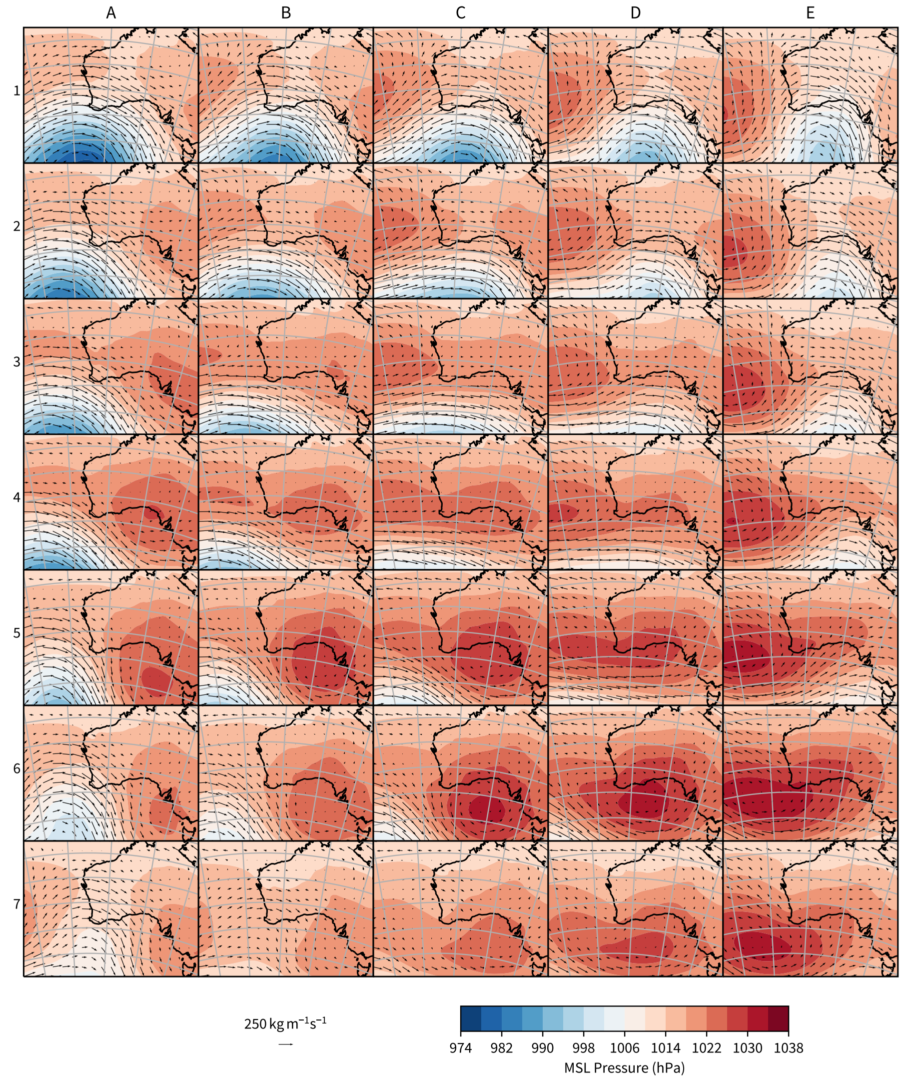

Figure 4. SOM–derived synoptic types, with mean-sea-level pressure and vertically-integrated water vapor transport.

