



17 **Abstract**

18 The factors driving variability in rainfall stable water isotopes (specifically  $\delta^{18}\text{O}$   
 19 and deuterium excess,  $d = \delta^2\text{H} - 8\delta^{18}\text{O}$ ) were studied in a 13-year dataset of daily rain-  
 20 fall samples from coastal southwestern Western Australia (SWWA). Backwards disper-  
 21 sion modelling, automatic synoptic type classification, and a statistical model were used  
 22 to establish causes of variability on a daily scale; and predictions from the model were  
 23 aggregated to longer temporal scales to discover the cause of variability on multiple timescales.  
 24 Factors differ between  $\delta^{18}\text{O}$  and  $d$  and differ according to temporal scale. Rainfall in-  
 25 tensity, both at the observation site and upwind, was most important for determining  
 26  $\delta^{18}\text{O}$  and this relationship was robust across all time scales (daily, seasonal, and inter-  
 27 annual) as well as generalizing to a second observation site. The sensitivity of  $\delta^{18}\text{O}$  to  
 28 rainfall intensity makes annual mean values particularly sensitive to the year's largest  
 29 events. Projecting the rainfall intensity relationship back through  $\sim 100$  years of pre-  
 30 cipitation observations can explain  $\sim 0.2\text{--}0.4\text{‰}$  shifts in rainfall  $\delta^{18}\text{O}$ . Twentieth cen-  
 31 tury speleothem records from the region exhibit signals of a similar magnitude, indicat-  
 32 ing that rainfall intensity should be taken into account during the interpretation of re-  
 33 gional climate archives. For  $d$ , humidity during evaporation from the ocean was the most  
 34 important driver of variability at the daily scale, as well as explaining the seasonal cy-  
 35 cle, but source humidity failed to explain the longer-term interannual variability.

36 **Plain Language Summary**

37 In cave deposits, as with several other natural systems, the relative abundance of  
 38 the heavy isotopes oxygen-18 and deuterium can be used to determine past changes in  
 39 climate. This is because the isotopic composition of these systems is linked to that of  
 40 rainfall, while the abundance of heavy isotopes in rainfall is driven by climate param-  
 41 eters such as temperature and rainfall characteristics. For this to be possible, the fac-  
 42 tors which drive rainfall isotopic variability need to be well known. This study uses a  
 43 13-year data set of daily rainfall samples from coastal southwestern Western Australia  
 44 to better understand isotopic variability for this region. Oxygen-18 variations here are  
 45 driven mainly by rainfall intensity (the amount of rain each day) both according to mea-  
 46 surements at the site and upwind simulations. Deuterium excess, a second order param-  
 47 eter which is often linked to conditions in the evaporation source region, was well-predicted  
 48 by source region humidity at the daily scale but not when aggregated to annual totals.  
 49 The relationship between rainfall intensity and oxygen-18 appears to be important over  
 50 the 20th century, based on a comparison between observed rainfall and a cave record.

51 **1 Introduction**

52 In systems where material is sequestered from the environment, for instance dur-  
 53 ing speleothem growth or groundwater infiltration, the stable isotope ratios  $\delta^{18}\text{O}$  and  
 54  $\delta^2\text{H}$  act as markers of environmental change. Speleothems, that is cave decorations such  
 55 as stalagmites and flowstones, record changes in the oxygen isotopic composition as they  
 56 grow, and these changes can in turn be linked to changes in rainfall isotopic composi-  
 57 tion (Lachniet, 2009; Orland et al., 2009; Z. Zhang et al., 2018). Karst regions occur through-  
 58 out the midlatitudes (Chen et al., 2017) meaning that cave records can be used to in-  
 59 fer past changes in certain aspects of the hydrological cycle, in areas where this is not  
 60 achievable using materials such as coral and ice (Treble, Chappell, et al., 2005; Lorrey  
 61 et al., 2008; Fohlmeister et al., 2012; McCabe-Glynn et al., 2013; H. Zhang et al., 2018).  
 62 Speleothem use is widespread, with the SISALv2 database alone containing 691 time-  
 63 series of  $\delta^{18}\text{O}$  in speleothem calcite (Comas-Bru et al., 2020).

64 The ratio of deuterium to hydrogen,  $\delta^2\text{H}$ , also reflects changes in hydrological pro-  
 65 cesses. Even though it is not directly preserved in speleothem carbonite, calcareous speleothems

66 nevertheless record the history of  $\delta^2\text{H}$  in infiltrating water because of the formation of  
67 fluid inclusions within the speleothem which trap enough water for isotopic analysis (Vonhof  
68 et al., 2006; van Breukelen et al., 2008). Alternatively, groundwater can be sampled and  
69 dated to obtain a low-resolution record of both  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  (Priestley et al., 2020).

70 Interpreting these records, of oxygen-18 in calcite and deuterium in fluid inclusions,  
71 relies on an understanding of how climatic and atmosphere processes drive isotope vari-  
72 ability in rainfall. At the laboratory scale, this is well understood. In a closed system,  
73 heavier isotopes are concentrated in the more condensed phase according to the temperature-  
74 dependent equilibrium fractionation factor (Majoube, 1971; Horita & Wesolowski, 1994). In  
75 well-controlled conditions where diffusive transport is important, the difference in molec-  
76 ular diffusivity between isotopologues (Merlivat, 1978b) leads to quantitatively-predictable  
77 kinetic fractionation. In the climate system, however, precisely which climatic and at-  
78 mospheric processes emerge with the strongest link to isotopic variations is less clear and  
79 differs between regions.

80 Towards the poles, over long time scales, oxygen and hydrogen isotopes in ice have  
81 been used as an indicator of temperature (Brook & Buizert, 2018; Jouzel et al., 2007);  
82 whereas tropical rainfall isotopes have classically been thought of as being controlled by  
83 precipitation amount (Dansgaard, 1964). Other factors are also important, though, some  
84 of which are location-dependent. In the tropics, these factors include the degree of con-  
85 vective organization (Moerman et al., 2013) or monsoon activity (Okazaki et al., 2015).  
86 In both the tropics and midlatitudes, the type of precipitation (Aggarwal et al., 2016)  
87 and atmospheric residence time (Aggarwal et al., 2012) are important. In studies from  
88 the midlatitudes, the moisture source (Krklec & Domínguez-Villar, 2014) and, more gen-  
89 erally, the airmass history (Deininger et al., 2016; Good et al., 2014) have been identi-  
90 fied as drivers of isotopic variability.

91 One way to simplify the analysis of many individual factors, and potentially mak-  
92 ing interpretation more robust or straightforward, is to examine the link between the type  
93 of synoptic-scale weather system and water isotopes in precipitation. Using this approach  
94 in Southern Australia (Barras & Simmonds, 2008, 2009; Treble, Budd, et al., 2005; Guan  
95 et al., 2013), and elsewhere (Lykoudis et al., 2010; Farlin et al., 2013; Tyler et al., 2016;  
96 Wang et al., 2017; Schlosser et al., 2017), has indeed revealed that an association exists.  
97 It arises because several of the factors mentioned above systematically differ between syn-  
98 optic types.

99 This study is concerned with southwestern Western Australia (SWWA) in the South-  
100 ern Hemisphere midlatitudes. Here,  $\delta^{18}\text{O}$  values in speleothem records (Treble, Chap-  
101 pell, et al., 2005) have low frequency variations that are likely to be linked to climate,  
102 but a robust understanding of the mechanism is incomplete. Treble, Chappell, et al. (2005)  
103 showed that the stable water isotopes measured in SWWA daily rainfall samples, over  
104 a one-year study period, are associated with rainfall intensity, but other drivers may also  
105 play a role. It is also unclear whether the intensity dependence holds over longer time  
106 periods. An understanding of these drivers is particularly important for this region; win-  
107 ter rainfall here has dropped significantly since the 1970s (Bates et al., 2008) and plac-  
108 ing this in the context of the region’s long-term natural variability is important for fully  
109 understanding the change. This is a challenging task because of the region’s strong in-  
110 ternal variability, demonstrated in climate models (Cai et al., 2005; England et al., 2006),  
111 combined with a short ( $\sim 100$  yr) instrumental record (Haylock & Nicholls, 2000).

112 There are several approaches for determining climate variability using paleoclimate  
113 records or reconstructions. Changes in rainfall have been inferred from distant measure-  
114 ments of snow accumulation (Zheng et al., 2021), which is possible because of an anti-  
115 correlation between SWWA May–October rainfall and snowfall at Law Dome, Antarc-  
116 tica (van Ommen & Morgan, 2010). Speleothem records, an in situ climate proxy, are  
117 found in caves which develop in Tamala Limestone (Geoscience Australia & Australian

118 Stratigraphy Commission, 2017), an eolian carbonate deposited in the Middle to Late  
 119 Pleistocene,  $\sim 10$ –250 ka before present (Smith et al., 2012). Tamala Limestone is exten-  
 120 sively distributed along several hundred kilometers of the Western Australian coastline  
 121 (Fig. 1). Meanwhile, groundwater from the confined aquifers of the Perth Basin (Priestley  
 122 et al., 2020) has been interpreted as a low-resolution record of infiltration. Both speleothem  
 123 and groundwater records would benefit from a better understanding of the climate drivers  
 124 of stable water isotopes.

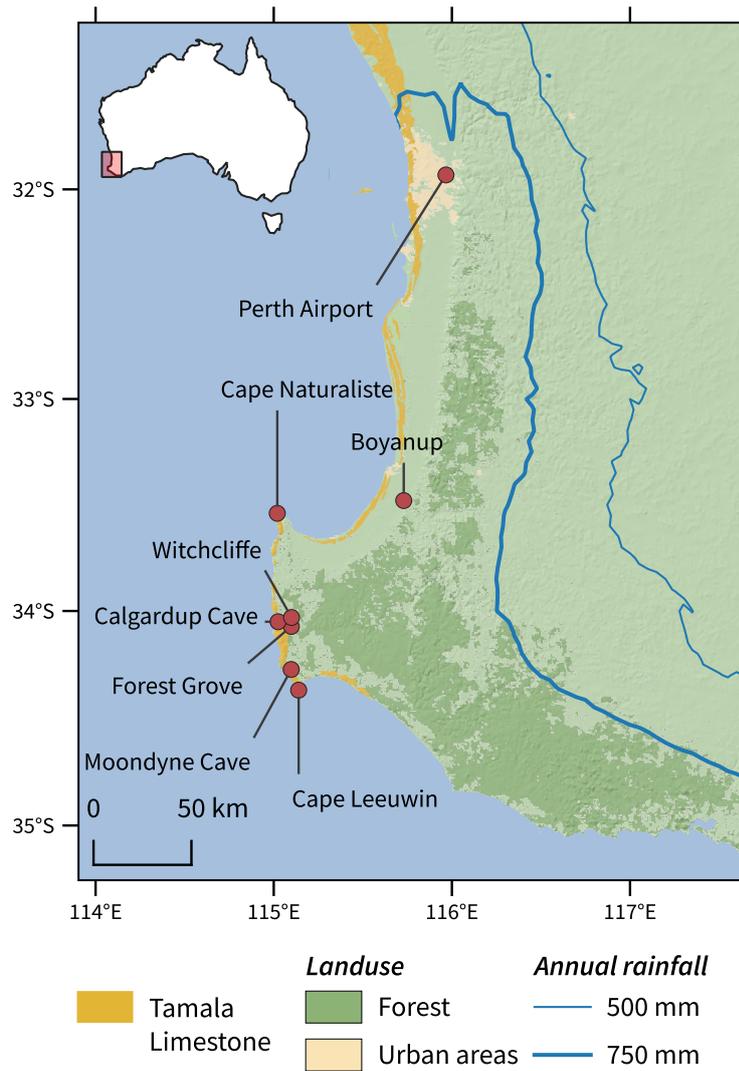
125 The purpose of this paper, then, is to investigate the factors which influence the  
 126 abundance of stable water isotopes ( $^2\text{H}\text{H}\text{O}$ ,  $\text{H}_2^{18}\text{O}$ ) in a modern 13 yr record of SWWA  
 127 rainfall, taking into account day-to-day variations in synoptic types, upstream conditions,  
 128 and site parameters. In particular, our goal is to identify factors which are important  
 129 both at the daily, seasonal, and annual scales. This is most relevant to understanding  
 130 speleothem records from the region, although we expect the measurements to be more  
 131 widely useful.

132 The remainder of this paper is organized as follows: Sect. 2 describes the charac-  
 133 teristics of the study region; Sect. 3 introduces the methods used in this study, includ-  
 134 ing a Lagrangian trajectory model and statistical methods; Sect. 4 describes the main  
 135 results and illustrates links between water isotopes and their drivers; and Sect. 5 com-  
 136 pares our results with the literature, tests the ability of our interpretation to general-  
 137 ize to another site, as well as summarizing implications for speleothem record interpre-  
 138 tation.

## 139 2 Regional setting

140 The coastal region of southwestern Western Australia (SWWA, Fig. 1), has an an-  
 141 nual rainfall of more than 750 mm making it a wet and productive region in compari-  
 142 son to the arid inland. The region is too warm for snow, so precipitation falls as rain and  
 143 this is mostly during the cooler months of May–October (Bates et al., 2008, Fig. S1). The  
 144 total cool-season rainfall is closely related to the number of fronts which cross the coast,  
 145 which in turn is coupled to the strength and extent of the Hadley-Walker circulation (Rudeva  
 146 et al., 2019). Along the coastline south of Perth, about 50% of winter rainfall is asso-  
 147 ciated with fronts, which can be accompanied by thunderstorms (Pepler et al., 2020),  
 148 20% with cutoff lows (low pressure systems formed at upper tropospheric levels), and  
 149 the remainder with warm troughs and other synoptic systems (Pook et al., 2012). Fur-  
 150 ther inland, the proportion of frontal rainfall is lower, and the climate is dryer. Other  
 151 studies, although differing in how synoptic systems are defined (Hope et al., 2014), have  
 152 generally classified rainfall-bearing systems into similar synoptic types (Hope et al., 2006;  
 153 Raut et al., 2014) and agree on the importance of frontal rainfall during the rainy win-  
 154 ter season. In summer, when the subtropical ridge lies over the region, monthly rainfall  
 155 of 20 mm or less is typical and frontal rainfall makes up a smaller proportion of the to-  
 156 tal. Instead, rainfall comes from a mixture of thunderstorms, extratropical cyclones, (Pepler  
 157 et al., 2020) and warm troughs (Raut et al., 2014). Also more likely during summer are  
 158 the rare, but potentially extreme, events from ex-tropical cyclones (Foley & Hanstrum,  
 159 1994).