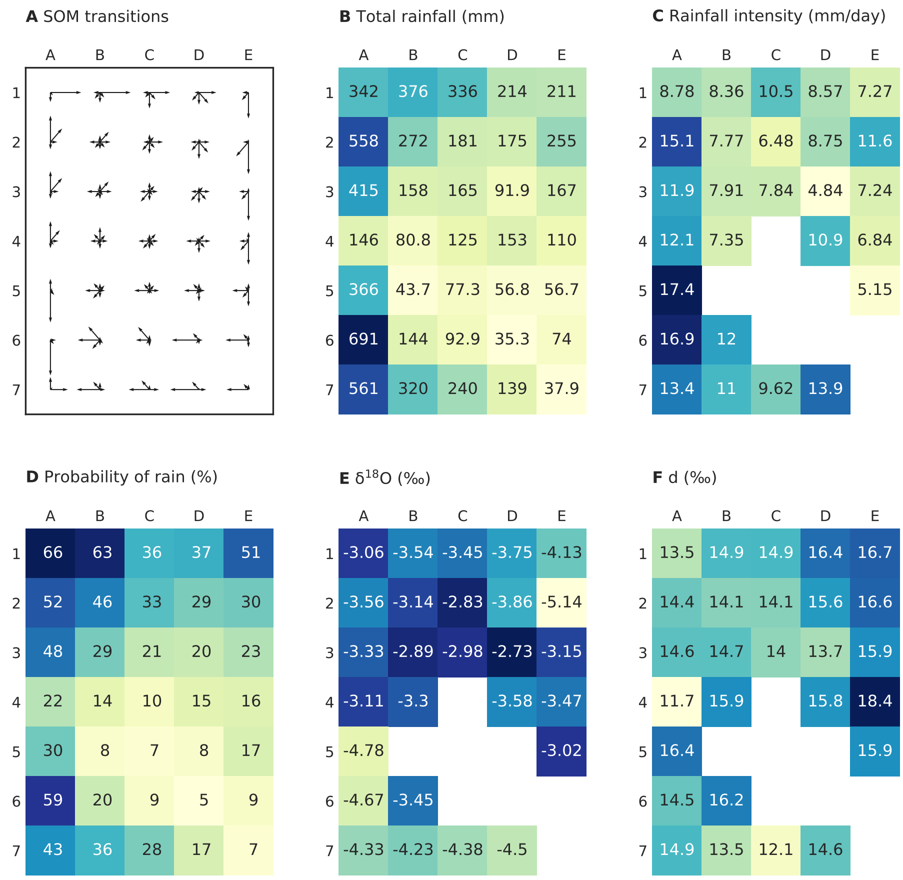

Figure 5. Rainfall, isotope, and SOM properties, 2006-18, by synoptic type. Panels show: **A** relative transition probability (longer arrows show more likely transitions); **B** accumulated precipitation; **C** rainfall intensity (mean rainfall per day); **D** probability of rainfall; **E** arithmetic mean δ^{18}O; **F** arithmetic mean deuterium excess, *d*. Colors are used to highlight patterns in the data, the number of days in each class ranges from 51 to 101, and cells with less than 10 observations are left blank in panels C, G, and H.

occupy a larger number of classes. Based on manual classifications, Pook et al. (2012) also found that fronts were responsible for most winter rainfall.

Water isotopes show a weaker dependence on synoptic type than precipitation itself, but a relationship nevertheless exists (Fig. 5e and 5f). For δ^{18}O, frontal rainfall shows a trend towards lower δ^{18}O and higher d after the passage of the front, seen in the top row of these figures. Another pattern revealed by the SOM is that non-frontal rainfall is lower in δ^{18}O. Trends in d (Fig. 5f) are in the opposite direction, with the non-frontal class A5 having higher d than the frontal rainfall classes A1-A3.

These observations are consistent with other studies (Treble, Budd, et al., 2005; Barras & Simmonds, 2008) which have demonstrated, in the Australian region, that different types of synoptic systems can have distinct isotopic signatures, an effect which is replicated at sites elsewhere in the world (Baldini et al., 2010; Scholl et al., 2009). In particular, the anomalously low rainfall δ^{18}O observed from intense low pressure systems lying off the eastern coast of Australia (Crawford et al., 2017) is a similar finding to the low δ^{18}O and intense rainfall seen in classes A6 and A7.

The SOM analysis, while showing an association between synoptic types and isotopes, does not by itself identify the reasons behind the association. Furthermore, although there is a relatively large difference between frontal and non-frontal rainfall, δ^{18}O differing by 1–1.7‰, this difference is not large enough to explain the year-by-year variability (Fig. 3). Year-by-year changes can reach 1‰, meaning that rainfall would need to switch from almost exclusively frontal rainfall to non-frontal to explain the changes in annual mean δ^{18}O, and this is not something which is observed. In the next section, upstream conditions, diagnosed from dispersion modelling, are combined with site-based observations and synoptic types to gain more insight into the underlying processes.

4.5 Generalized additive model for δ^{18}O

Generalized additive models (GAMs) trained to predict daily rainfall δ^{18}O are shown in Fig. 6. These curves are the model's 'smooth terms', that is the smooth functions expressing the relationship between predictor variables and the response variable. Two models are shown, one with synoptic types (trained on data from the wet months, Apr–Oct) and another without synoptic types (trained on data from the entire year). In this figure, smooth terms are ordered by their explained deviance (Wood, 2006); a measure of importance for predicting daily δ^{18}O. Metrics for judging the importance of terms are shown in Fig. 7.

For predictions of daily δ^{18}O, the most important smooth terms in this model are: the locally-recorded rainfall intensity, P; the mean rainfall intensity along the retroplume, $\overline{P}$; and then source humidity, h_s relative to the sea surface temperature. Local rainfall intensity is the best predictor of daily δ^{18}O, the seasonal cycle, and year-to-year variability (Fig. 7), it follows a relationship which is close to $\delta^{18}\mathrm{O} \propto \log(P)$. Adding the retroplume rainfall intensity improves the model's fit to interannual variability, by almost as much as P, but does not affect its fit to the seasonal cycle. The third term, h_s is defined as the humidity in the evaporation region, within the lowest model level, relative to the sea surface temperature, and weighted by evaporation rate. Source humidity is important for the δ^{18}O seasonal cycle, but not interannual variability. Indeed, the inclusion of h_s increases the model error for the prediction of interannual variability.

Remaining terms do not make a major difference to the model's predictive ability at interannual scales (Fig. 7). Nevertheless, the starting latitude and longitude of the plume, along with the source temperature and retroplume overland fraction, are detected in the model as having an influence on δ^{18}O, and are discussed further in Sect. 5.