160 As well as having a pronounced seasonal cycle, the region’s rainfall has changed  
 161 on interannual to decadal timescales. Since 1970, the water inflow to Perth’s dams has  
 162 decreased by half (Power et al., 2005), due to the combined effect of reduced winter rain-  
 163 fall and increased evaporation. The rainfall intensity distribution has also changed over  
 164 the instrumental period, but with differences between stations within SWWA (Philip &  
 165 Yu, 2020). A number of studies, reviewed by Dey et al. (2019), show the rainfall decrease,  
 166 in winter, is associated with a change in regional circulation including a poleward shift  
 167 in westerly winds. The resulting decrease in the frequency of strong fronts (Raut et al.,  
 168 2014) has been related to a significant warming of the Southern Hemisphere troposphere



**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).

169 south of 30°S followed by a decrease in the strength of the jetstream, which, in turn, de-  
 170 creases the instability and makes the formation of synoptic disturbances less likely (Frederiksen  
 171 & Frederiksen, 2007). This is in agreement with a recent study by Lucas et al. (2021),  
 172 who described a reduction in the intensity of the upward midlatitude circulation branch  
 173 in the Southern Hemisphere at 30°S. Climate model projections indicate that the dry-  
 174 ing trend will continue (Bates et al., 2008; Raut et al., 2016).

### 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 for most of the record, however Western Australia observed daylight saving time during the summers of 2006–2009) and on days with at least 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 at 3 °C until analysis. For this study, measurements were included from the years 2006–2018 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  $\delta^{18}\text{O}$  of  $-1.2\text{‰}$  with 52 mm of rainfall, compared to an expected value of about  $-5\text{‰}$  for this amount of rainfall. Three rain-days later a sample was anomalously low ( $-5.0\text{‰}$  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 ( $\text{H}_2^{18}\text{O}/\text{H}_2\text{O}$ ) and deuterium ( $^2\text{HHO}/\text{H}_2\text{O}$ ) relative to Vienna Standard Mean Ocean Water (VSMOW; IAEA, 2006) in rainwater. We use delta notation where  $\delta = R/R_{\text{VSMOW}} - 1$ , with  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  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  $\pm 1.0\text{‰}$  for  $\delta^2\text{H}$  and  $\pm 0.1\text{‰}$  for  $\delta^{18}\text{O}$ ). All samples were filtered prior to analysis and data were reported against in-house standards calibrated to VSMOW/VSMOW2 and SLAP/SLAP2.

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

$$d = \delta^2\text{H} - 8 \delta^{18}\text{O}. \quad (1)$$

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  $\delta^{18}\text{O}$  at cool temperatures. This trend reverses, however, once Rayleigh distillation proceeds far enough (less than about 10% of vapor remaining) meaning that Rayleigh theory predicts that dry mid-tropospheric air has low  $\delta^{18}\text{O}$  and high  $d$ , in general agreement with observations (Sodemann et al., 2017).

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).

225 In this paper, the amount of rainfall collected each day is called the ‘rainfall inten-  
 226 sity’, in contrast to ‘rainfall total’ which is the accumulated rainfall over a longer period.  
 227 Where averages of  $\delta^{18}\text{O}$  and  $d$  are computed, these are weighted by rainfall amount un-  
 228 less noted otherwise.

### 229 3.2 Source region diagnostic

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

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

249 Three of the uncertainties in the approach are that: the time of rainfall is only known  
 250 to within a 24 h sampling window; the appropriate height for beginning the backwards  
 251 plume has to be estimated; and the error in the plume grows as the model is integrated  
 252 further back in time. After some experimentation, the beginning time was taken from  
 253 the time in the WRF simulation with the largest rain rate, and the starting height was  
 254 taken to be the cloud base in WRF, estimated at the height when relative humidity reaches  
 255 80%. Then, to verify that the model indeed produces a useful diagnostic, we checked the  
 256 correlation between  $d$  and humidity relative to saturation at the sea surface tempera-  
 257 ture,  $h_s$ , as a function of back trajectory length. This is a useful diagnostic because  $d$ ,  
 258 in vapor, and  $h_s$ , at the evaporation site, are strongly correlated (Pfahl & Wernli, 2008),  
 259 and we assume that  $d$  will be approximately conserved during the conversion of water  
 260 vapor into clouds and then rainfall.

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

### 265 3.3 Synoptic classifications

266 On each day, the synoptic type was classified with a Self Organizing Map (SOM),  
 267 using SOM-PAK (Kohonen et al., 1996), following the approach described by Hope et  
 268 al. (2006). Synoptic types were derived from the 1200 UTC mean sea level pressure (MSLP)  
 269 anomaly fields of the ERA-Interim reanalysis on a  $0.75^\circ$  latitude/longitude grid in the  
 270 region  $90\text{--}130^\circ\text{E}$ ,  $50\text{--}15^\circ\text{S}$ . The SOM is an unsupervised classification method, produc-  
 271 ing synoptic types that are arranged in a two-dimensional grid. The arrangement of types  
 272 into a grid, where similar synoptic types are arranged close to each other, is the main  
 273 way in which the SOM differs from other statistical classification techniques (Philipp et

274 al., 2016). Synoptic classification was only applied to the rainy months (April–October),  
 275 due to the presence of seasonally persistent features in the surface pressure field asso-  
 276 ciated with the meridional movement of the subtropical high pressure ridge. Training  
 277 was performed using data from the years 1979–2018 and grid cells were weighted by area.

278 In addition to the SOM classifications, fronts were detected in the reanalysis fields  
 279 and used as an aid to interpret the SOM classifications. The position of fronts was found  
 280 using the wind shift method (Simmonds et al., 2011) based on ERA-Interim 3-hourly 10 m  
 281 wind fields. This is a straightforward method which is applicable to SWWA (Hope et  
 282 al., 2014). It does not produce spurious fronts along the coastline, that are often found  
 283 by a more commonly used methods based on the temperature gradients (Pepler et al.,  
 284 2020). The wind-based method works well to define meridionally elongated fronts, that  
 285 are mainly cold fronts, and is particularly well suited for the Southern Hemisphere (Schemm  
 286 et al., 2015).

### 287 3.4 Generalized additive models

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

296 The GAM implementation was provided by `mgcv`, a package for R (R Core Team,  
 297 2014). Relationships between predictor and response variables are modelled with penal-  
 298 ized regression splines in which the smoothness is estimated during the fitting process  
 299 using restricted maximum likelihood (REML; Wood, 2011), and models used the iden-  
 300 tity link function. In this implementation, predictors which can be modelled with a lin-  
 301 ear response are modelled that way, and predictors with insufficient explanatory power  
 302 are dropped from the model. The `mgcv` models can also incorporate categorical variables,  
 303 allowing the synoptic classification to be included within the same framework.

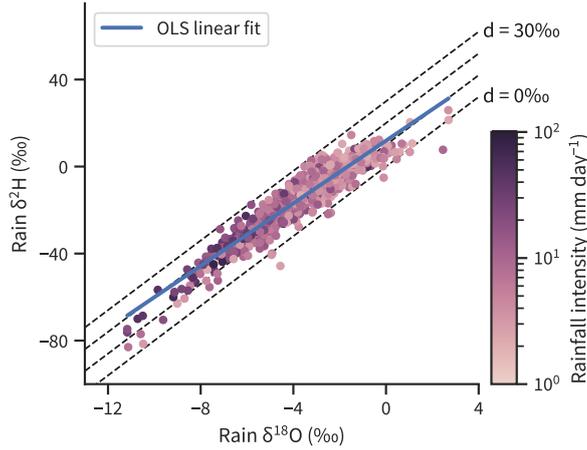
304 In this study, we also assessed the importance of terms for explaining the obser-  
 305 vations on different timescales. As well as allowing the models to drop unimportant terms  
 306 (using REML) we followed a procedure where models were constructed term-by-term.  
 307 Beginning with an empty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

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

$$314 \text{MSE} = \frac{1}{N} \sum_{i=1}^N (\hat{y}_i - y_i)^2 \quad (2)$$

315 where  $N$  is the number of observations,  $y_i$  is the  $i$ th day’s observation and  $\hat{y}_i$  is the  $i$ th  
 316 day’s model prediction. This is repeated for all years in the data set, and the MSE is taken  
 317 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.

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



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

321

### 3.5 Modelled precipitation isotopes

322

323

324

325

326

327

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  $2.5^\circ$ .

328

329

330

331

332

At other sites, IsoGSM reproduces daily, monthly, and seasonal variability in water isotope ratios, with more skill at simulating  $\delta^{18}\text{O}$  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.

333

## 4 Results

334

335

336

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.

337

### 4.1 Daily $\delta^{18}\text{O}$ , $\delta^2\text{H}$ , and precipitation

338

339

340

341

342

343

Over the 13-year monitoring period (2006-18 inclusive, days with  $\geq 2 \text{ mm day}^{-1}$  of rainfall) the precipitation-weighted mean (the mean weighted by the daily precipitation amount)  $\delta^{18}\text{O}$  was  $-4.45\text{‰}$ ,  $d$  was  $15.4\text{‰}$ , and  $\delta^2\text{H}$  was  $-20.2\text{‰}$ . 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  $\delta^{18}\text{O} \sim \delta^2\text{H}$  space, are strongly correlated and lie about the so-called local meteoric water line (LMWL; Fig. 2).

344

345

346

There is a tendency for intense rainfall to have lower  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  and for low intensity rainfall to both have high  $\delta^{18}\text{O}$  and, above  $-2\text{‰}$ , depart from the straight line trend. Deuterium excess for these high  $\delta^{18}\text{O}$  samples tends towards the  $d = 0\text{‰}$  line,

347 contrasting to the overall mean  $d$ . For comparison, the global meteoric water line (GMWL)  
 348 of Craig (1961) lies on the  $d = 10\%$  line. In common with many Australian sites (Hollins  
 349 et al., 2018), the slope of the LMWL when calculated with ordinary least-squares (OLS)  
 350 is lower than the GMWL. The parameters for straight-line fits to the daily rainfall sam-  
 351 ples are shown in Tab. S1, with both ordinary least-squares and precipitation weighted  
 352 least squares (WLS, Hughes & Crawford, 2012), although these are not the only options  
 353 for characterizing the LMWL and the slope is dependent on the regression method (Crawford  
 354 et al., 2014). Taking uncertainty into account, the slope of the LMWL at Calgardup Cave  
 355 is indistinguishable from the Perth Airport LMWL, but there is an offset between  
 356 the two sites since the intercept differs by about two standard deviations. The cause of  
 357 this offset is explained in Sect. 5.3.

358 As noted in Sect. 3.1, the temperature-dependence of equilibrium fractionation would  
 359 lead to an increase in  $d$  with  $\delta^{18}\text{O}$ . Here we see the opposite trend, which is indicative  
 360 of non-equilibrium processes, such as sub-cloud raindrop re-evaporation (Lee & Fung,  
 361 2008), becoming relatively more important during light rainfall.

## 362 4.2 Seasonal cycle

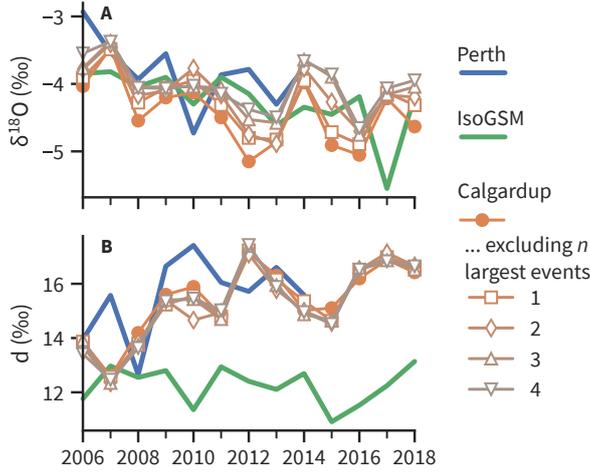
363 The composite seasonal cycle of  $\delta^{18}\text{O}$ ,  $d$ , and rainfall has been published previously  
 364 for Perth (Hollins et al., 2018; Liu et al., 2010) and the seasonal cycle at Calgardup is  
 365 broadly similar (shown later in Fig. 8, but also in Fig. S1). The similarity is consistent  
 366 with isotopes at the two sites being driven by similar factors. As shown in these figures,  
 367 the  $\delta^{18}\text{O}$  minimum occurs in May or June, which is earlier than the July peak in rain-  
 368 fall. January stands out as an exception with anomalously low—and variable—rainfall  $\delta^{18}\text{O}$   
 369 when compared with the surrounding months, likely because of the occurrence of rare,  
 370 but intense, rainfall events. The seasonal cycle of  $d$  also has a large amplitude, but mir-  
 371 rors  $\delta^{18}\text{O}$  with a peak in the rainy months. Unlike  $\delta^{18}\text{O}$ , summer variability is not es-  
 372 pecially pronounced.

## 373 4.3 Annual mean time series

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

382 On average, annual  $\delta^{18}\text{O}$  values are 0.61‰ higher at Perth implying a meridional  
 383 gradient in  $\delta^{18}\text{O}$  of 0.29‰ per degree of latitude. This agrees with a persistent feature  
 384 of isotope enabled GCMs which simulate a  $\delta^{18}\text{O}$  maximum over the Indian Ocean north  
 385 of Perth, near 30°S and under the descending branch of the Hadley Cell, with decreas-  
 386 ing values towards the pole (Werner et al., 2011; Lee et al., 2007; Noone & Simmonds,  
 387 2002; Risi et al., 2012). The offset between mean values for Perth and Calgardup Cave  
 388 shows no trend through time, implying that the meridional gradient has remained con-  
 389 sistent over the monitoring period.

390 Annual mean departures from the trend are not consistent between sites (Fig. 3a),  
 391 suggesting that  $\delta^{18}\text{O}$  anomalies are related to local processes. At least in part, the low  
 392 correlation between sites is because annual mean  $\delta^{18}\text{O}$  is particularly sensitive to the heav-  
 393 iest events of the year, shown by plotting four similar time series in which the heaviest  
 394 1–4 rainfall events from each year are excluded. Excluding the heavy events shifts the  
 395 mean  $\delta^{18}\text{O}$  higher and, in years like 2015 and 2018, can change annual means from anoma-



**Figure 3.** Annual precipitation-weighted mean **A**  $\delta^{18}\text{O}$  and **B**  $d$  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  $\delta^{18}\text{O}$  variations, but not  $d$  variations, to a few events. Results from the IsoGSM isotope-enabled general circulation model are also shown.

396 lously low to high. As with any rainfall event, these heavy events will be sampled dif-  
 397 ferently by the two monitoring sites (Good et al., 2014), so stochastic variability is a ma-  
 398 jor contributor to the annual precipitation-weighted mean  $\delta^{18}\text{O}$ . In contrast to  $\delta^{18}\text{O}$ , the  
 399 interannual variability in  $d$  is not as strongly affected by these intense rainfall events (Fig. 3b),  
 400 so the annual-mean difference between Perth and Calgardup Cave time-series are not  
 401 as sensitive to stochastic variability.