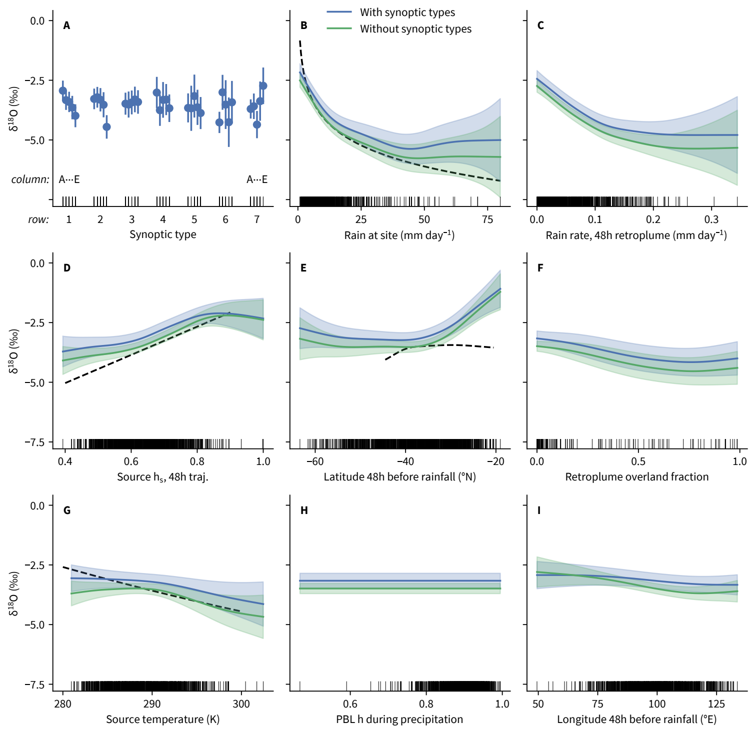

Figure 6. Categorical and smooth terms for a GAM predicting daily δ^{18}O. The categorical term is shown first, then smooth terms are ordered from the highest to lowest explained deviance. Error bars or shading indicates the 95% confidence interval. Upward ticks on the x-axis of each plot indicate measurements and black dashed lines show other relationships: **B** $\log P$; **D** kinetic fractionation (Merlivat & Jouzel, 1979; Benetti et al., 2014); **E** δ^{18}O latitudinal variation in Indian Ocean surface waters (LeGrande & Schmidt, 2006); **G** equilibrium fractionation factor dependence on temperature (Horita & Wesolowski, 1994).

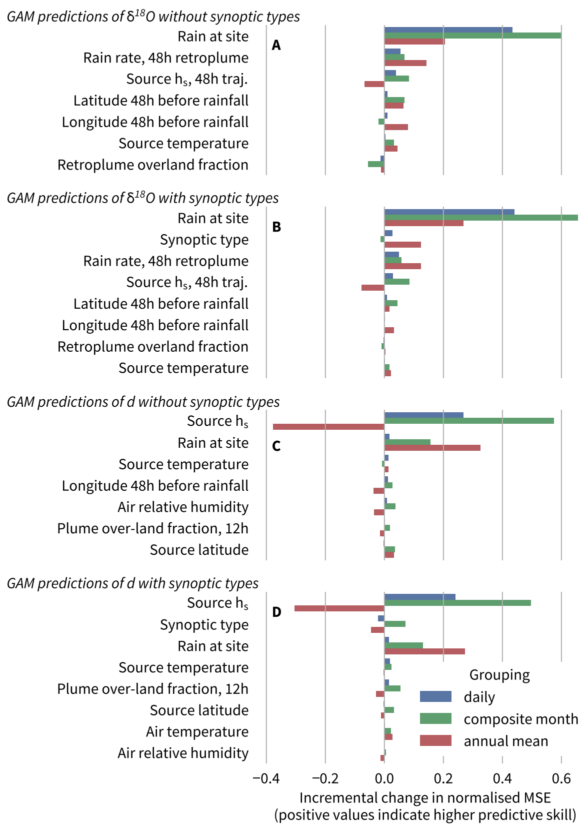

Figure 7. Prediction accuracy of GAM: **A** δ^{18}O predictions; **B** like A, but the additional 'synoptic type' predictor; **C** d predictions; **D** like C, but with synoptic types. Plots show the improvement in the cross-validated mean squared error (MSE) compared with a simpler model without that term, starting from a model which predicts the mean. MSE is normalized the by MSE of the 'constant value' model. In each category, three precipitation-weighted groupings are considered: 1. the ungrouped daily data; 2. monthly groups for a composite year; and 3. annual totals.