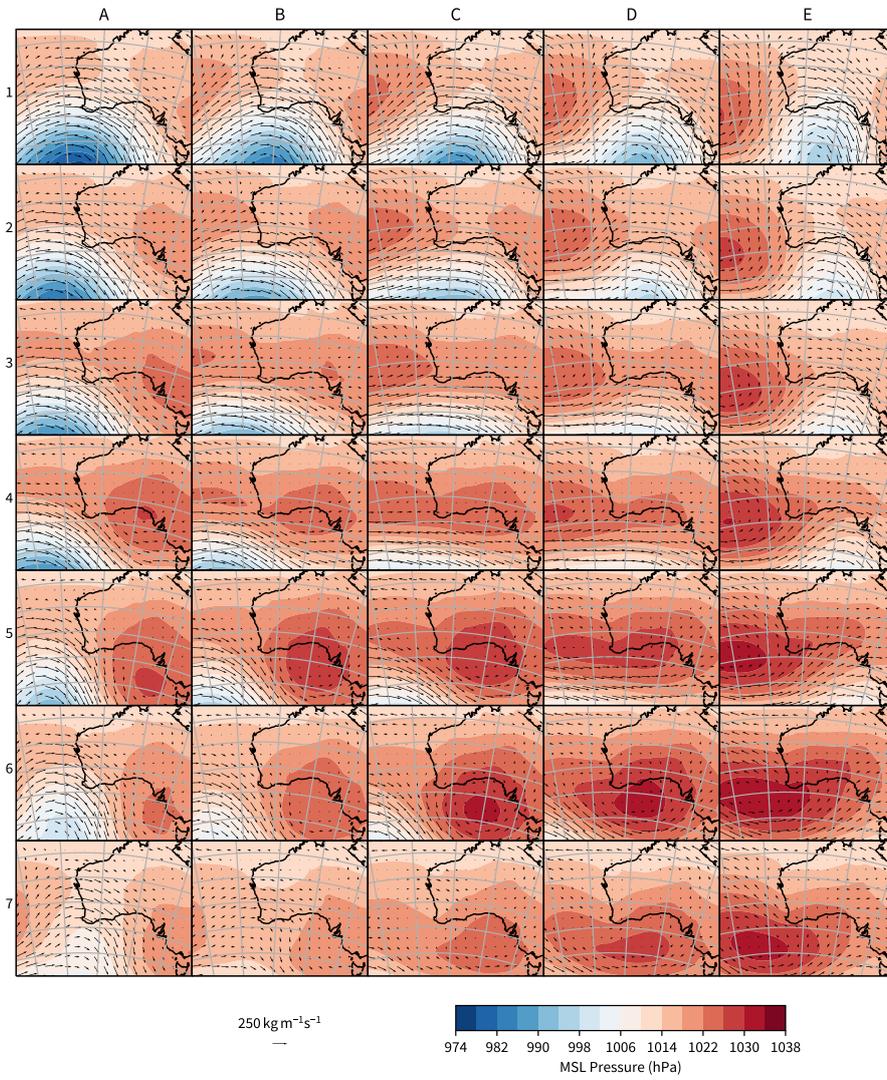
402 To examine the factors which drive these long-term changes, and the seasonal cy-  
 403 cle, we analyze the conditions on each rainy day in the following sections.

404 **4.4 Synoptic systems**

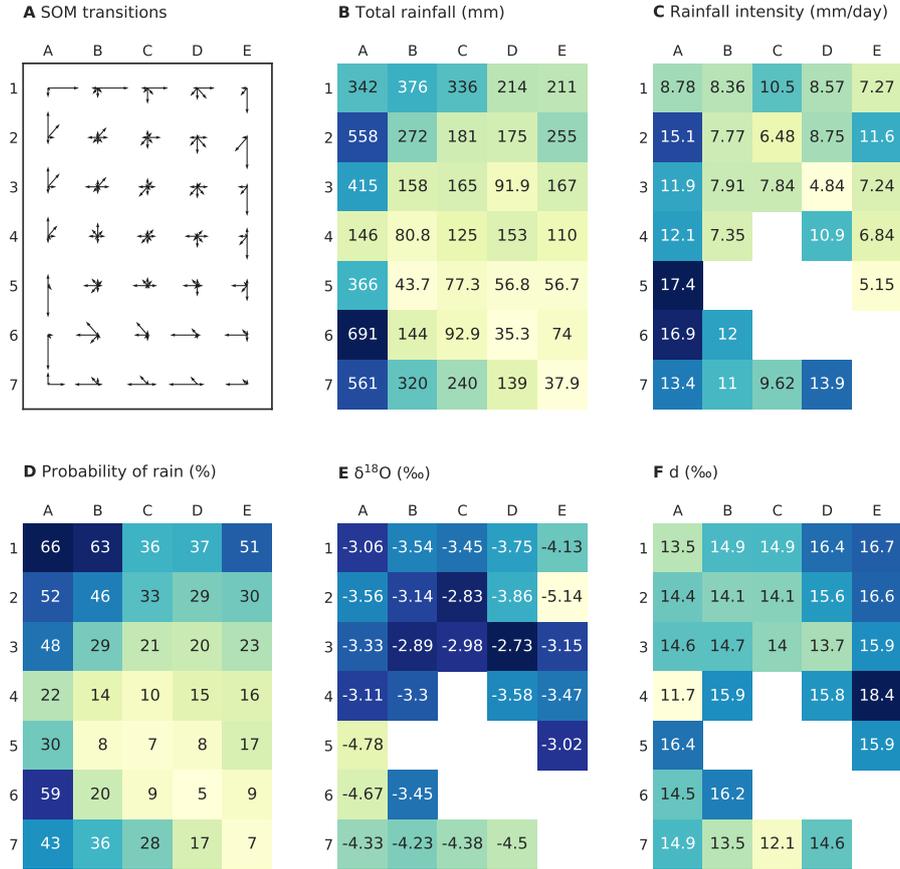
405 Self organizing maps (SOMs) were used to classify synoptic regimes. We identified  
 406 35 synoptic types using MSLP fields from ERA-Interim, and each day was associated  
 407 with one of types shown in Fig. 4. Supplementary interpretation is provided by the frontal  
 408 density and 500 hPa height fields in Fig. S3, and Fig 5 summarizes several observations  
 409 according to synoptic type.

410 Although the SOM is not derived directly from frontal information, the location  
 411 of fronts is related to the surface pressure field and the synoptic types are therefore as-  
 412 sociated with front positions. The top two rows in the SOM are most strongly associ-  
 413 ated with the presence of rain-bearing cold fronts directly over SWWA, while the sequence  
 414 around the outside edge of the SOM, A4 ··· A1 ··· E1, tracks the progress of cold fronts  
 415 beginning offshore to the west and moving east across the region. This is a common oc-  
 416 currence, and appears as a path with high transition probabilities in Fig. 5a. Types in  
 417 the top left are more representative of pre-frontal rainfall, while types in the top right  
 418 are post-frontal.

419 Synoptic types away from the top rows are not as strongly associated with frontal  
 420 rainfall (Fig. S3); although fronts are detected they are generally away from SWWA. No-  
 421 tably, the pressure pattern for classes A5, A6 resembles a trough, associated with mois-



**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  $\delta^{18}\text{O}$ ; **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, E, and F.

422 ture transport from the northwest, and A7 is a blend between a trough and cutoff low.  
 423 These three classes, in general, are related to upper-tropospheric processes with fronts  
 424 being detected too far to the west to be responsible for rainfall.

425 As shown in Fig. 5 synoptic types are a reasonable predictor of rainfall properties,  
 426 several of which show a strong dependence on SOM classification. In particular, the wettest  
 427 class (A1) has a rainfall probability of 66%, much higher than the driest class with 5%  
 428 probability of rain (Fig. 5d). Rainfall intensity (Fig. 5c) is also sensitive to synoptic type,  
 429 with column A showing the most intense rainfall, especially for classes A5 and A6. Al-  
 430 though these non-frontal classes are associated with heavy rain, and A6 accounts for the  
 431 highest total precipitation, frontal events are responsible for more rainfall overall as they  
 432 occupy a larger number of classes. Based on manual classifications, Pook et al. (2012)  
 433 also found that fronts were responsible for most winter rainfall.

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

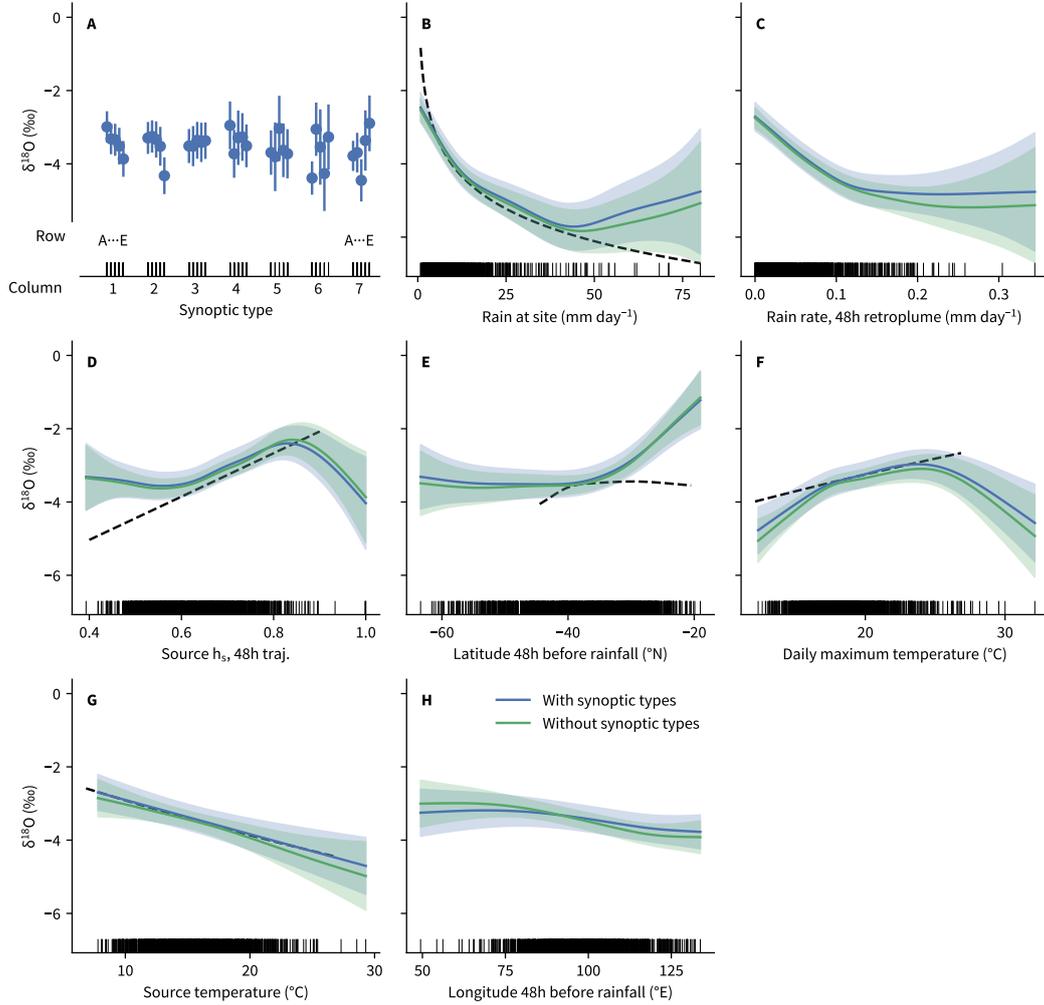
440 These observations are consistent with other studies (Treble, Budd, et al., 2005;  
 441 Barras & Simmonds, 2008) which have demonstrated, in the Australian region, that dif-  
 442 ferent types of synoptic systems can have distinct isotopic signatures, an effect which is  
 443 replicated at sites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

447 The SOM analysis, while showing an association between synoptic types and iso-  
 448 topes, does not by itself identify the reasons behind the association. Furthermore, although  
 449 there is a relatively large difference between frontal and non-frontal rainfall,  $\delta^{18}\text{O}$  dif-  
 450 fering by 1–1.7‰, this difference is not large enough to explain the year-by-year variabil-  
 451 ity (Fig. 3). Year-by-year changes can reach 1‰, meaning that rainfall would need to  
 452 switch from almost exclusively frontal rainfall to non-frontal to explain the changes in  
 453 annual mean  $\delta^{18}\text{O}$ , and this is not something which is observed. In the next section, up-  
 454 stream conditions, diagnosed from dispersion modelling, are combined with site-based  
 455 observations and synoptic types to gain more insight into the underlying processes.

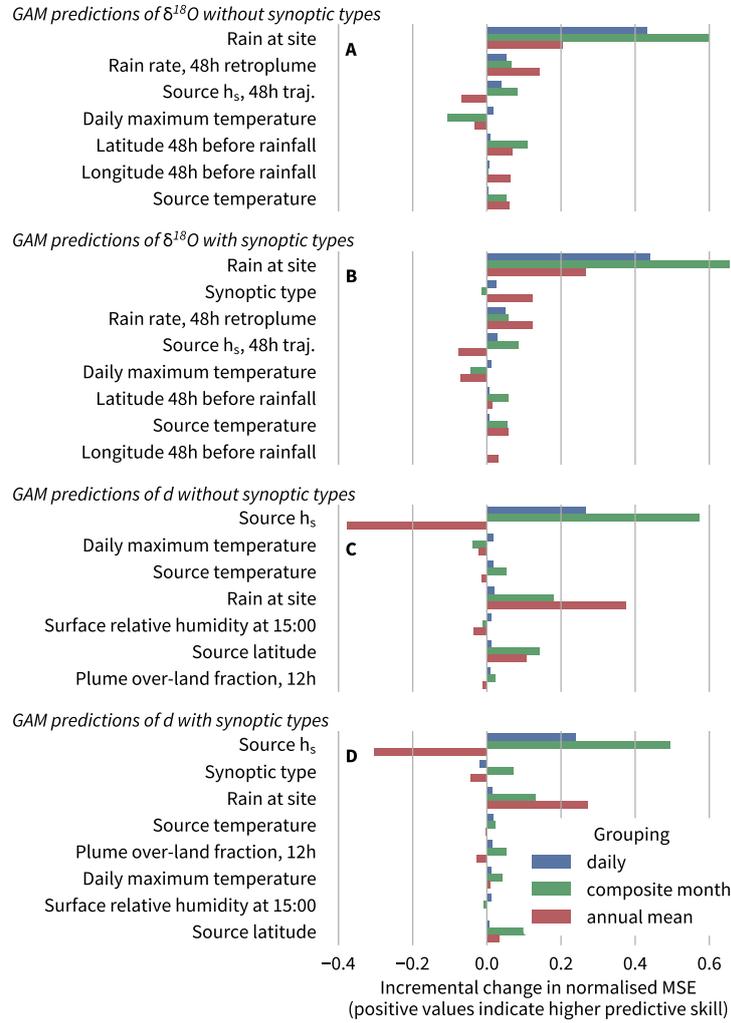
#### 456 4.5 Generalized additive model for $\delta^{18}\text{O}$

457 Generalized additive models (GAMs) trained to predict daily rainfall  $\delta^{18}\text{O}$  are shown  
 458 in Fig. 6. These curves are the model’s ‘smooth terms’, that is the smooth functions ex-  
 459 pressing the relationship between predictor variables and the response variable. Two mod-  
 460 els are shown, one with synoptic types (trained on data from the wet months, April–October)  
 461 and another without synoptic types (trained on data from the entire year). In this fig-  
 462 ure, smooth terms are ordered according to how much they improve the daily mean-square  
 463 error. This, and other metrics for judging the importance of terms, is shown in Fig. 7.