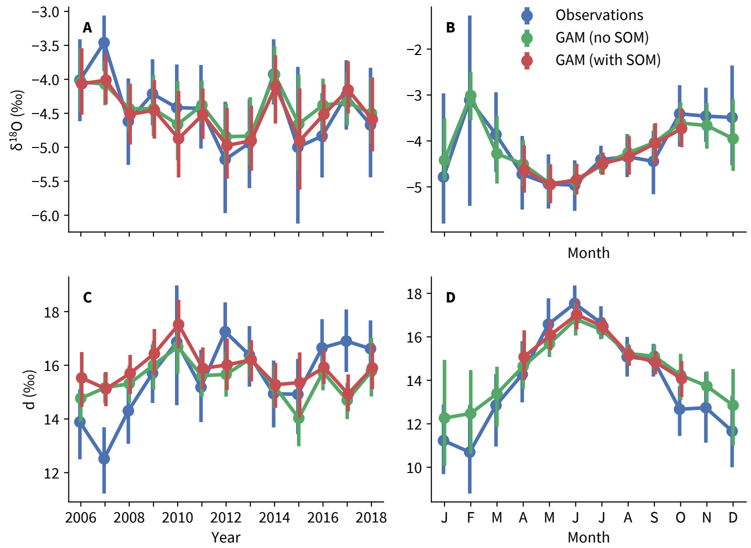

Figure 8. Precipitation-weighted GAM predictions versus observations: **A** annual δ^{18}O; **B** seasonal δ^{18}O; **C** annual d; and **D** seasonal d. Error bars show the 95% confidence interval from bootstrapping daily values.

Despite being statistically-significant, including synoptic types as a predictor variable does not appreciably improve the overall model performance (Fig. 7), suggesting that synoptic types contain redundant information already contained in the smooth terms. The shape of the smooth terms is also insensitive to the presence of synoptic types, as seen in Fig. 6 where the GAM with synoptic types has similar smooth terms to the GAM without. There are also similarities in the patterns of Fig. 6a, which show the effect of synoptic type marginalized for the effect of other variables, to the patterns in Fig. 5e which showed the mean δ^{18}O in each synoptic type.

A comparison of GAM predicted oxygen isotopes with observed timeseries is shown in Fig. 8a and 8b showing that the GAM successfully tracks δ^{18}O interannual variability and the seasonal cycle.

In summary, the combination of the GAM analysis with synoptic types supports the conclusions of earlier studies which have found that isotopic composition is related to synoptic types, but it also shows that there are underlying continuous variables which explain the isotopic composition, for this region, without needing to incorporate synoptic types. The continuous predictor variables have the advantages that they can be used in all months of the year and are less likely to cause over-fitting.

4.6 Generalized additive model for deuterium excess

Deuterium excess, d, differs from δ^{18}O both in terms of which predictors are important, and how well a GAM trained on daily data is able to predict interannual variability. As with δ^{18}O, our approach was to train a GAM using daily data and then use this model to make predictions aggregated over longer periods. Next, another GAM was trained with the additional predictor of synoptic types.

The leading predictor of daily d is source humidity, h_s relative to saturation at the sea surface. It is followed by rainfall intensity, P. Compared with h_s, P only weakly improves the MSE at the daily scale (Fig. 7c) but it is the term which improves the annual-

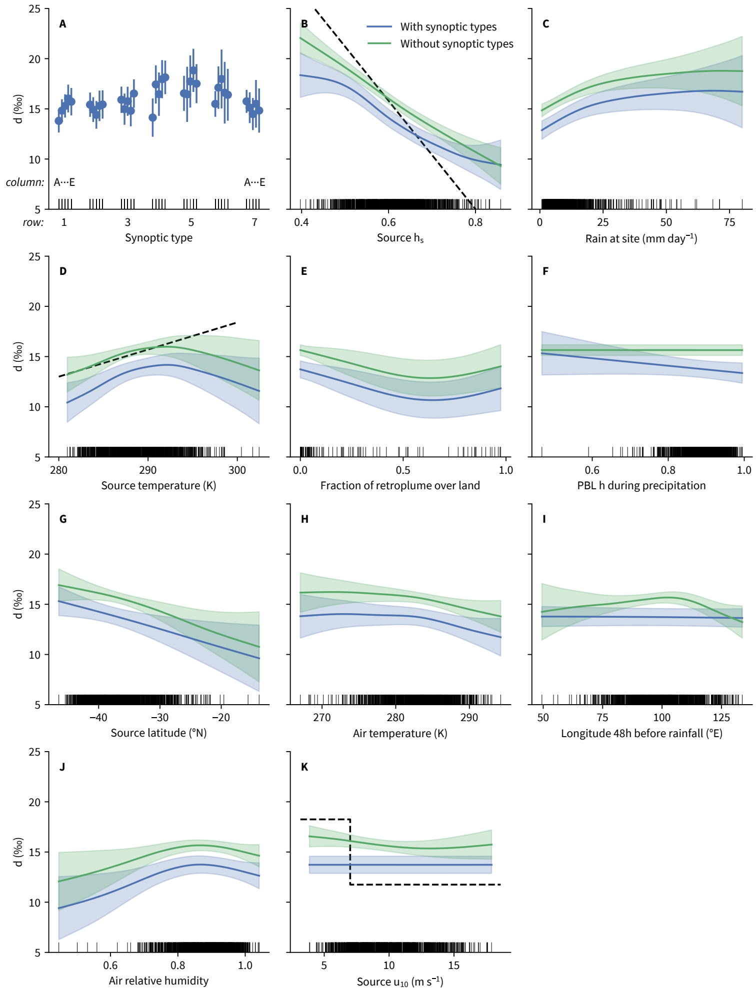

Figure 9. Smooth terms for GAM predicting daily d. Other relationships shown are: **B** empirical relationship from Pfahl and Sodemann (2014); **D** empirical relationships from Bonne et al. (2019); **K** parameterisation from Merlivat and Jouzel (1979) at $h_s = 0.6$ (Benetti et al., 2014).

mean predictions the most as well as contributing to a better prediction of the seasonal cycle.