464 For predictions of daily  $\delta^{18}\text{O}$ , the most important smooth terms in this model are:  
 465 the locally-recorded rainfall intensity,  $P$ ; the mean rainfall intensity along the backwards  
 466 plume,  $\bar{P}$ ; then source humidity,  $h_s$  relative to the sea surface temperature. Local rain-  
 467 fall intensity is the best predictor of daily  $\delta^{18}\text{O}$ , the seasonal cycle, and year-to-year vari-  
 468 ability (Fig. 7), it follows a relationship which is close to  $\delta^{18}\text{O} \propto \log(P)$ . Adding the  
 469 rainfall intensity, along the backwards plume, improves the model’s fit to interannual vari-  
 470 ability, by almost as much as  $P$ , but does not affect its fit to the seasonal cycle. The third  
 471 term,  $h_s$  is defined as the humidity in the evaporation region relative to the sea surface



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



**Figure 7.** Prediction accuracy of GAM: **A**  $\delta^{18}\text{O}$  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) due to the addition of a predictor, compared with a simpler model which does not include that predictor. The simplest model, which begins the sequence, is 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.

472 temperature. It is calculated from atmospheric properties within the lowest model level  
 473 and weighted by evaporation rate. Source humidity is important for the  $\delta^{18}\text{O}$  seasonal  
 474 cycle, but not interannual variability. Even more than that, the inclusion of  $h_s$  increases  
 475 the model error for the prediction of interannual variability.

476 Remaining terms do not make a major difference to the model’s predictive abil-  
 477 ity at interannual scales (Fig. 7). Nevertheless, the starting latitude and longitude of the  
 478 plume, along with the source temperature and backwards-plume overland fraction, are  
 479 detected in the model as having an influence on  $\delta^{18}\text{O}$ , and are discussed further in Sect. 5.

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

488 A comparison of GAM predicted  $\delta^{18}\text{O}$  with observed timeseries is shown in Fig. 8a  
 489 and 8b showing that the GAM successfully tracks  $\delta^{18}\text{O}$  interannual variability and the  
 490 seasonal cycle.

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

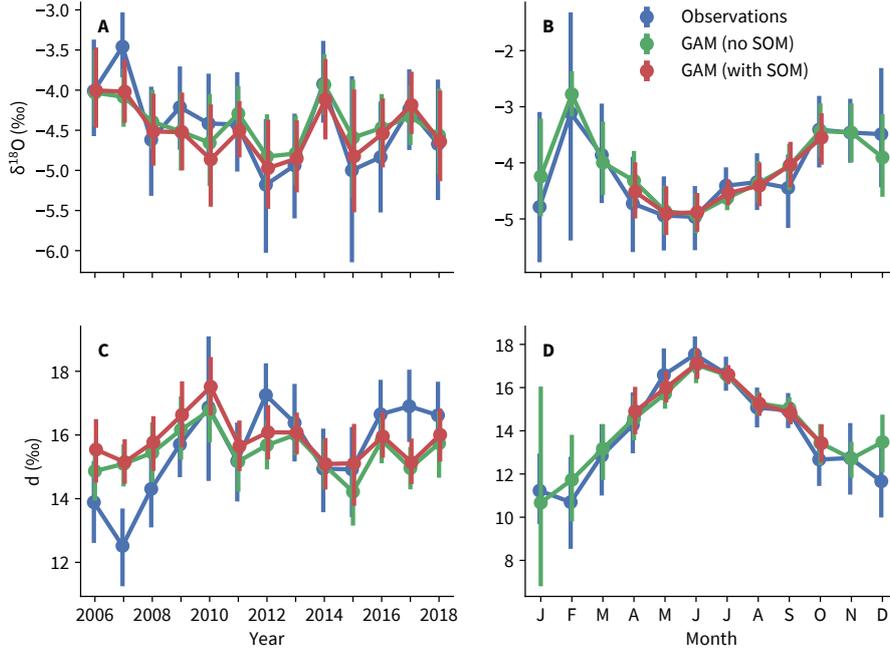
#### 497 4.6 Generalized additive model for deuterium excess

498 Rainfall  $d$  differs from  $\delta^{18}\text{O}$  both in terms of which predictors are important, and  
 499 how well a GAM trained on daily data is able to predict interannual variability. As with  
 500  $\delta^{18}\text{O}$ , a GAM was trained using daily data and then used to predict aggregate values over  
 501 longer periods. This process was repeated with another GAM which included synoptic  
 502 types.

503 The leading predictor of daily  $d$  is source humidity relative to saturation at the sea  
 504 surface,  $h_s$ . This is followed by source temperature,  $T_s$ , site temperature (daily maxi-  
 505 mum temperature at Calgardup Cave from a gridded data set; Jones et al., 2009), and  
 506 rainfall intensity,  $P$ . Compared with  $h_s$ , the remaining terms only weakly improve the  
 507 MSE at the daily scale (Fig. 7c), but only the inclusion of  $P$  is able to improve the annual-  
 508 mean predictions, relative to a prediction of constant  $d$ .

509 The effect of adding synoptic types to the  $d$  model, which also means restricting  
 510 the model to rainy months, is shown in Fig. 7d. Synoptic types, although statistically  
 511 important according to the REML test, fail to improve the cross-validated MSE at the  
 512 daily or interannual time scales. As with  $\delta^{18}\text{O}$ , the information introduced to the model  
 513 by the synoptic types is redundant, and reduces the cross-validated performance of the  
 514 model, possibly because the large number of categories promotes over-fitting.

515 Of all the factors in this analysis, however, it is the source humidity which stands  
 516 out. It is strongly linked to  $d$  at the daily scale, it is apparently the main driver of the  
 517 observed seasonal cycle, but using it to predict interannual variability produces a very  
 518 poor model—one which has a larger error than a model without  $h_s$ . When plotted along-  
 519 side observations, the annual mean predictions of  $d$  (Fig. 8c) show that, in contrast to  
 520 the case of  $\delta^{18}\text{O}$ , the GAM is unable to follow the overall increasing trend in observed



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

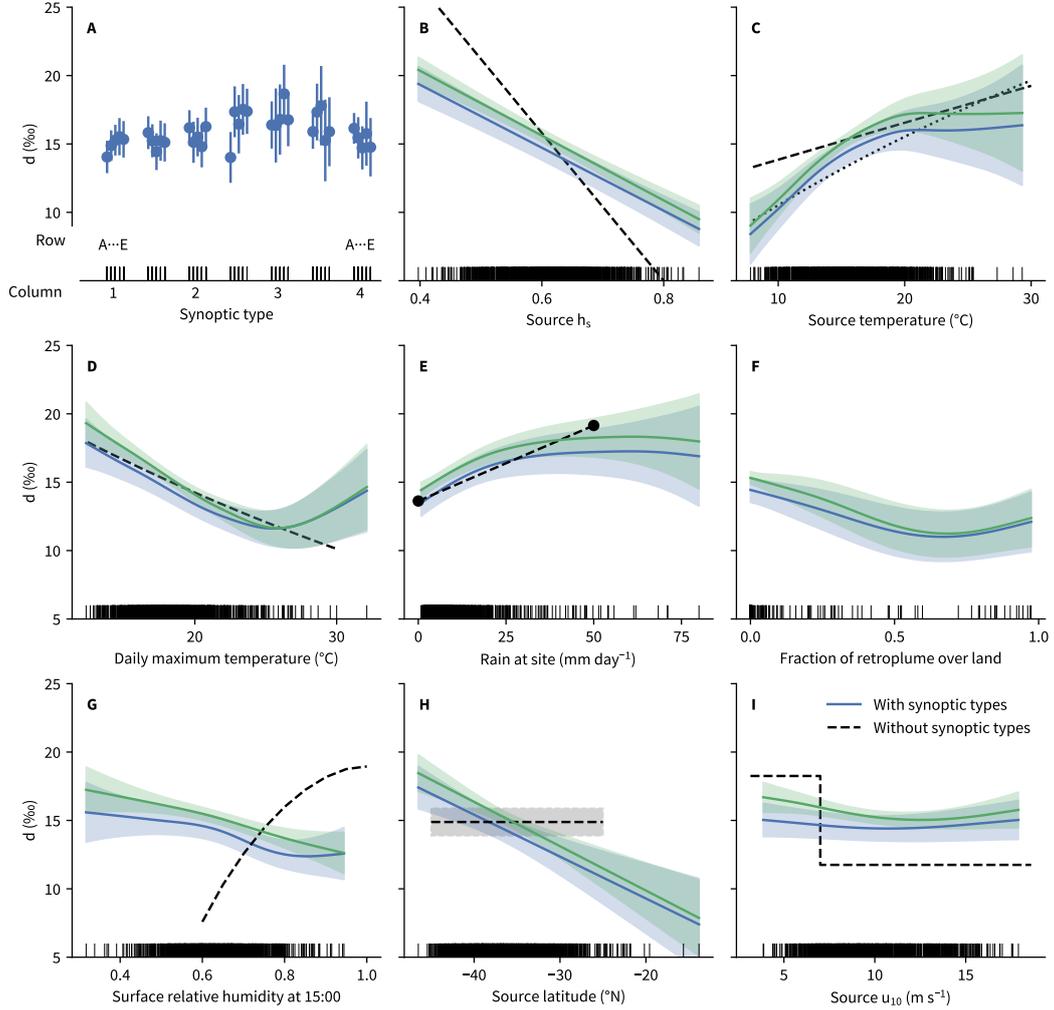
521 rainfall  $d$ , even though it is largely successful at reproducing the seasonal cycle. The GAM  
 522 predictions start above the observations and then are biased low by the end of the ob-  
 523 servation period (model residuals are shown more clearly in Fig. S4). An explanation  
 524 for this apparent contradiction is that there is a missing term which is correlated with  
 525 both  $h_s$  and  $d$  at the annual-mean timescale.

## 526 5 Discussion

### 527 5.1 Physical processes driving $\delta^{18}\text{O}$

528 The predictor variables with the strongest link to rainfall  $\delta^{18}\text{O}$  were rainfall inten-  
 529 sity, observed at the site,  $P$ , and rainfall intensity modelled along the backwards plume,  
 530  $\bar{P}$ . These two predictors are only moderately correlated ( $R = 0.30$ , 95% CI [0.25, 0.35])  
 531 meaning that they are statistically different enough to represent different underlying pro-  
 532 cesses, and yet they are conceptually similar enough to be driven by a single process. If  
 533 both  $P$  and  $\bar{P}$  are driven by the same process, this is likely a modified version of Rayleigh  
 534 distillation (Eriksson, 1965).

535 During idealized Rayleigh distillation, an air mass is continually cooled, condensate  
 536 forms in isotopic equilibrium with the vapor, this condensate is immediately removed  
 537 from the system by rainout, and  $\delta^{18}\text{O}$  of the remaining vapor can be expressed as func-  
 538 tion of the fraction of remaining moisture. But in the case of coastal rainfall, the sys-  
 539 tem departs from the ideal in several ways. It is possible for moisture to be continually  
 540 renewed by evaporation from the ocean (Moore et al., 2014), rainfall is not instantaneously  
 541 removed allowing for partial evaporation of rainfall below the cloud base and recycling  
 542 of moisture (Lee & Fung, 2008), and there is three-dimensional transport within synop-  
 543 tic systems which differs from the idealized model (Dütsch et al., 2016). Although the



**Figure 9.** Smooth terms for GAM predicting daily  $d$ . Other relationships shown are: **B** empirical  $h_s$  relationship (Pfahl & Sodemann, 2014); **C** empirical  $T_s$  relationship (Bonne et al., 2019, dashes) and the effect of the temperature dependence of equilibrium fractionation factors (Horita et al., 2008, dots); **D** the effect of the temperature dependence of equilibrium fractionation factors applied to raindrops (Horita et al., 2008); **E** Xia and Winnick (2021) subcloud evaporation model with raindrop diameter at cloud base of 0.6 mm and 2.6 mm (surface temperature 18 °C, surface humidity 60%); **F** Xia and Winnick (2021) subcloud evaporation model humidity dependence (raindrop diameter 2.1 mm, surface temperature 18 °C); **H** indicative range of  $d$  observed in surface waters in the Atlantic Ocean (Bonne et al., 2019) and modelled in the Indian Ocean (Xu et al., 2012); **I** parameterization from Merlivat and Jouzel (1979) at  $h_s = 0.6$  (Benetti et al., 2014).

544 observations are not comprehensive enough to draw strong conclusions, they are consis-  
 545 tent with the interpretation that  $P$  is correlated with past rainout, and hence Rayleigh-  
 546 type processes, as well as being directly indicative of the importance of local post-condensation  
 547 processes. The preferential rainout of heavy isotopes upstream of Calgardup Cave is closely  
 548 related to  $\bar{P}$ , but this parameter is only available via model output and therefore has a  
 549 large error. The fact that  $\bar{P}$  has predictive power, almost as much as  $P$  at the interan-  
 550 nual timescale despite being derived from model output, indicates that past rainfall is  
 551 important.

552 Another way of considering the role of Rayleigh distillation is through the so-called  
 553 continental effect (Winnick et al., 2014) which often appears as an important term driv-  
 554 ing  $\delta^{18}\text{O}$  (Good et al., 2014, e.g.). Here, the fraction of the backwards plume over land,  
 555  $f_l$  (calculated after 3 h of travel), shows that airmasses which have spent more time over  
 556 land have lower  $\delta^{18}\text{O}$ , meaning that the trend in our results is consistent with isotopic  
 557 depletion driven by rainout. On the whole,  $f_l$  is of only minor importance because the  
 558 vast majority of trajectories do not pass over land before arriving at the rainfall site. Just  
 559 over 90% of backwards plumes spend less than 0.1% of their time over land within 12 h  
 560 of arrival. Because of the lack of overland trajectories in the data, it is unlikely that the  
 561 GAM has been able to learn an accurate relationship, or be able to generalize well to in-  
 562 land sites, but the presence of a relationship between  $\delta^{18}\text{O}$  and  $f_l$  indicates that rain-  
 563 out is able to drive depletion, making it likely that this process plays a role in the sen-  
 564 sitivity of  $\delta^{18}\text{O}$  to rainfall intensity.

565 This sensitivity to rainfall intensity acts on a timescale of individual storms. When  
 566 aggregated from daily to monthly precipitation-weighted values, rainfall intensity has a  
 567 stronger association with  $\delta^{18}\text{O}$  than does total monthly precipitation. This is in agree-  
 568 ment with Fischer and Treble (2008) who studied monthly  $\delta^{18}\text{O}$  data from Perth and  
 569 a short record of daily measurements from Cape Leeuwin. Also similar is that Fischer  
 570 and Treble (2008) found a nonlinear relationship between precipitation and  $\delta^{18}\text{O}$ , using  
 571  $\delta^{18}\text{O} \propto P^{\frac{1}{2}}$ . In our data set, due to scatter,  $\delta^{18}\text{O} \propto P^{\frac{1}{2}}$  fits the data almost as well  
 572 as  $\delta^{18}\text{O} \propto \log(P)$ , and we plot the log form mainly out of preference because of its ap-  
 573 pearance in Rayleigh distillation and also the use of a log transformation when  $\delta^{18}\text{O}$  is  
 574 regressed against moisture residence time,  $\tau$ . Aggarwal et al. (2012) found that  $\delta^{18}\text{O} \propto$   
 575  $\tau = \log(Q/P)$  where  $Q$  is the total column water vapor and  $P$  is the long-term mean  
 576 precipitation rate. In our data, variability in  $Q$  is small enough that  $\log(P/P_0)$  is strongly  
 577 correlated with  $\tau$  so there is no advantage in changing variables to  $\tau$  ( $R = -0.94$ , 95%  
 578 CI  $[-0.95, -0.93]$ ,  $Q$  from ERA-Interim).