The effect of adding synoptic types to the d model, which also means restricting the model to rainy months, is shown in Fig. 7d. Synoptic type, although statistically important according to the REML test, fails to improve the cross-validated MSE at the daily or interannual time scales. As with δ^{18}O, the information introduced to the model by the synoptic types is redundant, and harms the cross-validated performance of the model, possibly because the large number of categories promotes over-fitting.

Of all the factors in this analysis, however, it is the source humidity which stands out. It is strongly linked to d at the daily scale, it is apparently the main driver of the observed seasonal cycle, but it produces a very poor model of interannual variability—one which has a larger error than a model without h_s. When plotted alongside observations, the annual mean predictions of d (Fig. 8c) show that, in contrast to the case of δ^{18}O, the GAM is unable to follow the overall increasing trend in observed rainfall d, even though it is largely successful at reproducing the seasonal cycle. The GAM predictions start above the observations and then are biased low by the end of the observation period (model residuals are shown more clearly in Fig. S4). An explanation for this apparent contradiction is that there is a missing term which is correlated with both h_s and δ^{18}O at the annual-mean timescale.

5 Discussion

5.1 Physical processes driving δ^{18}O

The process with the strongest link to rainfall δ^{18}O was the rainfall intensity, both that measured at the site (P) and along the retroplume ($\overline{P}$). Although P and $\overline{P}$ are closely related conceptually, they are only moderately correlated ($R = 0.33$) and it is possible that both are imperfect proxies of the same underlying factor driving isotopic fractionation, such as the proportion of water remaining in the system (Lee & Fung, 2008) in analogy with Rayleigh distillation, or the relative importance of moisture convergence versus local evaporation, which has been described in steady-state over an idealized tropical ocean (Moore et al., 2014).

Rainfall intensity, that is rainfall measured each day, has a stronger association with δ^{18}O than does total monthly precipitation, in agreement with Fischer and Treble (2008) who studied monthly δ^{18}O data from Perth and a short record of daily measurements from Cape Leeuwin. Also similar is that Fischer and Treble (2008) found a nonlinear relationship between precipitation and δ^{18}O, using $\delta^{18}\mathrm{O} \propto P^{\frac{1}{2}}$. In our data set, due to scatter, $\delta^{18}\mathrm{O} \propto P^{\frac{1}{2}}$ fits the data almost as well as $\delta^{18}\mathrm{O} \propto \log(P)$, and we plot the log form mainly out of preference because of its appearance in Rayleigh distillation (Eriksson, 1965) and also the use of a log transformation when δ^{18}O is regressed against moisture residence time, τ. Aggarwal et al. (2012) found that $\delta^{18}\mathrm{O} \propto \tau = \log(Q/P)$ where Q is the total column water vapor and P is the long-term mean precipitation rate. In our data, variability in Q is small enough that $\log(P/P_0)$ is strongly correlated with τ ($R = -0.94$, Q from ERA-Interim) and there is no advantage in using τ.

Source humidity, h_s affects δ^{18}O through kinetic fractionation. The relationship determined by the GAM is similar to the expression for kinetic fraction used by Benetti et al. (2014), as shown by the dashed line in Fig. 6d. In contrast, the relationship between latitude and δ^{18}O (Fig. 6e) does not follow the meridional variation in Indian Ocean surface water δ^{18}O (LeGrande & Schmidt, 2006). Fischer and Treble (2008) also reported a difference in δ^{18}O between airmasses travelling equatorward or poleward, but our results suggest that isotopic differences in the source waters are not responsible. Instead, there are multiple co-varying parameters which may obfuscate the direct effect of source water δ^{18}O; latitude is strongly correlated with the oceanic source temperature ($R =$

0.91), wind speed ($R = -0.61$), and humidity ($R = 0.48$). In addition, there is a strong and persistent meridional gradient in mean atmospheric δ^{18}O, with higher values towards the pole, and this is a large driver of isotopic variability in idealized simulations (Dütsch et al., 2016).

The so-called continental effect (Winnick et al., 2014), often appears as an important term driving δ^{18}O (Good et al., 2014, e.g.). Here, the fraction of the retroplume over land, f_l (calculated after 3 h of travel), has the expected sign making our results consistent with isotopic depletion driven by rainout. But, on the whole, f_l is of only minor importance because the vast majority of trajectories do not pass over land before arriving at the rainfall site. Because of the lack of overland trajectories in the data, it is unlikely that the GAM has been able to learn an accurate relationship.

Also present in the GAM is the evaporation-weighted sea surface temperature, T_s. As indicated by the dashed line in Fig. 6G, this term is consistent with the temperature dependence of equilibrium fractionation of water vapor from the ocean surface (Majoube, 1971; Horita & Wesolowski, 1994), though the presence of latitude in the model (strongly correlated with T_s) means that this relationship may be distorted.

Notably absent from the GAM is the planetary boundary layer (PBL) humidity on the day of rainfall collection (Fig. 6H). This parameter has the potential to affect the degree of rain droplet re-evaporation, and therefore rainfall isotopes, and was the leading parameter affecting δ^{18}O at an arid inland site in Eastern Australia (Crawford et al., 2017). Both sites are at a similar latitude, so Calgardup Cave's location on the coast is likely to be the reason for the absence of a link between PBL humidity and δ^{18}O.

5.2 Physical processes driving deuterium excess

The strongest predictor of daily d is the source humidity, h_s, although the relationship between d and h_s shows a lower slope (-30 ‰) than seen in studies of water vapor; the dashed line in Fig. 9b shows a typical slope of -54 ‰ (Pfahl & Sodemann, 2014). There are three potential explanations for this. First, this difference may be due to uncertainty in the h_s estimate. The standard deviation of the difference between FLEXPART and FLEXPART-WRF derived values, accounting for part of the uncertainty, is 0.04 which is large enough, based on tests with synthetic data, to reduce the slope of the line of best fit. Second, low humidity air during rainfall (small h) causes strong re-evaporation of rainfall (Risi et al., 2008). At this coastal site, h is correlated with h_s ($R = 0.33$), so the two effects together act to reduce the observed slope between h_s and d. Third, the slope between d and h_s may be a genuine trait of the source region. Steen-Larsen et al. (2014) report a flatter slope for the $d \sim h_s$ relationship, with a slope of -42.6 ‰, and Aemisegger and Sjolte (2018) demonstrate the $d \sim h_s$ slope varies by region. Even accounting for regional variation however, -30 ‰ is sufficiently outside the range of other observations that a combination of the other factors too, h_s uncertainty or $h \sim h_s$ correlation, is likely to be important.

The effect of sea surface temperature, T_s, and wind speed, u_{10}, have been investigated in the past and their importance is still debated. Uemura et al. (2008) reported a positive correlation between d and T_s in field measurements, in agreement with Bonne et al. (2019), whereas Pfahl and Sodemann (2014) argue that the T_s is of minor importance compared with h_s. Figure 9d shows that our data do indicate a positive correlation between d and T_s for $T_s < 20$°C. The reversal at higher temperatures is physically implausible and likely to be an artifact caused by the sparse data at higher temperatures and correlation between T_s and latitude.

The relationship between u_{10} and d is weak in the GAM, and arguably inconsistent with the widely-applied results of Merlivat (1978a), in which kinetic fractionation, and hence d in evaporation, is lower at high wind speeds. In their parametrization, low wind speeds below about $7\,\mathrm{ms}^{-1}$ correspond to a smooth regime (and higher d) whereas high wind speeds are modelled by a rough regime (with lower d) (Merlivat & Jouzel, 1979). The u_{10} relationship here is too weak to match the parametrization, it disappears from the model when synoptic types are included, and there is little evidence from other studies of the Merlivat (1978a) parametrization being directly applicable in the field. Benetti et al. (2014) present data which lies between the rough and smooth, Steen-Larsen et al. (2014) find no statistical difference in d in low versus high winds, and Bonne et al. (2019) also find there to be no effect on d from wind speed, with their data being best explained by the rough regime of the Merlivat and Jouzel (1979) model. Considered in the context of these other studies, then, the existence of a strong relationship between d and u_{10} seems unlikely.

5.3 Generalizability of the model

As a test of the model's performance away from the observation site, predictions of δ^{18}O for Perth Airport were computed based on observed daily rainfall and FLEXPART backwards plumes terminating at Perth Airport on each rain day. GAM predictions were clipped to the range of observations to prevent extrapolation errors. In particular, on days with less than 2 mm of rainfall δ^{18}O was set to the same value as if 2 mm of rainfall was observed. This was necessary because many of the monthly accumulations included a nontrivial contribution from days with light rainfall.

When compared with monthly δ^{18}O observations, the GAM performed well during the wet months but had large errors during the dry months (Fig. S5). On some occasions, this was because of highly depleted rainfall sourced from the ocean off the northwest coast of Western Australia which had made a long transit over land. In general, the failure of the model to perform well during the summer months can be attributed to lack of summer rainfall in the training data. The stronger influence of tropical processes in summer, on Perth rainfall, may also play a role.

When aggregated to annual data, the poor performance during the dry months becomes inconsequential and the model generalizes well; performance in Perth shows a similar predictive skill to Calgardup Cave (Fig. S6). Furthermore, the GAM is able to reproduce the offset in mean δ^{18}O observed between Perth and Calgardup Cave. To reproduce the offset, the model needs to include rainfall intensity, rainfall along the retroplume, source humidity, and source latitude. This suggests that a combination of these variables is responsible for the observed offset, but it also turned out that rainfall intensity, being responsible for about 10% of the offset, is not the main driver of the offset. The good performance of the model for the Perth observations makes it more promising for the interpretation of longer-term data from the coastal zone between Calgardup and Perth.

For deuterium excess, GAM predictions at Perth Airport show a similar error to the Calgardup Cave timeseries tending to have a low bias at the start of the observation period and a high bias towards the end.