579 Besides processes which happen during rainfall or in-transit, the properties of source  
 580 moisture are also potential drivers of variability, and some of these are identified by the  
 581 GAM. Source humidity,  $h_s$  affects  $\delta^{18}\text{O}$  through kinetic fractionation. The relationship  
 582 determined by the GAM is similar to the expression for kinetic fraction used by Benetti  
 583 et al. (2014), as shown by the dashed line in Fig. 6d. In contrast, the relationship be-  
 584 tween latitude and  $\delta^{18}\text{O}$  (Fig. 6e) does not follow the meridional variation in Indian Ocean  
 585 surface water  $\delta^{18}\text{O}$  (LeGrande & Schmidt, 2006). Fischer and Treble (2008) also reported  
 586 a difference in  $\delta^{18}\text{O}$  between airmasses travelling equatorward or poleward, but our re-  
 587 sults suggest that isotopic differences in the source waters are not responsible, meaning  
 588 that perhaps it is the atmospheric  $\delta^{18}\text{O}$  values at the beginning of the backwards plume  
 589 which is important. This is plausible because of a strong and persistent meridional gra-  
 590 dient in mean atmospheric  $\delta^{18}\text{O}$ , with higher values towards the pole, which is a large  
 591 driver of isotopic variability in idealized simulations (Dütsch et al., 2016). There are also  
 592 several co-varying parameters which may obfuscate the direct effect of source water  $\delta^{18}\text{O}$ ;  
 593 latitude is strongly correlated with the oceanic source temperature ( $R = 0.91$  95% CI  
 594  $[0.9, 0.92]$ ), wind speed ( $R = -0.61$  95% CI  $[-0.64, -0.57]$ ), and humidity ( $R = 0.34$   
 595 95% CI  $[0.28, 0.39]$ ).

596 Also present in the GAM is the evaporation-weighted sea surface temperature,  $T_s$ .  
 597 As indicated by the dashed line in Fig. 6g, this term is consistent with the temperature  
 598 dependence of equilibrium fractionation of water vapor from the ocean surface (Majoube,  
 599 1971; Horita & Wesolowski, 1994). Fortuitously, and despite the presence of latitude in  
 600 the model, the smooth term in the GAM matches the slope of the theoretical relation-  
 601 ship very well.

## 602 5.2 Physical processes driving deuterium excess

603 The strongest predictor of daily  $d$  is the source humidity,  $h_s$ , although the relation-  
 604 ship between  $d$  and  $h_s$  shows a lower slope ( $-30\%$ ) than seen in studies of water vapor;  
 605 the dashed line in Fig. 9b shows a typical slope of  $-54\%$  (Pfahl & Sodemann, 2014).  
 606 There are three potential explanations for this. First, this difference may be due to un-  
 607 certainty in the  $h_s$  estimate. The standard deviation of the difference between FLEX-  
 608 PART and FLEXPART-WRF derived values, accounting for part of the uncertainty, is  
 609 0.04 which is large enough, based on tests with synthetic data, to reduce the slope of the  
 610 line of best fit. Second, low humidity air during rainfall (small  $h$ ) causes strong re-evaporation  
 611 of rainfall (Risi et al., 2008). At this coastal site,  $h$  is moderately correlated with  $h_s$  (us-  
 612 ing modelled  $h$ , since  $h_s$  is model-derived,  $R = 0.31$  95% CI [0.26, 0.36]), so the two ef-  
 613 fects together act to reduce the observed slope between  $h_s$  and  $d$ . Third, the slope be-  
 614 tween  $d$  and  $h_s$  may be a genuine trait of the source region. Steen-Larsen et al. (2014)  
 615 report a flatter slope for the  $d \sim h_s$  relationship, with a slope of  $-42.6\%$ , and Aemisegger  
 616 and Sjolte (2018) demonstrate the  $d \sim h_s$  slope varies by region. Even accounting for  
 617 regional variation however,  $-30\%$  is sufficiently outside the range of other observations  
 618 that a combination of the other factors too,  $h_s$  uncertainty or  $h \sim h_s$  correlation, is likely  
 619 to be important.

620 The effect of other sea surface parameters, temperature,  $T_s$ , and wind speed,  $u_{10}$ ,  
 621 have been investigated in the past and their importance is still debated. Uemura et al.  
 622 (2008) reported a positive correlation between  $d$  and  $T_s$  in field measurements, in agree-  
 623 ment with Bonne et al. (2019), whereas Pfahl and Sodemann (2014) argue that the  $T_s$   
 624 is of minor importance compared with  $h_s$ . Figure 9d shows that our data do indicate a  
 625 positive correlation between  $d$  and  $T_s$  for  $T_s < 20^\circ\text{C}$ . The relationship between  $u_{10}$  and  
 626  $d$  is weak in the GAM (Fig. 9i), and arguably inconsistent with the Merlivat (1978a) re-  
 627 lationship, in which kinetic fractionation, and hence  $d$  in evaporation, is lower at high  
 628 wind speeds. In their parameterization, low wind speeds below about  $7\text{ ms}^{-1}$  correspond  
 629 to a smooth regime (and higher  $d$ ) whereas high wind speeds are modelled by a rough  
 630 regime (with lower  $d$ ) (Merlivat & Jouzel, 1979). The  $u_{10}$  relationship here is too weak  
 631 to match the parameterization, and it weakens further when synoptic types are included.  
 632 These findings are in agreement with other recent studies which have found that the Merlivat  
 633 (1978a) parameterization is not directly applicable to field observations. Benetti et al.  
 634 (2014) present data which lies between the rough and smooth regimes, Steen-Larsen et  
 635 al. (2014) find no statistical difference in  $d$  in low versus high winds, and Bonne et al.  
 636 (2019) also find there to be no effect on  $d$  from wind speed, with their data being best  
 637 explained by the rough regime of the Merlivat and Jouzel (1979) model. Considered in  
 638 the context of these other studies, then, the existence of a strong relationship between  
 639  $d$  and  $u_{10}$  seems unlikely.

640 Variability in  $d$  is also driven by the latitude of origin, which may either be linked  
 641 to meridional variations in oceanic  $d$  or meridional variations in atmospheric  $d$ . Figure 9h  
 642 shows that the change in  $d$  with latitude is much larger than observed and modelled vari-  
 643 ations in surface waters, meaning that atmospheric processes are more likely to be re-  
 644 sponsible.

645 Besides the conditions at the moisture source, a second driver of  $d$  is the post-condensation  
 646 re-evaporation of droplets in the subcloud layer. The re-evaporation model of Xia and

647 Winnick (2021) is used for comparison with the GAM smooth terms, showing that the  
 648 increase in  $d$  as a function of rainfall intensity is consistent with subcloud reevaporation.  
 649 The model is a reasonable match to the smooth term when physically reasonable rain-  
 650 drop diameters of 0.6 and 2.6 mm are assumed for rain rates of 2 and 50 mm day<sup>-1</sup> (Fig. 9e).  
 651 In contrast, a calculation using the same model of the relationship between  $d$  and sur-  
 652 face humidity (based on humidity at the surface measured at 1500 local time) does not  
 653 even match the sign of the relationship between  $d$  and (Fig. 9g). This is likely to be the  
 654 result of the predictor variable, based on dewpoint temperature at 1500 local time, be-  
 655 ing a poor stand in for the humidity which is actually experienced during rainfall, which  
 656 happens at an unknown time each day and over a relatively deep atmospheric layer be-  
 657 tween the cloud base and surface.

### 658 5.3 Generalizability of the model

659 As a test of the model’s performance away from the observation site, predictions  
 660 of  $\delta^{18}\text{O}$  for Perth Airport were computed based on observed daily rainfall and FLEX-  
 661 PART backwards plumes terminating at Perth Airport on each rain day. Only  $\delta^{18}\text{O}$  was  
 662 analyzed in detail, since the model was unable to reproduce interannual variations in  $d$   
 663 over the monitoring period at Calgardup Cave. GAM predictions were clipped to the  
 664 range of observations to prevent extrapolation errors. In particular, on days with less  
 665 than 2 mm of rainfall  $\delta^{18}\text{O}$  was set to the same value as if 2 mm of rainfall was observed.  
 666 This was necessary because many of the monthly accumulations included a nontrivial  
 667 contribution from days with light rainfall.

668 When compared with monthly  $\delta^{18}\text{O}$  observations, the GAM performed well dur-  
 669 ing the wet months but had large errors during the dry months (Fig. S5). On some oc-  
 670 casions, this was because of highly depleted rainfall sourced from the ocean off the north-  
 671 west coast of Western Australia which had made a long transit over land. In general, the  
 672 failure of the model to perform well during the summer months can be attributed to a  
 673 lack of summer rainfall in the training data. The stronger influence of tropical processes  
 674 in summer, on Perth rainfall, may also play a role.

675 When aggregated to annual data, the poor performance during dry months becomes  
 676 inconsequential and the model generalizes well; performance in Perth shows a similar pre-  
 677 dictive skill to Calgardup Cave (Fig. S6). Furthermore, the GAM is able to reproduce  
 678 the offset in mean  $\delta^{18}\text{O}$  observed between Perth and Calgardup Cave. To reproduce the  
 679 offset, the model needs to include rainfall intensity, rainfall along the backwards plume,  
 680 source humidity, and source latitude. In particular, the difference in rainfall intensity be-  
 681 tween Perth and Calgardup Cave is only responsible for about 10% of the offset. The  
 682 good performance of the model for the Perth observations makes it likely to be suitable  
 683 for the interpretation of longer-term data from the coastal zone between Calgardup Cave  
 684 and Perth.

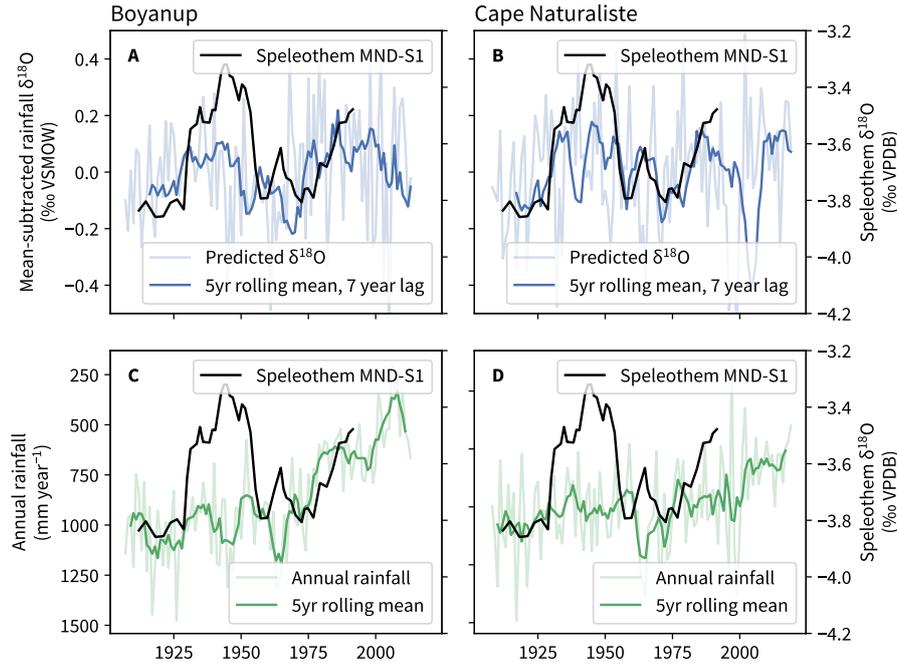
685 For deuterium excess, GAM predictions at Perth Airport show a similar error to  
 686 the Calgardup Cave timeseries tending to have a low bias at the start of the observation  
 687 period and a high bias towards the end.

### 688 5.4 Interpretation of water isotopes in paleoclimate studies

689 Based on data from the 13 year observing period, this study confirms that rain-  
 690 fall intensity is a primary driver of  $\delta^{18}\text{O}$  in precipitation. The nonlinear relationship can  
 691 be approximated as

$$692 \delta^{18}\text{O} = \begin{cases} \alpha \log(P/P_0) + \beta, & P \geq 2 \text{ mm day}^{-1} \\ -2.05\text{‰}, & P < 2 \text{ mm day}^{-1}, \end{cases} \quad (3)$$

693 where  $P_0 = 1 \text{ mm day}^{-1}$ ,  $\alpha = -2.85\text{‰}$ , and  $\beta = -1.19\text{‰}$ . Importantly, years with  
 more intense rainfall are not necessarily wetter overall. In our data, rainfall intensity (pre-



**Figure 10.** Speleothem MND-S1 from Moondyne Cave  $\delta^{18}\text{O}$  (Treble, Chappell, et al., 2005) compared with **A** rainfall  $\delta^{18}\text{O}$  inferred from Boyanup rainfall intensity, **B** Boyanup annual rainfall, **C** rainfall  $\delta^{18}\text{O}$  inferred from Cape Naturaliste rainfall intensity, **D** Cape Naturaliste annual rainfall. A lag of 7 yr and smoothing with a 5 yr rolling mean has been applied to the inferred  $\delta^{18}\text{O}$  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  $\delta^{18}\text{O}$  values.

694 precipitation weighted) has no significant correlation with annual rainfall ( $R = 0.23$ , 95%  
695 CI  $[-0.37, 0.69]$ ).

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

705 Although taking only the leading predictor into account, rainfall  $\delta^{18}\text{O}$  inferred from  
706 the Boyanup record displays an intriguing similarity to the Moondyne Cave record, par-  
707 ticularly the period of relatively higher speleothem  $\delta^{18}\text{O}$  from 1930–55 and the upwards  
708 shift from the mid 1970s. There is also a marked similarity when rainfall intensity is taken  
709 from the Cape Naturaliste record, although with a divergence during the 1930–55 pe-  
710 riod. The disagreement which remains may be the result of nonlinear filtering caused by  
711 karst hydrological processes, which has only been accounted for crudely here by a com-  
712 bination of temporal averaging and introducing a time lag. Indeed, the time lag, of 7 years,  
713 is longer than suggested by the field evidence which perhaps indicates that uncertain-

714 ties in the chronology play a role (Nagra et al., 2016, approx. 5 yr). Another complica-  
 715 tion is that changes in rainfall intensity, inferred from the instrumental record (Philip  
 716 & Yu, 2020), are not spatially smooth and, as demonstrated in Fig. 3, even at the annual  
 717 scale the  $\delta^{18}\text{O}$  timeseries is sensitive to the heaviest events which would impact sites  
 718 differently, even over short spatial scales.