5.4 Interpretation of water isotopes in paleoclimate studies

Based on data from the 13 year observing period, this study confirms that rainfall intensity is a primary driver of δ^{18}O in precipitation. The nonlinear relationship can be approximated as

$$\delta^{18}\mathrm{O} = \begin{cases} \alpha \log\left(P/P_0\right) + \beta, & P \geq 2\,\mathrm{mm\,day}^{-1} \\ -2.05‰, & P < 2\,\mathrm{mm\,day}^{-1}, \end{cases} \tag{3}$$

where $P_0 = 1\,\mathrm{mm\,day}^{-1}$, $\alpha = -2.85‰$, and $\beta = -1.19‰$. Importantly, years with more intense rainfall are not necessarily wetter overall. In our data, rainfall intensity (precipitation weighted) has a weak correlation with annual rainfall ($R = 0.23$).

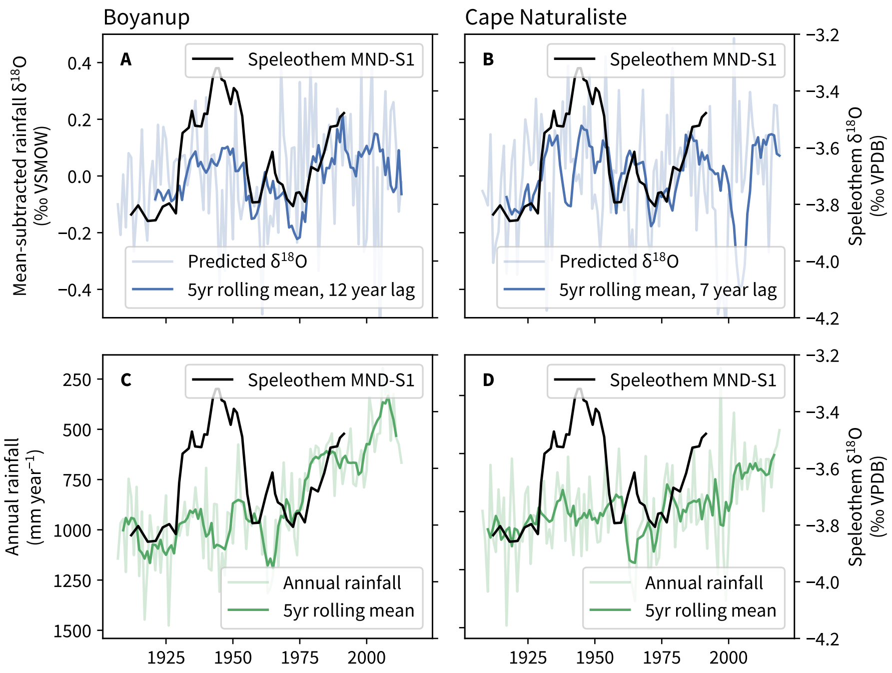

Figure 10. Speleothem MND-S1 from Moondyne Cave δ^{18}O (Treble, Chappell, et al., 2005) compared with **A** rainfall δ^{18}O inferred from Boyanup rainfall intensity, **B** Boyanup annual rainfall, **C** rainfall δ^{18}O inferred from Cape Naturaliste rainfall intensity, **D** Cape Naturaliste annual rainfall. A lag of 12 yr and smoothing with a 5 yr rolling mean has been applied to the inferred δ^{18}O timeseries for comparison with the lower resolution speleothem record which contains both analytical smoothing and attenuation due to karst flow paths. The y-axis for annual total rainfall is inverted to facilitate comparison with the speleothem d18O values.

In Fig. 10 we use 100 yr records of daily rainfall, along with Eq. 3, to hindcast the δ^{18}O timeseries at Boyanup and Cape Naturaliste and compare the δ^{18}O hindcast to a speleothem record from Moondyne Cave to the south (Treble, Chappell, et al., 2005). Although not the closest observations stations to the cave where the speleothem was collected, these are high quality stations (Lavery et al., 1997) in the Australian network (locations shown in Fig. 1) meaning they are sites with long observation records and have been screened for spurious trends. To generate the hindcast, we used only days marked in the record as single-day accumulations, and checked for a weekday dependence to avoid some known quality problems in the Australian record (Viney & Bates, 2004).

Although taking only the leading predictor into account, rainfall δ^{18}O inferred from the Boyanup record displays an intriguing similarity to the Moondyne Cave record, particularly the period of relatively higher speleothem δ^{18}O from 1930–55 and the upwards shift from the mid 1970s. There is also a marked similarity when rainfall intensity is taken from the Cape Naturaliste record, although with a divergence during the 1930–55 period. The disagreement which remains may be the result of nonlinear filtering caused by karst hydrological processes, which has only been accounted for crudely here by a combination of temporal averaging and introducing a time lag. Indeed the time lag, of 12 or 7 years, is longer than suggested by the field evidence which perhaps indicates that uncertainties in the chronology play a role (Nagra et al., 2016, report a 5 year mismatch between the counting method and the documented history of the speleothem). Another complication is that changes in rainfall intensity, inferred from the instrumental record

(Philip & Yu, 2020), are not spatially smooth and, as demonstrated in Fig. 3, even at the annual scale the δ^{18}O timeseries is sensitive to the heaviest events which would impact sites differently, even over short spatial scales.