719 Supporting the interpretation that rainfall intensity is key to determining  $\delta^{18}\text{O}$ , on  
 720 daily through to decadal timescales, the trends in annual rainfall accumulations show  
 721 a weaker relationship with  $\delta^{18}\text{O}$  (Fig. 10c and 10d). Post 1970, for the Boyanup hind-  
 722 cast, a drying trend coincides with an upwards shift in speleothem  $\delta^{18}\text{O}$ . This may be  
 723 a sign that the interaction between karst hydrology and  $\delta^{18}\text{O}$  changes as the system dries  
 724 out, but needs detailed investigation before making firm conclusions. A sustained change  
 725 in intense rainfall events could be further amplified by karst flowpaths as intense rain-  
 726 fall events are likely to be more effective at initiating recharge of karst stores (Treble et  
 727 al., 2013).

728 In the case of deuterium excess, the interpretation of multidecadal records in this  
 729 region continues to be hampered by an incomplete understanding of governing processes.  
 730 The strongest predictor on a daily scale, source humidity, makes model predictions worse  
 731 on an interannual scale. Out of the predictors that we considered, rainfall intensity, mea-  
 732 sured at the collection site but not along the backwards plume, has the strongest effect  
 733 on  $d$ . This driver is consistent with subcloud re-evaporation being important for driv-  
 734 ing interannual variability and, if this relationship holds over longer timeseries, it would  
 735 drive an anticorrelation between  $d$  and  $\delta^{18}\text{O}$ . Such an anticorrelation was indeed reported  
 736 by Priestley et al. (2020), in a 35 ka groundwater record, which indicates that the ob-  
 737 served relationships between  $\delta^{18}\text{O}$ ,  $d$ , and  $P$  may also be present over much longer time  
 738 scales.

## 739 6 Conclusions

740 Water isotopes in precipitation were measured daily over thirteen years (2006–2018).  
 741 Daily variability was found to be superimposed on weaker low-frequency trends which  
 742 were driven by anomalous conditions in the first three years of monitoring:  $\delta^{18}\text{O}$  decreases  
 743 by  $0.06 \pm 0.03 \text{‰yr}^{-1}$  and  $d$  increases by  $0.24 \pm 0.07 \text{‰yr}^{-1}$ , and trends tend to weaken  
 744 or reverse in the second half of the monitoring period. The factors which drive  $\delta^{18}\text{O}$  and  
 745  $d$  variability, on a range of timescales, were investigated using generalized additive mod-  
 746 els (GAMs), with upstream conditions diagnosed with backwards dispersion modelling  
 747 and synoptic types determined using a statistical method. Although water isotopes demon-  
 748 strated an association with synoptic types, these were ultimately not a strong driver of  
 749 variability because, we infer, the synoptic types contained redundant information which  
 750 was better expressed by continuous values derived from backwards-plume diagnostics.

751 Daily variability in  $\delta^{18}\text{O}$  was driven primarily by rainfall intensity, both at the mea-  
 752 surement site and upstream, in agreement with the main finding of Fischer and Treble  
 753 (2008), which was based on a smaller data set. The  $\delta^{18}\text{O}$  seasonal cycle was driven by  
 754 seasonal changes in both rainfall intensity and source humidity. The relationship between  
 755 rainfall intensity, at a daily scale, and  $\delta^{18}\text{O}$  was robust. It applied at both the primary  
 756 measurement station, Calgardup Cave, and to monthly accumulations from Perth Air-  
 757 port. The relationship also appears to be robust over longer time periods, as shown by  
 758 projecting the  $\delta^{18}\text{O} \propto \log(P/P_0)$  relationship back through the  $\sim 100$  yr period with  
 759 rainfall observations and comparing to a speleothem record. Because of the relationship  
 760 between rainfall intensity and  $\delta^{18}\text{O}$ , annual accumulations of  $\delta^{18}\text{O}$  are more sensitive to  
 761 the heaviest rainfall events each year than annual accumulated rainfall is, which has im-  
 762 plications both for the interpretation of  $\delta^{18}\text{O}$  records and for how much nearby sites can  
 763 be expected to agree with each other.

764 The behavior of  $d$  differed from  $\delta^{18}\text{O}$  in several ways. On a daily scale, variability  
 765 was driven primarily by  $h_s$ , although with a flatter slope than reported in studies of  
 766 water vapor. The  $d$  seasonal cycle was also well explained mainly by  $h_s$ , with a weaker  
 767 contribution from rainfall intensity. In contrast, year-to-year changes in  $h_s$  failed to ex-  
 768 plain the interannual signal in precipitation-weighted annual mean  $d$ , with the implica-  
 769 tion that multidecadal, or longer, records of  $d$  should not be interpreted as a straight-  
 770 forward proxy record of  $h_s$  in this region. Furthermore, the link between rainfall inten-  
 771 sity and  $d$  was too weak to drive the observed changes in  $d$ , meaning that the driver for  
 772 low-frequency changes in  $d$  was not fully explained. Further investigation of  $d$  is warranted  
 773 because  $d$  has other desirable properties;  $d$  is not as sensitive as  $\delta^{18}\text{O}$  to extreme events,  
 774 and there is a low-frequency signal in the observations at both Calgardup Caves and Perth  
 775 which may be climate-related; meaning that the  $d$  signal carries information which sup-  
 776 plements  $\delta^{18}\text{O}$ .

## 777 7 Acknowledgements

778 The outcomes of this study contribute to ARC DP200100203 awarded to PCT. Wa-  
 779 ter isotope measurements were undertaken at ANSTO and simulations were carried out  
 780 at ANSTO's High Performance Computing facility. We gratefully acknowledge the as-  
 781 sistance from the staff at Calgardup Caves (Department of Biodiversity, Conservations  
 782 and Attractions) to conduct the cave monitoring. IsoGSM data which appears in this  
 783 article was provided thanks to Kei Yoshimura. This paper has benefited from the insight-  
 784 ful comments of three anonymous reviewers whose contribution is appreciated. We also  
 785 acknowledge the open-source software used in this project, especially: R; mgcv; Python;  
 786 Pandas; SOM-PAK; and QGIS.

## 787 8 Data availability

788 The water isotope measurements from Calgardup Cave are available from the IAEA  
 789 Water Isotope System for data analysis, visualization and Electronic Retrieval, [https://  
 790 nucleus.iaea.org/wiser/](https://nucleus.iaea.org/wiser/) using station code 9564101. Water isotopes measured at Perth  
 791 Airport are available from <https://openscience.ansto.gov.au/collection/881>. The  
 792 models used in this study are available for download, FLEXPART and FLEXPART-WRF  
 793 from <http://www.flexpart.eu/> and WRF from <http://www.mmm.ucar.edu/wrf/users/>.  
 794 Reanalysis data are archived by the European Centre for Medium Range Weather Fore-  
 795 casting (ERA-Interim, [https://www.ecmwf.int/en/forecasts/datasets/reanalysis  
 796 -datasets/era-interim](https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era-interim)) and the National Oceanic and Atmospheric Administration  
 797 (CFSR, [https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/climate  
 798 -forecast-system-version2-cfsv2](https://www.ncdc.noaa.gov/data-access/model-data/model-datasets/climate-forecast-system-version2-cfsv2)). Weather station and gridded climate data are  
 799 available from the Australian Bureau of Meteorology, <http://www.bom.gov.au>.

## 800 References

- 801 Aemisegger, F., & Sjolte, J. (2018). A Climatology of Strong Large-Scale  
 802 Ocean Evaporation Events. Part II: Relevance for the Deuterium Excess  
 803 Signature of the Evaporation Flux. *J. Climate*, *31*(18), 7313–7336. doi:  
 804 10.1175/JCLI-D-17-0592.1
- 805 Aggarwal, P. K., Alduchov, O. A., Froehlich, K. O., Araguas-Araguas, L. J., Stur-  
 806 chio, N. C., & Kurita, N. (2012). Stable isotopes in global precipitation: A  
 807 unified interpretation based on atmospheric moisture residence time. *Geophys.*  
 808 *Res. Lett.*, *39*(11). doi: 10.1029/2012GL051937
- 809 Aggarwal, P. K., Romatschke, U., Araguas-Araguas, L., Belachew, D., Longstaffe,  
 810 F. J., Berg, P., ... Funk, A. (2016). Proportions of convective and stratiform  
 811 precipitation revealed in water isotope ratios. *Nature Geosci.*, *9*(8), 624–629.  
 812 doi: 10.1038/ngeo2739

- 813 Baldini, L. M., McDermott, F., Baldini, J. U. L., Fischer, M. J., & Möllhoff, M.  
814 (2010). An investigation of the controls on Irish precipitation  $\delta^{18}\text{O}$  val-  
815 ues on monthly and event timescales. *Clim Dyn*, *35*(6), 977–993. doi:  
816 10.1007/s00382-010-0774-6
- 817 Barras, V., & Simmonds, I. (2008). Synoptic controls upon  $\delta^{18}\text{O}$  in southern Tasma-  
818 nian precipitation. *Geophys. Res. Lett.*, *35*(2). doi: 10.1029/2007GL031835
- 819 Barras, V., & Simmonds, I. (2009). Observation and modeling of stable water iso-  
820 topes as diagnostics of rainfall dynamics over southeastern Australia. *J. Geo-*  
821 *phys. Res. Atmospheres*, *114*(D23), D23308. doi: 10.1029/2009JD012132
- 822 Bates, B. C., Hope, P., Ryan, B., Smith, I., & Charles, S. (2008). Key findings from  
823 the Indian Ocean Climate Initiative and their impact on policy development in  
824 Australia. *Clim. Change*, *89*(3), 339–354.
- 825 Benetti, M., Reverdin, G., Pierre, C., Merlivat, L., Risi, C., Steen-Larsen, H. C., &  
826 Vimeux, F. (2014). Deuterium excess in marine water vapor: Dependency on  
827 relative humidity and surface wind speed during evaporation. *J. Geophys. Res.*  
828 *Atmos.*, *119*(2), 584–593. doi: 10.1002/2013JD020535
- 829 Bonne, J.-L., Behrens, M., Meyer, H., Kipfstuhl, S., Rabe, B., Schönike, L., ...  
830 Werner, M. (2019). Resolving the controls of water vapour isotopes in the  
831 Atlantic sector. *Nat Commun*, *10*(1), 1–10. doi: 10.1038/s41467-019-09242-6
- 832 Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., ...  
833 Wotawa, G. (2013). The Lagrangian particle dispersion model FLEXPART-  
834 WRF version 3.1. *Geosci. Model Dev.*, *6*(6), 1889–1904. doi: 10.5194/  
835 gmd-6-1889-2013
- 836 Brook, E. J., & Buizert, C. (2018). Antarctic and global climate history viewed from  
837 ice cores. *Nature*, *558*(7709), 200. doi: 10.1038/s41586-018-0172-5
- 838 Cai, W., Shi, G., & Li, Y. (2005). Multidecadal fluctuations of winter rainfall over  
839 southwest Western Australia simulated in the CSIRO Mark 3 coupled model.  
840 *Geophys. Res. Lett.*, *32*(12). doi: 10.1029/2005GL022712
- 841 Chen, Z., Auler, A. S., Bakalowicz, M., Drew, D., Griger, F., Hartmann, J., ...  
842 Goldscheider, N. (2017). The World Karst Aquifer Mapping project: Concept,  
843 mapping procedure and map of Europe. *Hydrogeol J*, *25*(3), 771–785. doi:  
844 10.1007/s10040-016-1519-3
- 845 Comas-Bru, L., Rehfeld, K., Roesch, C., Amirnezhad-Mozhdehi, S., Harrison, S. P.,  
846 Atsawawanunt, K., ... SISAL Working Group members (2020). SISALv2: A  
847 comprehensive speleothem isotope database with multiple age–depth models.  
848 *Earth Syst. Sci. Data*, *12*(4), 2579–2606. doi: 10.5194/essd-12-2579-2020
- 849 Craig, H. (1961). Isotopic Variations in Meteoric Waters. *Science*, *133*(3465), 1702–  
850 1703. doi: 10.1126/science.133.3465.1702
- 851 Crawford, J., Hollins, S. E., Meredith, K. T., & Hughes, C. E. (2017). Precipitation  
852 stable isotope variability and subcloud evaporation processes in a semi-arid  
853 region. *Hydrol. Process.*, *31*(1), 20–34. doi: 10.1002/hyp.10885
- 854 Crawford, J., Hughes, C. E., & Lykoudis, S. (2014). Alternative least squares meth-  
855 ods for determining the meteoric water line, demonstrated using GNIP data.  
856 *Journal of Hydrology*, *519*, 2331–2340. doi: 10.1016/j.jhydrol.2014.10.033
- 857 Dansgaard, W. (1964). Stable isotopes in precipitation. *Tellus*, *16*(4), 436–468. doi:  
858 10.3402/tellusa.v16i4.8993
- 859 Dee, D. P., Uppala, S. M., Simmons, A. J., Berrisford, P., Poli, P., Kobayashi, S., ...  
860 Vitart, F. (2011). The ERA-Interim reanalysis: Configuration and performance  
861 of the data assimilation system. *Q.J.R. Meteorol. Soc.*, *137*(656), 553–597. doi:  
862 10.1002/qj.828
- 863 Deininger, M., Werner, M., & McDermott, F. (2016). North Atlantic Oscillation  
864 controls on oxygen and hydrogen isotope gradients in winter precipitation  
865 across Europe; implications for palaeoclimate studies. *Clim. Past*, *12*(11),  
866 2127–2143. doi: 10.5194/cp-12-2127-2016
- 867 Dey, R., Lewis, S. C., Arblaster, J. M., & Abram, N. J. (2019). A review of past and