Supporting the interpretation that rainfall intensity is key to determining δ^{18}O, on daily through to decadal timescales, the trends in annual rainfall accumulations show a weaker relationship with δ^{18}O (Fig. 10c and 10d). Post 1970, for the Boyanup hindcast, a drying trend coincides with a upwards shift in speleothem δ^{18}O. This may be a sign that the interaction between karst hydrology and δ^{18}O changes as the system dries out, but needs detailed investigation before making firm conclusions.

The link between δ^{18}O and rainfall intensity makes δ^{18}O more sensitive to extreme events than would otherwise be the case. In particular, δ^{18}O is more sensitive to extreme events than rainfall accumulations, and therefore is not as smooth spatially. This has implications for the degree of agreement that can be expected between nearby sites, especially over periods of a few decades, because the heaviest few events each year will be sampled differently at different sites. A sustained change in intense rainfall events could be further amplified by karst flowpaths as intense rainfall events are likely to be more effective at initiating recharge of karst stores (Treble et al., 2013).

In the case of deuterium excess, the interpretation of multidecadal records in this region continues to be hampered by an incomplete understanding of governing processes. The strongest predictor on a daily scale, source humidity, makes model predictions worse on an interannual scale. Out of the predictors that we considered, rainfall intensity, measured at the collection site but not along the retroplume, has the strongest effect on d. If this relationship holds over longer timeseries, it would drive an anticorrelation between d and δ^{18}O. Such an anticorrelation was indeed reported by Priestley et al. (2020), in a 35 ka groundwater record, and supports the interpretation by Priestley et al that variations in groundwater isotopes through time, for the Perth Basin, are driven by precipitation intensity.

6 Conclusions

Water isotopes in precipitation were measured daily over thirteen years (2006–18). Daily variability was found to be superimposed on weaker low-frequency trends driven by anomalous conditions in the first three years of monitoring: δ^{18}O decreases by $0.06\pm 0.03\,‰\mathrm{yr}^{-1}$ and d increases by $0.24\pm0.07\,‰\mathrm{yr}^{-1}$, and trends tend to weaken or reverse in the second half of the monitoring period. The factors which drive δ^{18}O and d variability, on a range of timescales, were investigated using generalized additive models (GAMs), with upstream conditions diagnosed with backwards dispersion modelling and synoptic types determined using a statistical method. Although water isotopes demonstrated an association with synoptic types, these were ultimately not a strong driver of variability because, we infer, the synoptic types contained redundant information which was better expressed by continuous values derived from retroplume diagnostics.

Daily variability in δ^{18}O was driven primarily by rainfall intensity, both at the measurement site and upstream, in agreement with the main finding of Fischer and Treble (2008), which was based on a smaller data set. The δ^{18}O seasonal cycle was driven by seasonal changes in both rainfall intensity and source humidity. The relationship between rainfall intensity, at a daily scale, and δ^{18}O was robust. It applied at both the primary measurement station, Calgardup Cave, and to monthly accumulations from Perth Airport. The relationship also appears to be robust over longer time periods, as shown by projecting the $\delta^{18}\mathrm{O} \propto \log(P/P_0)$ relationship back through the ~ 100 yr period with rainfall observations and comparing to a speleothem record. Because of the relationship between rainfall intensity and δ^{18}O, annual accumulations of δ^{18}O are more sensitive to the heaviest rainfall events each year than annual accumulated rainfall is, which has implications both for the interpretation of δ^{18}O records and for how much nearby sites can be expected to agree with each other.

Deuterium excess, d, differed from δ^{18}O in several respects. On a daily scale, variability was driven primarily by h_s, although with a flatter slope than reported in studies of water vapor. The d seasonal cycle was also well explained mainly by h_s, with a weaker contribution from rainfall intensity. In contrast, year-to-year changes in h_s failed to explain the interannual signal in annual mean d, with the implication that multi-decadal, or longer, records of d should not be interpreted as a proxy record of h_s in this region. Furthermore, the link between rainfall intensity and d was too weak to drive the observed changes in d, meaning that the driver for low-frequency changes in d was not fully explained. Further investigation of d is warranted; d is not as sensitive as δ^{18}O to extreme events, there is a low-frequency signal in the observations at both Calgardup Caves and Perth which may be climate-related; meaning that the d signal carries information which supplements δ^{18}O.

7 Acknowledgements

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8 Data availability

The water isotope measurements from Calgardup Cave will be archived with the Global Network for Isotopes in Precipitation (GNIP) should this manuscript be accepted for publication (https://www.iaea.org/services/networks/gnip). Water isotopes measured at Perth Airport are available through GNIP, https://openscience.ansto.gov.au/collection/881. The models used in this study are available for download from http://www.flexpart.eu/ (FLEXPART and FLEXPART-WRF) and http://www.mmm.ucar.edu/wrf/users/ (WRF). Reanalysis data are archived by the European Centre for Medium Range Weather Forecasting (ERA-Interim, https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-interim) and the National Oceanic and Atmospheric Administration (CFSR, https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/climate-forecast-system-version2-cfsv2).

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