- 868 projected changes in Australia’s rainfall. *Wiley Interdiscip. Rev. Clim. Change*,  
869 *10*(3), e577. doi: 10.1002/wcc.577
- 870 Dütsch, M., Pfahl, S., & Sodemann, H. (2017). The impact of nonequilibrium and  
871 equilibrium fractionation on two different deuterium excess definitions. *J. Geo-*  
872 *phys. Res. Atmospheres*, *122*(23), 12,732–12,746. doi: 10.1002/2017JD027085
- 873 Dütsch, M., Pfahl, S., & Wernli, H. (2016). Drivers of  $\delta^2\text{H}$  variations in an idealized  
874 extratropical cyclone. *Geophys. Res. Lett.*, *43*(10), 5401–5408. doi: 10.1002/  
875 2016GL068600
- 876 England, M. H., Ummenhofer, C. C., & Santoso, A. (2006). Interannual rainfall  
877 extremes over southwest Western Australia linked to Indian Ocean climate  
878 variability. *J. Climate*, *19*(10), 1948–1969. doi: 10.1175/JCLI3700.1
- 879 Eriksson, E. (1965). Deuterium and oxygen-18 in precipitation and other natural  
880 waters Some theoretical considerations1. *Tellus*, *17*(4), 498–512. doi: 10.1111/  
881 j.2153-3490.1965.tb00212.x
- 882 Farlin, J., Lai, C.-T., & Yoshimura, K. (2013). Influence of synoptic weather  
883 events on the isotopic composition of atmospheric moisture in a coastal city  
884 of the western United States. *Water Resour. Res.*, *49*(6), 3685–3696. doi:  
885 10.1002/wrcr.20305
- 886 Fischer, M. J., & Treble, P. C. (2008). Calibrating climate- $\delta^{18}\text{O}$  regression models  
887 for the interpretation of high-resolution speleothem  $\delta^{18}\text{O}$  time series. *J. Geo-*  
888 *phys. Res.-Atmospheres*, *113*(D17). doi: 10.1029/2007JD009694
- 889 Fohlmeister, J., Schröder-Ritzrau, A., Scholz, D., Spötl, C., Riechelmann, D. F. C.,  
890 Mudelsee, M., . . . Mangini, A. (2012). Bunker Cave stalagmites: An archive  
891 for central European Holocene climate variability. *Clim. Past*, *8*(5), 1751–  
892 1764. doi: 10.5194/cp-8-1751-2012
- 893 Foley, G. R., & Hanstrum, B. N. (1994). The capture of tropical cyclones by cold  
894 fronts off the west coast of Australia. *Wea. Forecasting*, *9*(4), 577–592. doi: 10  
895 .1175/1520-0434(1994)009(0577:TCOTCB)2.0.CO;2
- 896 Forster, C., Stohl, A., & Seibert, P. (2007). Parameterization of convective trans-  
897 port in a Lagrangian particle dispersion model and its evaluation. *J. Appl. Me-*  
898 *teorol. Climatol.*, *46*(4), 403–422. doi: 10.1175/JAM2470.1
- 899 Frederiksen, J. S., & Frederiksen, C. S. (2007). Interdecadal changes in southern  
900 hemisphere winter storm track modes. *Tellus Dyn. Meteorol. Oceanogr.*, *59*(5),  
901 599–617. doi: 10.1111/j.1600-0870.2007.00264.x
- 902 Geoscience Australia. (2012). *Surface Geology of Australia, 1:1 000 000 scale, 2012*  
903 *edition*. <http://data.bioregionalassessments.gov.au/dataset/8284767e-b5b1-4d8b-b8e6-b334fa972611>.
- 904 Geoscience Australia, & Australian Stratigraphy Commission. (2017). *Australian*  
905 *Stratigraphic Units Database*. [http://www.ga.gov.au/products-services/data-](http://www.ga.gov.au/products-services/data-applications/reference-databases/stratigraphic-units.html)  
906 [applications/reference-databases/stratigraphic-units.html](http://www.ga.gov.au/products-services/data-applications/reference-databases/stratigraphic-units.html).
- 907 Good, S. P., Mallia, D. V., Lin, J. C., & Bowen, G. J. (2014). Stable isotope  
908 analysis of precipitation samples obtained via crowdsourcing reveals the spa-  
909 tiotemporal evolution of Superstorm Sandy. *PLoS ONE*, *9*(3), e91117. doi:  
910 10.1371/journal.pone.0091117
- 911 Guan, H., Zhang, X., Skrzypek, G., Sun, Z., & Xu, X. (2013). Deuterium excess  
912 variations of rainfall events in a coastal area of South Australia and its rela-  
913 tionship with synoptic weather systems and atmospheric moisture sources. *J.*  
914 *Geophys. Res. Atmospheres*, *118*(2), 1123–1138. doi: 10.1002/jgrd.50137
- 915 Haylock, M., & Nicholls, N. (2000). Trends in extreme rainfall indices for an up-  
916 dated high quality data set for Australia, 1910–1998. *Int. J. Climatol.*, *20*(13),  
917 1533–1541. doi: 10.1002/1097-0088(20001115)20:13<1533::AID-JOC586>3.0.CO;  
918 2-J
- 919 Hollins, S. E., Hughes, C. E., Crawford, J., Cendón, D. I., & Meredith, K. T. (2018).  
920 Rainfall isotope variations over the Australian continent – Implications for  
921 hydrology and isoscape applications. *Science of The Total Environment*, *645*,

- 923 630–645. doi: 10.1016/j.scitotenv.2018.07.082
- 924 Hope, P., Drosowsky, W., & Nicholls, N. (2006). Shifts in the synoptic systems in-  
925 fluencing southwest Western Australia. *Clim. Dyn.*, *26*(7-8), 751–764. doi: 10  
926 .1007/s00382-006-0115-y
- 927 Hope, P., Keay, K., Pook, M., Catto, J., Simmonds, I., Mills, G., . . . Berry, G.  
928 (2014). A comparison of automated methods of front recognition for climate  
929 studies: A case study in southwest Western Australia. *Mon. Wea. Rev.*,  
930 *142*(1), 343–363. doi: 10.1175/MWR-D-12-00252.1
- 931 Horita, J., Rozanski, K., & Cohen, S. (2008). Isotope effects in the evaporation  
932 of water: A status report of the Craig-Gordon model. *Isotopes Environ Health  
933 Stud.*, *44*(1), 23–49. doi: 10.1080/10256010801887174
- 934 Horita, J., & Wesolowski, D. J. (1994). Liquid-vapor fractionation of oxygen  
935 and hydrogen isotopes of water from the freezing to the critical temperature.  
936 *Geochimica et Cosmochimica Acta*, *58*(16), 3425–3437.
- 937 Hughes, C. E., & Crawford, J. (2012). A new precipitation weighted method for  
938 determining the meteoric water line for hydrological applications demonstrated  
939 using Australian and global GNIP data. *Journal of Hydrology*, *464–465*,  
940 344–351. doi: 10.1016/j.jhydrol.2012.07.029
- 941 IAEA. (2006). *Reference sheet for international measurement standards* (Tech.  
942 Rep.). Vienna, Austria.
- 943 Jones, D., Wang, W., & Fawcett, R. (2009). High-quality spatial climate data-sets  
944 for Australia. *AMOJ*, *58*(04), 233–248. doi: 10.22499/2.5804.003
- 945 Jouzel, J., Masson-Delmotte, V., Cattani, O., Dreyfus, G., Falourd, S., Hoffmann,  
946 G., . . . Wolff, E. W. (2007). Orbital and Millennial Antarctic Climate  
947 Variability over the Past 800,000 Years. *Science*, *317*(5839), 793–796. doi:  
948 10.1126/science.1141038
- 949 Kohonen, T., Hynninen, J., Kangas, J., & Laaksonen, J. (1996).  
950 *SOM\_PAK: The Self-Organizing Map Program Package*.  
951 [https://www.semanticscholar.org/paper/SOM\\_PAK%3A-](https://www.semanticscholar.org/paper/SOM_PAK%3A-The-Self-Organizing-Map-Program-Package-Kohonen-Hynninen/7bd2bb8319a75d9140fd4c30431c7283a6b25710)  
952 [The-Self-Organizing-Map-Program-Package-Kohonen-](https://www.semanticscholar.org/paper/SOM_PAK%3A-The-Self-Organizing-Map-Program-Package-Kohonen-Hynninen/7bd2bb8319a75d9140fd4c30431c7283a6b25710)  
953 [Hynninen/7bd2bb8319a75d9140fd4c30431c7283a6b25710](https://www.semanticscholar.org/paper/SOM_PAK%3A-The-Self-Organizing-Map-Program-Package-Kohonen-Hynninen/7bd2bb8319a75d9140fd4c30431c7283a6b25710).
- 954 Krklec, K., & Domínguez-Villar, D. (2014). Quantification of the impact of moisture  
955 source regions on the oxygen isotope composition of precipitation over Eagle  
956 Cave, central Spain. *Geochimica et Cosmochimica Acta*, *134*, 39–54. doi:  
957 10.1016/j.gca.2014.03.011
- 958 Lachniet, M. S. (2009). Climatic and environmental controls on speleothem oxygen-  
959 isotope values. *Quaternary Science Reviews*, *28*(5–6), 412–432. doi: 10.1016/j  
960 .quascirev.2008.10.021
- 961 Lavery, B., Joung, G., & Nicholls, N. (1997). An extended high-quality historical  
962 rainfall dataset for Australia. *Australian Meteorological Magazine*, *46*(1), 27–  
963 38.
- 964 Lee, J.-E., & Fung, I. (2008). “Amount effect” of water isotopes and quantitative  
965 analysis of post-condensation processes. *Hydrol. Process.*, *22*(1), 1–8. doi: 10  
966 .1002/hyp.6637
- 967 Lee, J.-E., Fung, I., DePaolo, D. J., & Henning, C. C. (2007). Analysis of the global  
968 distribution of water isotopes using the NCAR atmospheric general circulation  
969 model. *J. Geophys. Res.*, *112*(D16), D16306. doi: 10.1029/2006JD007657
- 970 LeGrande, A. N., & Schmidt, G. A. (2006). Global gridded data set of the oxygen  
971 isotopic composition in seawater. *Geophys. Res. Lett.*, *33*(12). doi: 10.1029/  
972 2006GL026011
- 973 Liu, J., Fu, G., Song, X., Charles, S. P., Zhang, Y., Han, D., & Wang, S. (2010).  
974 Stable isotopic compositions in Australian precipitation. *J. Geophys. Res.*  
975 *Atmospheres*, *115*(D23), D23307. doi: 10.1029/2010JD014403
- 976 Lorrey, A., Williams, P., Salinger, J., Martin, T., Palmer, J., Fowler, A., . . . Neil,  
977 H. (2008). Speleothem stable isotope records interpreted within a multi-proxy

- 978 framework and implications for New Zealand palaeoclimate reconstruction.  
 979 *Quaternary International*, 187(1), 52–75. doi: 10.1016/j.quaint.2007.09.039
- 980 Lucas, C., Rudeva, I., Nguyen, H., Boschat, G., & Hope, P. (2021). Variability  
 981 and changes to the mean meridional circulation in isentropic coordinates. *Clim*  
 982 *Dyn.* doi: 10.1007/s00382-021-05903-9
- 983 Lykoudis, S. P., Kostopoulou, E., & Argiriou, A. A. (2010). Stable isotopic signature  
 984 of precipitation under various synoptic classifications. *Physics and Chemistry*  
 985 *of the Earth, Parts A/B/C*, 35(9), 530–535. doi: 10.1016/j.pce.2009.09.002
- 986 Majoube, M. (1971). Fractionnement en oxygene-18 et en deuterium entre l'eau et  
 987 sa vapeur. *J. Chim. phys.*, 68(10), 1423–1436.
- 988 McCabe-Glynn, S., Johnson, K. R., Strong, C., Berkelhammer, M., Sinha, A.,  
 989 Cheng, H., & Edwards, R. L. (2013). Variable North Pacific influence on  
 990 drought in southwestern North America since AD 854. *Nat. Geosci.*, 6(8),  
 991 617–621. doi: 10.1038/ngeo1862
- 992 Merlivat, L. (1978a). The dependence of bulk evaporation coefficients on air-water  
 993 interfacial conditions as determined by the isotopic method. *J. Geophys. Res.*,  
 994 83(C6), 2977–2980. doi: 10.1029/JC083iC06p02977
- 995 Merlivat, L. (1978b). Molecular diffusivities of H<sub>2</sub><sup>16</sup>O, HD<sup>16</sup>O, and H<sub>2</sub><sup>18</sup>O in gases.  
 996 *The Journal of Chemical Physics*, 69(6), 2864–2871. doi: 10.1063/1.436884
- 997 Merlivat, L., & Jouzel, J. (1979). Global climatic interpretation of the deuterium-  
 998 oxygen 18 relationship for precipitation. *J. Geophys. Res.*, 84(C8), 5029–5033.  
 999 doi: 10.1029/JC084iC08p05029
- 1000 Moerman, J. W., Cobb, K. M., Adkins, J. F., Sodemann, H., Clark, B., & Tuen,  
 1001 A. A. (2013). Diurnal to interannual rainfall  $\delta^{18}\text{O}$  variations in northern  
 1002 Borneo driven by regional hydrology. *Earth and Planetary Science Letters*,  
 1003 369–370, 108–119. doi: 10.1016/j.epsl.2013.03.014
- 1004 Moore, M., Kuang, Z., & Blossey, P. N. (2014). A moisture budget perspective  
 1005 of the amount effect. *Geophys. Res. Lett.*, 41(4), 1329–1335. doi: 10.1002/  
 1006 2013GL058302
- 1007 Nagra, G., Treble, P. C., Andersen, M. S., Fairchild, I. J., Coleborn, K., & Baker, A.  
 1008 (2016). A post-wildfire response in cave dripwater chemistry. *Hydrol. Earth*  
 1009 *Syst. Sci.*, 20(7), 2745–2758. doi: 10.5194/hess-20-2745-2016
- 1010 Noone, D., & Simmonds, I. (2002). Associations between  $\delta^{18}\text{O}$  of Water and Cli-  
 1011 mate Parameters in a Simulation of Atmospheric Circulation for 1979–95.  
 1012 *J. Climate*, 15(22), 3150–3169. doi: 10.1175/1520-0442(2002)015<3150:  
 1013 ABOOWA>2.0.CO;2
- 1014 Okazaki, A., Satoh, Y., Tremoy, G., Vimeux, F., Scheepmaker, R., & Yoshimura,  
 1015 K. (2015). Interannual variability of isotopic composition in water vapor over  
 1016 western Africa and its relationship to ENSO. *Atmos. Chem. Phys.*, 15(6),  
 1017 3193–3204. doi: 10.5194/acp-15-3193-2015
- 1018 Orland, I. J., Bar-Matthews, M., Kita, N. T., Ayalon, A., Matthews, A., & Valley,  
 1019 J. W. (2009). Climate deterioration in the Eastern Mediterranean as revealed  
 1020 by ion microprobe analysis of a speleothem that grew from 2.2 to 0.9 ka in  
 1021 Soreq Cave, Israel. *Quat. Res.*, 71(1), 27–35. doi: 10.1016/j.yqres.2008.08.005
- 1022 Paget, M. J. (2008). *MODIS Land data sets for the Australian region* (Tech. Rep.).  
 1023 Canberra: CSIRO Marine and Atmospheric Research.
- 1024 Pepler, A. S., Dowdy, A. J., van Rensch, P., Rudeva, I., Catto, J. L., & Hope,  
 1025 P. (2020). The contributions of fronts, lows and thunderstorms to south-  
 1026 ern Australian rainfall. *Clim Dyn.*, 55(5), 1489–1505. doi: 10.1007/  
 1027 s00382-020-05338-8
- 1028 Pfahl, S., & Sodemann, H. (2014). What controls deuterium excess in global precipi-  
 1029 tation? *Clim. Past*, 10(2), 771–781. doi: 10.5194/cp-10-771-2014
- 1030 Pfahl, S., & Wernli, H. (2008). Air parcel trajectory analysis of stable isotopes  
 1031 in water vapor in the eastern Mediterranean. *J. Geophys. Res. Atmospheres*,  
 1032 113(D20), D20104. doi: 10.1029/2008JD009839

- 1033 Pfahl, S., & Wernli, H. (2009). Lagrangian simulations of stable isotopes in water  
 1034 vapor: An evaluation of nonequilibrium fractionation in the Craig-Gordon  
 1035 model. *J. Geophys. Res.*, *114*(D20), D20108.
- 1036 Philip, P., & Yu, B. (2020). Interannual variations in rainfall of different intensities  
 1037 in South West of Western Australia. *Int. J. Climatol.*, *40*(6), 3052–3071. doi:  
 1038 10.1002/joc.6382
- 1039 Philipp, A., Beck, C., Huth, R., & Jacobeit, J. (2016). Development and comparison  
 1040 of circulation type classifications using the COST 733 dataset and software.  
 1041 *Int. J. Climatol.*, *36*(7), 2673–2691. doi: 10.1002/joc.3920
- 1042 Pook, M. J., Risbey, J. S., & McIntosh, P. C. (2012). The synoptic climatology of  
 1043 cool-season rainfall in the central wheatbelt of Western Australia. *Mon. Wea.*  
 1044 *Rev.*, *140*(1), 28–43. doi: 10.1175/MWR-D-11-00048.1
- 1045 Power, S., Sadler, B., & Nicholls, N. (2005). The influence of climate science on  
 1046 water management in Western Australia: Lessons for climate scientists. *Bull.*  
 1047 *Amer. Meteor. Soc.*, *86*(6), 839–844. doi: 10.1175/BAMS-86-6-839
- 1048 Priestley, S. C., Meredith, K. T., Treble, P. C., Cendón, D. I., Griffiths, A. D.,  
 1049 Hollins, S. E., . . . Pigois, J.-P. (2020). A 35 ka record of groundwater recharge  
 1050 in south-west Australia using stable water isotopes. *Sci. Total Environ.*, *717*,  
 1051 135105. doi: 10.1016/j.scitotenv.2019.135105
- 1052 R Core Team. (2014). *R: A language and environment for statistical computing*  
 1053 (Tech. Rep.). Vienna, Austria.
- 1054 Raut, B. A., Jakob, C., & Reeder, M. J. (2014). Rainfall changes over southwestern  
 1055 Australia and their relationship to the Southern Annular Mode and ENSO. *J.*  
 1056 *Climate*, *27*(15), 5801–5814. doi: 10.1175/JCLI-D-13-00773.1
- 1057 Raut, B. A., Reeder, M. J., & Jakob, C. (2016). Trends in CMIP5 Rainfall Patterns  
 1058 over Southwestern Australia. *J. Climate*, *30*(5), 1779–1788. doi: 10.1175/JCLI  
 1059 -D-16-0584.1
- 1060 Risi, C., Bony, S., & Vimeux, F. (2008). Influence of convective processes on the  
 1061 isotopic composition ( $\delta^{18}\text{O}$  and  $\delta\text{D}$ ) of precipitation and water vapor in the  
 1062 tropics: 2. Physical interpretation of the amount effect. *J. Geophys. Res.*,  
 1063 *113*(D19), D19306. doi: 10.1029/2008JD009943
- 1064 Risi, C., Bony, S., Vimeux, F., & Jouzel, J. (2010). Water-stable isotopes in  
 1065 the LMDZ4 general circulation model: Model evaluation for present-day  
 1066 and past climates and applications to climatic interpretations of tropical  
 1067 isotopic records. *J. Geophys. Res. Atmospheres*, *115*(D12), D12118. doi:  
 1068 10.1029/2009JD013255
- 1069 Risi, C., Noone, D., Worden, J., Frankenberg, C., Stiller, G., Kiefer, M., . . . Sturm,  
 1070 C. (2012). Process-evaluation of tropospheric humidity simulated by general  
 1071 circulation models using water vapor isotopologues: 1. Comparison between  
 1072 models and observations. *J. Geophys. Res. Atmospheres*, *117*(D5). doi:  
 1073 10.1029/2011JD016621
- 1074 Rudeva, I., Simmonds, I., Crock, D., & Boschat, G. (2019). Midlatitude fronts and  
 1075 variability in the Southern Hemisphere tropical width. *J. Clim.*, *32*(23), 8243–  
 1076 8260. doi: 10.1175/JCLI-D-18-0782.1
- 1077 Saha, S., Moorthi, S., Pan, H.-L., Wu, X., Wang, J., Nadiga, S., . . . Goldberg, M.  
 1078 (2010). The NCEP Climate Forecast System Reanalysis. *Bull. Amer. Meteor.*  
 1079 *Soc.*, *91*(8), 1015–1058. doi: 10.1175/2010BAMS3001.1
- 1080 Schemm, S., Rudeva, I., & Simmonds, I. (2015). Extratropical fronts in the lower  
 1081 troposphere—global perspectives obtained from two automated methods. *Q. J.*  
 1082 *R. Meteorol. Soc.*, *141*(690), 1686–1698. doi: 10.1002/qj.2471
- 1083 Schlosser, E., Dittmann, A., Stenni, B., Powers, J. G., Manning, K. W., Masson-  
 1084 Delmotte, V., . . . Scarchilli, C. (2017). The influence of the synoptic regime  
 1085 on stable water isotopes in precipitation at Dome C, East Antarctica. *The*  
 1086 *Cryosphere*, *11*(5), 2345–2361. doi: 10.5194/tc-11-2345-2017
- 1087 Schmidt, G. A., LeGrande, A. N., & Hoffmann, G. (2007). Water isotope expres-

- 1088 sions of intrinsic and forced variability in a coupled ocean-atmosphere model.  
 1089 *J. Geophys. Res.*, *112*(D10), D10103. doi: 10.1029/2006JD007781
- 1090 Scholl, M. A., Shanley, J. B., Zegarra, J. P., & Coplen, T. B. (2009). The stable  
 1091 isotope amount effect: New insights from NEXRAD echo tops, Luquillo Moun-  
 1092 tains, Puerto Rico. *Water Resour. Res.*, *45*(12). doi: 10.1029/2008WR007515
- 1093 Seibert, P., & Frank, A. (2004). Source-receptor matrix calculation with a La-  
 1094 grangian particle dispersion model in backward mode. *Atmos. Chem. Phys.*,  
 1095 *4*(1), 51–63. doi: 10.5194/acp-4-51-2004
- 1096 Simmonds, I., Keay, K., & Tristram Bye, J. A. (2011). Identification and climatol-  
 1097 ogy of Southern Hemisphere mobile fronts in a modern reanalysis. *J. Climate*,  
 1098 *25*(6), 1945–1962. doi: 10.1175/JCLI-D-11-00100.1
- 1099 Skamarock, W. C., & Klemp, J. B. (2008). A time-split nonhydrostatic atmospheric  
 1100 model for weather research and forecasting applications. *Journal of Computa-  
 1101 tional Physics*, *227*(7), 3465–3485. doi: 10.1016/j.jcp.2007.01.037
- 1102 Smith, A., Massuel, S., Pollock, D., & Dillon, P. (2012). Geohydrology of the  
 1103 Tamala Limestone Formation in the Perth Region: Origin and role of sec-  
 1104 ondary porosity.  
 1105 doi: 10.4225/08/599dd0fd98a44
- 1106 Sodemann, H., Aemisegger, F., Pfahl, S., Bitter, M., Corsmeier, U., Feuerle, T., ...  
 1107 Wernli, H. (2017). The stable isotopic composition of water vapour above  
 1108 Corsica during the HyMeX SOP1 campaign: Insight into vertical mixing pro-  
 1109 cesses from lower-tropospheric survey flights. *Atmospheric Chem. Phys.*, *17*(9),  
 1110 6125–6151. doi: 10.5194/acp-17-6125-2017
- 1111 Sodemann, H., Schwierz, C., & Wernli, H. (2008). Interannual variability of Green-  
 1112 land winter precipitation sources: Lagrangian moisture diagnostic and North  
 1113 Atlantic Oscillation influence. *J. Geophys. Res. Atmospheres*, *113*(D3). doi:  
 1114 10.1029/2007JD008503
- 1115 Steen-Larsen, H. C., Sveinbjörnsdóttir, A. E., Peters, A. J., Masson-Delmotte, V.,  
 1116 Guishard, M. P., Hsiao, G., ... White, J. W. C. (2014). Climatic controls  
 1117 on water vapor deuterium excess in the marine boundary layer of the North  
 1118 Atlantic based on 500 days of in situ, continuous measurements. *Atmos. Chem.  
 1119 Phys.*, *14*(15), 7741–7756. doi: 10.5194/acp-14-7741-2014
- 1120 Stohl, A., Eckhardt, S., Forster, C., James, P., Spichtinger, N., & Seibert, P. (2002).  
 1121 A replacement for simple back trajectory calculations in the interpretation  
 1122 of atmospheric trace substance measurements. *Atmospheric Environment*,  
 1123 *36*(29), 4635–4648. doi: 10.1016/S1352-2310(02)00416-8
- 1124 Sturm, K., Hoffmann, G., Langmann, B., & Stichler, W. (2005). Simulation of  $\delta^{18}\text{O}$   
 1125 in precipitation by the regional circulation model REMO\_iso. *Hydrol. Process.*,  
 1126 *19*(17), 3425–3444. doi: 10.1002/hyp.5979
- 1127 Treble, P. C., Bradley, C., Wood, A., Baker, A., Jex, C. N., Fairchild, I. J., ...  
 1128 Azcurra, C. (2013). An isotopic and modelling study of flow paths and stor-  
 1129 age in Quaternary calcarenite, SW Australia: Implications for speleothem  
 1130 paleoclimate records. *Quaternary Science Reviews*, *64*, 90–103. doi:  
 1131 10.1016/j.quascirev.2012.12.015
- 1132 Treble, P. C., Budd, W. F., Hope, P. K., & Rustomji, P. K. (2005). Synoptic-  
 1133 scale climate patterns associated with rainfall  $\delta^{18}\text{O}$  in southern Australia. *J.  
 1134 Hydrol.*, *302*(1-4), 270–282. doi: 10.1016/j.jhydrol.2004.07.003
- 1135 Treble, P. C., Chappell, J., Gagan, M. K., McKeegan, K. D., & Harrison, T. M.  
 1136 (2005). In situ measurement of seasonal  $\delta^{18}\text{O}$  variations and analysis of iso-  
 1137 topic trends in a modern speleothem from southwest Australia. *Earth and  
 1138 Planetary Science Letters*, *233*(1–2), 17–32. doi: 10.1016/j.epsl.2005.02.013
- 1139 Tyler, J. J., Jones, M., Arrowsmith, C., Allott, T., & Leng, M. J. (2016). Spatial  
 1140 patterns in the oxygen isotope composition of daily rainfall in the British Isles.  
 1141 *Clim. Dyn.*, *47*(5-6), 1971–1987. doi: 10.1007/s00382-015-2945-y

- 1142 Uemura, R., Matsui, Y., Yoshimura, K., Motoyama, H., & Yoshida, N. (2008).  
 1143 Evidence of deuterium excess in water vapor as an indicator of ocean sur-  
 1144 face conditions. *J. Geophys. Res. Atmospheres*, *113*(D19), D19114. doi:  
 1145 10.1029/2008JD010209
- 1146 van Breukelen, M. R., Vonhof, H. B., Hellstrom, J. C., Wester, W. C. G., & Kroon,  
 1147 D. (2008). Fossil dripwater in stalagmites reveals Holocene temperature and  
 1148 rainfall variation in Amazonia. *Earth and Planetary Science Letters*, *275*(1),  
 1149 54–60. doi: 10.1016/j.epsl.2008.07.060
- 1150 van Ommen, T. D., & Morgan, V. (2010). Snowfall increase in coastal East Antarc-  
 1151 tica linked with southwest Western Australian drought. *Nat. Geosci.*, *3*(4),  
 1152 267–272. doi: 10.1038/ngeo761
- 1153 Viney, N. R., & Bates, B. C. (2004). It never rains on Sunday: The prevalence  
 1154 and implications of untagged multi-day rainfall accumulations in the Aus-  
 1155 tralian high quality data set. *Int. J. Climatol.*, *24*(9), 1171–1192. doi:  
 1156 10.1002/joc.1053
- 1157 Vonhof, H. B., van Breukelen, M. R., Postma, O., Rowe, P. J., Atkinson, T. C., &  
 1158 Kroon, D. (2006). A continuous-flow crushing device for on-line  $\delta^2\text{H}$  analysis of  
 1159 fluid inclusion water in speleothems. *Rapid Commun. Mass Spectrom.*, *20*(17),  
 1160 2553–2558. doi: 10.1002/rcm.2618
- 1161 Wang, S., Zhang, M., Crawford, J., Hughes, C. E., Du, M., & Liu, X. (2017).  
 1162 The effect of moisture source and synoptic conditions on precipitation iso-  
 1163 topes in arid central Asia. *J. Geophys. Res. Atmos.*, 2015JD024626. doi:  
 1164 10.1002/2015JD024626
- 1165 Werner, M., Langebroek, P. M., Carlsen, T., Herold, M., & Lohmann, G. (2011).  
 1166 Stable water isotopes in the ECHAM5 general circulation model: Toward high-  
 1167 resolution isotope modeling on a global scale. *J. Geophys. Res.*, *116*(D15),  
 1168 D15109. doi: 10.1029/2011JD015681
- 1169 Winnick, M. J., Chamberlain, C. P., Caves, J. K., & Welker, J. M. (2014). Quantify-  
 1170 ing the isotopic ‘continental effect’. *Earth and Planetary Science Letters*, *406*,  
 1171 123–133. doi: 10.1016/j.epsl.2014.09.005
- 1172 Wood, S. N. (2011). Fast stable restricted maximum likelihood and marginal like-  
 1173 lihood estimation of semiparametric generalized linear models. *J. R. Stat. Soc.*  
 1174 *Ser. B Stat. Methodol.*, *73*(1), 3–36. doi: 10.1111/j.1467-9868.2010.00749.x
- 1175 Wood, S. N. (2017). *Generalized additive models: An introduction with R*. Chapman  
 1176 and Hall/CRC.
- 1177 Xia, Z., & Winnick, M. J. (2021). The competing effects of terrestrial evapotran-  
 1178 spiration and raindrop re-evaporation on the deuterium excess of continen-  
 1179 tal precipitation. *Earth and Planetary Science Letters*, *572*, 117120. doi:  
 1180 10.1016/j.epsl.2021.117120
- 1181 Xu, X., Werner, M., Butzin, M., & Lohmann, G. (2012). Water isotope variations in  
 1182 the global ocean model MPI-OM. *Geosci. Model Dev.*, *5*(3), 809–818. doi: 10  
 1183 .5194/gmd-5-809-2012
- 1184 Yoshimura, K., Kanamitsu, M., Noone, D., & Oki, T. (2008). Historical isotope  
 1185 simulation using Reanalysis atmospheric data. *J. Geophys. Res.*, *113*(D19),  
 1186 D19108. doi: 10.1029/2008JD010074
- 1187 Zhang, H., Griffiths, M. L., Chiang, J. C. H., Kong, W., Wu, S., Atwood, A., ...  
 1188 Xie, S. (2018). East Asian hydroclimate modulated by the position of  
 1189 the westerlies during Termination I. *Science*, *362*(6414), 580–583. doi:  
 1190 10.1126/science.aat9393
- 1191 Zhang, Z., Li, G., Yan, H., & An, Z. (2018). Microcodium in Chinese loess as a  
 1192 recorder for the oxygen isotopic composition of monsoonal rainwater. *Quater-  
 1193 nary International*, *464*, 364–369. doi: 10.1016/j.quaint.2017.10.050
- 1194 Zheng, Y., Jong, L. M., Phipps, S. J., Roberts, J. L., Moy, A. D., Curran, M. A. J.,  
 1195 & van Ommen, T. D. (2021). Extending and understanding the South  
 1196 West Western Australian rainfall record using a snowfall reconstruction

1197  
1198

from Law Dome, East Antarctica.  
10.5194/cp-17-1973-2021

*Clim. Past*, 17(5), 1973–1987.

doi: