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- **1** Spatial patterns in the oxygen isotope composition of daily rainfall in the British
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28 Understanding the modern day relationship between climate and the oxygen isotopic composition of precipitation ($\delta^{18}O_P$) is crucial for obtaining rigorous palaeoclimate 29 30 reconstructions from a variety of archives. To date, the majority of empirical studies into the meteorological controls over $\delta^{18}O_P$ rely upon daily, event scale, or monthly 31 32 time series from individual locations, resulting in uncertainties concerning the 33 representativeness of statistical models and the mechanisms behind those 34 relationships. Here, we take an alternative approach by analysing daily patterns in $\delta^{18}O_P$ from multiple stations across the British Isles (n = 10 - 70 stations). We use 35 36 these data to examine the spatial and seasonal heterogeneity of regression statistics between $\delta^{18}O_P$ and common predictors (temperature, precipitation amount and the 37 38 North Atlantic Oscillation index; NAO). Temperature and NAO are poor predictors of daily $\delta^{18}O_P$ in the British Isles, exhibiting weak and/or inconsistent effects both 39 spatially and between seasons. By contrast $\delta^{18}O_P$ and rainfall amount consistently 40 41 correlate at most locations, and for all months analysed, with spatial and temporal 42 variability in the regression coefficients. The maps also allow comparison with daily synoptic weather types, and suggest characteristic $\delta^{18}O_P$ patterns, particularly 43 associated with Cylonic Lamb Weather Types. Mapping daily $\delta^{18}O_P$ across the British 44 Isles therefore provides a more coherent picture of the patterns in $\delta^{18}O_P$, which will 45 ultimately lead to a better understanding of the climatic controls. These observations 46 47 are another step forward towards developing a more detailed, mechanistic framework 48 for interpreting stable isotopes in rainfall as a palaeoclimate and hydrological tracer. 49

50 Keywords: oxygen isotopes, amount effect, NAO, British Isles

51 Introduction

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53 The relationship between climate and the oxygen isotope composition of precipitation $(\delta^{18}O_P)$ is central to a wide range of palaeoclimate interpretation techniques, from 54 55 direct archives of ancient precipitation preserved in ice (e.g. Barbante et al. 2006; 56 Dansgaard et al. 1993; Johnsen et al. 2001; Petit et al. 1999), through indirect archives 57 which include speleothem calcite, lake sediment biominerals and tree ring cellulose 58 (e.g. Baker et al. 2011; Evans and Schrag 2004; Jones et al. 2006; Robertson et al. 59 2001; Tyler et al. 2008; von Grafenstein et al. 1996; Wang et al. 2008). Improved 60 understanding of the climate-isotope relationship is therefore an important step 61 towards improving the accuracy and rigour of palaeoclimate reconstructions both 62 from individual records and through regional/global data-assimilation projects (e.g. 63 PAGES 2k Consortium 2013; Shakun and Carlson 2010). Much of our understanding 64 of this key climate-isotope interaction is built around two approaches: the 65 development and exploration of isotope-enabled climate models (Gedzelman and 66 Arnold 1994; Hoffmann et al. 2006; Jouzel et al. 2000; Langebroek et al. 2011; 67 Merlivat and Jouzel 1979; Schmidt et al. 2007) and the statistical examination of 68 isotope monitoring data (Dansgaard 1964; Fischer and Baldini 2011; Rozanski et al. 69 1993; Treble et al. 2005). Despite significant developments in incorporating isotope 70 systematics into climate models, the continued acquisition and exploration of 71 precipitation isotope monitoring data remains crucial, both to assist in the validation 72 and parameterisation of climate models but also to refine the interpretation of isotope 73 based palaeoclimate records.

Empirical studies into the links between $\delta^{18}O_P$ and climatic/meteorological parameters 75 are numerous and diverse. A widespread correlation between $\delta^{18}O_P$ and air 76 77 temperature is manifest globally and for select continental regions, particularly from a 78 spatial perspective and occasionally through time (Araguas-Araguas et al. 2000; 79 Dansgaard 1964; Kohn and Welker 2005; Rozanski et al. 1993). Air temperature is an 80 important factor in driving condensation within a vapour parcel and dictating the 81 liquid-vapour isotopic fractionation (Dansgaard 1964). However, uncertainties exist 82 regarding the association between air temperature at the land surface and vapour 83 condensation temperature, which varies as a function of altitude even during the 84 course of an individual rainfall event (Celle-Jeanton et al. 2004; Celle-Jeanton et al. 85 2001). Furthermore, the global correlation between air temperature and $\delta^{18}O_P$ is 86 subject to covariance with other key elements of the global isotope hydrological 87 cycle, including latitude, conditions at and distance from evaporation source, and 88 precipitation amount (Bowen and Wilkinson 2002).

89

Changes in precipitation amount are also expected to impart an influence upon $\delta^{18}O_P$, 90 91 due to the combined effects of Rayleigh distillation prior to precipitation at the 92 monitoring station, evaporation from falling raindrops and isotopic exchange between 93 raindrops and ambient vapour beneath cloud level (Callow et al. 2014; Dansgaard 94 1964). The so called 'amount effect' is most prominently associated with convective 95 tropical rainfall (Rozanski et al. 1993), however correlations between rainfall amount and $\delta^{18}O_P$ have also been frequently observed in data from maritime temperate 96 97 regions (Baldini et al. 2010; Baldini et al. 2008; Callow et al. 2014; Celle-Jeanton et 98 al. 2001; Crawford et al. 2013; Darling and Talbot 2003; Treble et al. 2005). In addition to effects at the site of precipitation, variability in $\delta^{18}O_P$ is subject to the 99

conditions at the source of moisture evaporation (e.g. sea surface temperatures and
relative humidity), the trajectory of the air mass, synoptic weather patterns and
interaction with the land surface (Celle-Jeanton et al. 2004; Celle-Jeanton et al. 2001;
Fischer and Baldini 2011; Heathcote and Lloyd 1986; Lachniet and Patterson 2009;
Liebminger et al. 2006; Sodemann et al. 2008; Treble et al. 2005).

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A major advance in our understanding of the controls over $\delta^{18}O_P$ has been the 106 107 proliferation of studies utilising daily or event-scale monitoring in an attempt to 108 address the mechanisms behind isotopic signatures at timescales relevant to the actual 109 process (Baldini et al. 2010; Fischer and Baldini 2011; Heathcote and Lloyd 1986). Longer term $\delta^{18}O_P$ data – be they monthly, annual, centennial or millennial – are best 110 111 viewed as composites of event scale processes, weighted by the amount of rainfall 112 during each event. For this reason, regression models built around monthly or annual 113 data can be subject to issues related to changes in seasonal weighting (Vachon et al. 114 2007) or simply an inability to capture the conditions during which precipitation 115 occurred (Baldini et al. 2010). Recently, approaches have emerged which enable the 116 integration of monthly resolved isotope data with daily meteorological data, therefore 117 modelling the processes at timescales relevant to synoptic conditions (Fischer and 118 Baldini 2011; Fischer and Treble 2008). However, with increased resolution comes 119 increased noise, thus the representativeness of empirical models based upon single 120 isotope time-series comes into question. There is therefore significant value in studies 121 which combine daily monitoring with multiple sites in order to evaluate the 122 relationships between regional meteorology and the isotopic composition of 123 precipitation (e.g. Good et al. 2014) and such studies are scarce.

124

125	The British Isles is an interesting study location for isotopes in rainfall, with a
126	maritime climate that is affected by the confluence of weather systems with distinctly
127	different origins depending on the direction of flow (Heathcote and Lloyd 1986).
128	Previous studies have addressed the climate-isotope relationships in the British Isles
129	using single site daily, event based and monthly monitoring (Baldini et al. 2010;
130	Darling and Talbot 2003; Fischer and Baldini 2011; Heathcote and Lloyd 1986; Jones
131	et al. 2007). On the basis of nearly two year's daily monitoring at Driby, Lincolnshire,
132	Heathcote and Lloyd (1986) observed no correlation between air temperature and
133	$\delta^{18}O_P$ and concluded that weather type and associated origin of moisture is the
134	primary factor responsible for changes in daily $\delta^{18}O_P$. At Wallingford, Oxfordshire,
135	Darling and Talbot (2003) observed weak and seasonally variable correlations
136	between daily $\delta^{18}O_P$, temperature and precipitation amount – correlations which
137	improve when monthly values are used. In a detailed analysis of event scale $\delta^{18}O_P$ in
138	Dublin, Ireland, Baldini et al. (2008; 2010) demonstrate the primary role of
139	precipitation amount and moisture source trajectory. On the basis of those data,
140	Fischer and Baldini (2011), developed a series of daily empirical functions of
141	increasing complexity to characterise $\delta^{18}O_P$ as a function of precipitation amount and
142	moisture source. The Fischer and Baldini (2011) daily functions, and approach in
143	general, offer tremendous potential for improving the interpretation of palaeoclimate
144	archives. However, the broader applicability of those functions, as with the traditional
145	Dansgaard (1964) type relationships, is dependent upon understanding how the
146	coefficients and model skill vary in space and through time. In particular, the causal
147	mechanism between rainfall amount and $\delta^{18}O_P$, and how that relationship evolves
148	through the lifespan of a rainfall event, remains poorly understood. Here, in an
149	attempt to better evaluate the spatial and temporal heterogeneity of daily isotope

150	functions, and to address the issue of signal vs. noise in daily $\delta^{18}O_P$ data, daily
151	monitoring of $\delta^{18}O_P$ was carried out at multiple locations across the relatively small
152	spatial gradient of the British Isles. We report daily $\delta^{18}O$ measurements over 57 days,
153	sampling rainfall events across each of the four seasons from up to 70 sites. These
154	data indicate daily spatial δ^{18} O gradients within the British Isles of up to 17‰,
155	highlighting the role of the evolution of weather systems in driving local scale
156	variability in $\delta^{18}O_P$. We use these data to evaluate the empirical relationships between
157	$\delta^{18}O_P$ and daily weather, and to qualitatively assess the potential for developing a
158	synoptic typology for $\delta^{18}O_P$ in the British Isles.
159	
160	Sites and Methodology
161	
162	Daily precipitation water samples were collected from up to 70 sites within England,
163	Wales, Scotland and Northern Ireland (Figure 1; Table 1). The samples were collected
164	as part of a pilot study for the British Isotopes in Rainfall Project (BIRP) - a
165	community engagement initiative, in collaboration with volunteer weather observers
166	and the UK Met Office. Initially, 17 volunteers were engaged, contacted via the
167	Climate Observers Link (COL) or via existing monitoring programmes (Table 1).
168	This group was subsequently augmented by further weather observers with ongoing
169	association with the U.K. Met Office. Precipitation water samples were collected
170	using a standard Met Office rain gauge at 9 am GMT each day. Having measured the
171	amount of rainfall for the previous 24 hours, the rain water samples were transferred
172	to 4 ml or 8 ml Nalgene ® HDPE bottles, depending on rainfall amount taking care to
173	ensure bottles were full to avoid any exchange of sample oxygen with air in the bottle.
174	The bottles were labelled, stored at 4°C and sent to the British Geological Survey at

the end of each month. Details of sampling practice were communicated to

176 participants via an online video (http://tinyurl.com/BIRP2010), with further

177 instructions provided via post to each sampler.

178

Precipitation samples were collected during the course of four campaigns, conducted 179 180 in March 2010, October 2010, July 2011 and January 2012, thereby capturing a 181 subsample of each of the seasons. Collection days were inevitably limited by the 182 occurrence of precipitation events and availability of volunteers, and thus the sample 183 set for each site and month range from 2-19 samples (Table 1). In addition to 184 collecting precipitation water, at the majority of stations a range of meteorological 185 data was recorded. Every station provided rainfall amount data, and the majority also 186 provided air temperature recordings. Where local temperature readings were not 187 taken, temperature data from a nearby Met Office station was used (Table 1). Because 188 precipitation samples were collected at 9 am each day, the date of precipitation in 189 each instance was assigned as the previous day, both for precipitation amount data 190 and isotope composition, following standard practice for the U.K. Met Office. The 191 daily North Atlantic Oscillation (NAO) index was obtained from the U.S. National 192 Oceanic and Atmospheric Administration (NOAA) Climate Prediction Center 193 (http://www.cpc.ncep.noaa.gov). 194

195 Oxygen and hydrogen isotopes of water were analysed at NERC Isotope Geosciences

196 Facility at the British Geological Survey. For D/H analysis, the hydrogen was

197 liberated by Cr reduction, while ${}^{18}O/{}^{16}O$ were equilibrated with CO₂ using an

198 ISOPREP 18 device. Mass spectrometry was performed on a VG SIRA (δ^{18} O) and

199	IsoPrime (δ^2 H) in conjunction with laboratory standards calibrated against NBS
200	standards. Long term analytical errors are 0.05‰ for δ^{18} O and <1‰ for δ^{2} H.

Spatial patterns in daily $\delta^{18}O_P$ were mapped using the *filled.contour3()* program in R, 202 203 which uses the function *akima()* to perform bivariate data interpolation (Akima 1978). 204 Backward trajectories of air parcels arriving at the British Isles were computed using 205 the web-based HYbrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) 206 model (Draxler and Hess 1997; Draxler and Hess 1998; Draxler and Rolph 2015; 207 Rolph 2015) for a matrix of 63 locations between 5.7°W, 50°N and 2.7°E, 58°N. Back 208 trajectories were computed for air parcels arriving at 1500 m.a.s.l. at six-hourly 209 intervals prior to the time of water sampling at 9:00 hrs. Principle locations of 210 moisture uptake were estimated as the first point in a particular trajectory whereby the 211 specific humidity increased by >0.5 g/kg and atmospheric pressure was >900 hPa, 212 following Krklec and Dominguez-Villar (2014). 213 214 **Results** 215 Regression between daily $\delta^{18}O_P$, maximum air temperature and precipitation amount 216 217 218 For the statistical analysis of daily precipitation isotope data, we treat the data from 219 January 2012 separately from those from March 2010, October 2010 and July 2011. 220 This is because the January 2012 sample contains observations from \sim 4 times as 221 many locations, yet over three days, which contrasts with the fewer (15-17) locations 222 over up to 19 days for the other sampling months. In all instances, relationships with precipitation amount are explored using a power coefficient ($P^{0.5}$), to ensure a 223

224 Gaussian distribution of regression residuals, following the reasoning outlined in 225 Fischer and Treble (2008) and Fischer and Baldini (2011); nonlinear relationships are 226 known in climate-isotope studies, such as in the discussion of Rayleigh fractionation 227 later in this paper, and root transformations are often applied to precipitation data in 228 climatology because precipitation data are typically skewed. Daily maximum 229 temperatures (T_{max}) are used, since they relate to the temperature around noon on the 230 day of precipitation, whereas mean daily temperatures for the period of sampling (9 231 am - 9 am) are not consistently available.

232

The relationship between δ^{18} O_P, T_{max} and $P^{0.5}$ is explored first using all daily data 233 234 (excluding January 2012), and then with data subset according to month and site in 235 order to ascertain the consistency of relationships in time and space. Due to the 236 paucity of data, monthly subsets from each site were not analysed individually. When 237 all daily data are combined, there are weak yet statistically significant (p < 0.001) relationships between $\delta^{18}O_P$, $P^{0.5}$ and T_{max} ($r^2 = 0.17$ and 0.02 respectively) but not 238 NAO (Figure 2). Significant regressions can also be observed between $\delta^{18}O_P$ and $P^{0.5}$ 239 when data are subset according to month, with slope coefficients varying between 240 March 2010 (slope = -1.45, $r^2 = 0.24$), October 2010 (slope = -0.97, $r^2 = 0.14$) and 241 July 2011 (slope = -0.85, $r^2 = 0.19$) (Figure 3a-c). Significant regressions (p < 0.05) 242 did not exist between $\delta^{18}O_P$ and T_{max} for monthly subsets for March or October 2010 243 (Figure 3d-e), however a weak ($r^2 = 0.04$, p = 0.01) relationship was observed for July 244 2011 (Figure 3f). The mean T_{max} for the three sampled months varied markedly, from 245 246 10.54°C in March 2010, 12.74°C in October 2010 and 17.21°C in July 2011 (Figure

247 3d-f). There were no significant regressions (p < 0.05) between $\delta^{18}O_P$ and NAO

248 where data were subset according to month (Figure 3g-i).

249

250 When data are subset according to sampling location (incorporating data from each month excluding January 2012), significant regressions between $\delta^{18}O_P$ and $P^{0.5}$ can be 251 252 observed with p < 0.05 at 11 of the 17 sites (Figure 4). Those sites that did not exhibit 253 significant precipitation effects were Edenbridge (EDEN), Glenmore Lodge (GLEN), 254 Horsham (HOR), Lunan Valley (LUNA), Wallingford (WALL) and West Moors (WMOR) (Figure 4). Five sites exhibited significant $P^{0.5}$ effects with $r^2 > 0.3$, and 255 256 those were Acton (ACT), Aboyne (ABYE), Carlton-in-Cleveland (CACL), Darvel 257 (DAR) and Lawkland (LAW). Given the paucity of data, it was not possible to rigorously test for differences in the within-site $P^{0.5}$ vs. $\delta^{18}O_P$ relationships between 258 259 months, although there is no obvious indication that data from March 2010, October 260 2010 or July 2011 exhibit markedly different response patterns to the regression 261 models based on all months combined (Figure 4). 262 Five of the 17 site-specific regressions between T_{max} and $\delta^{18}O_P$ are significant to $p < 10^{10}$ 263 264 0.05, data from Acton (ACT), Ebbr Vale (EBBV), Glenmore Lodge (GLEN), 265 Marlborough (MARL) and Wallingford (WALL) (Figure 5). The slope coefficients 266 for those relationships range between +0.14 at Glenmore Lodge (GLEN) to +0.37 at

Acton (ACT). For most sites, particularly those with significant T_{max} effects, there is

- visible clustering of data according to month (Figure 5). There is only one site,
- Edenbridge (EDEN) which exhibits a significant (p < 0.05, $r^2 = 0.58$) positive
- 270 relationship with NAO, however only a single month (March) was monitored at that
- site (Figure 6). Two additional sites West Moors (WMOR) and Horsham (HOR)

also exhibited significant regressions between March $\delta^{18}O_P$ and NAO (Supplementary Information, Figure S1).

274

The spatial distribution of regression coefficients for $P^{0.5}$ and T_{max} vs. δ^{18} O_P are 275 276 mapped in Figure 7. Sites which exhibit significant regression coefficients between δ^{18} O_P and $P^{0.5}$ are distributed across the British Isles, however the sites with the 277 largest r^2 and lowest (most negative) slopes vs. $P^{0.5}$ are those in northern England and 278 279 Scotland, with the exception of Acton (ACT) in central England (Figures 7a and 7b). 280 Two sites in Scotland, Glenmore Lodge (GLEN) and Lunan Valley (LUNA), exhibit no significant relationship with $P^{0.5}$ (Figures 7a and 7b). Only five sites produced a 281 significant regression between δ^{18} O_P and T_{max} , of which four are situated in central 282 and southern England (Figures 7c and 7d). One northern site – Glenmore Lodge 283 284 (GLEN), Scotland, also exhibited a significant relationship with temperature, whilst 285 three sites on the southern coast of England exhibited no significant temperature 286 effect (Figures 7c and 7d).

287

288 Spatial distribution of daily $\delta^{18}O_P$

289

Over the sampling period, the geographical distribution of $\delta^{18}O_P$ across Great Britain varied markedly from day to day. On some occasions, e.g. 13th March 2010 (Figure S2), 17th October 2010 (Figure S3) and 21st July 2011 (Figure S4), $\delta^{18}O_P$ values were largely homogenous (within a 2‰ range) across the entire spatial gradient. However, on other occasions, e.g. 4th October 2010, 27th October 2010 (Figure S3) and 9th July 2011 (Figure S4), marked spatial gradients in $\delta^{18}O_P$ occurred, up to a maximum range of 17‰. The degree of spatial homogeneity may, in part, relate to the sampling

297 frequency and location of precipitation samples for a particular day, however the day 298 to day differences in spatial range measured from ~15 sites is equivalent to that 299 observed over the three days sampled in January 2012 (Figure 8), where the spatial gradient varies from 17‰ on 23rd January 2012 to 10‰ on 25th January 2012, despite 300 over 60 stations being sampled on both occasions. Most frequently, daily $\delta^{18}O_P$ 301 302 patterns show a decrease along a south-west to north-east gradient, although there are 303 numerous exceptions to this rule, with occasional inversions in the south-west to 304 north-east gradient, changes in the direction of that gradient and bimodal distributions (e.g. 20th March 2010, 22nd October 2010 and 6th July 2011; Figures S2-S4). The most 305 depleted $\delta^{18}O_P$ values measured were collected at sites in Scotland and northern 306 307 England. Although a larger density of samples is preferable in order to trace spatial patterns in daily $\delta^{18}O_P$, reduction in sample number to as few as four sites still allows 308 309 for some coherent patterns to be observed. On days where precipitation fell at all locations, the pattern which emerges from 14 sampling sites (e.g. 17th July 2011, 310 Figure S4) is not dissimilar to those which can be observed based upon >80 sites (e.g. 311 24th January 2012; Figure 8c). 312

313

Spatial patterns in δ^{18} O_P are clearly best captured in January 2012, where the sample 314 density was highest (Figure 8). On 23^{rd} January 2012, the lowest $\delta^{18}O_P$ values of – 315 316 18.5‰ were obtained from rain falling in eastern-central Scotland, with a pattern of increasing $\delta^{18}O_P$ to the north, west and particularly to the south of that location 317 318 (Figure 8c). On this date, western England and Wales, Northern Ireland and southeast England experienced the highest $\delta^{18}O_P$ values of upto 0.58%, with a southward 319 pattern of increasing $\delta^{18}O_P$ along the eastern coast of England (Figure 8c). On 24th 320 January 2012, a marked longitudinal gradient was observed, with declining $\delta^{18}O_P$ 321

322	along an east-northeast trajectory (Figure 8f). Lowest $\delta^{18}O_P$ values on 24 th January
323	were observed along the eastern coast of England and Scotland (Figure 8f). The 25^{th}
324	January 2012 saw a shift towards low $\delta^{18}O_P(-15 \text{ to } -10\%)$ in the north west of
325	Scotland, Northern Ireland, north west England and northern Wales, with higher
326	$\delta^{18}O_P$ in the south of England (Figure 8i). HYSPLIT modelling indicates that two
327	moisture bearing air masses crossed the British Isles on January 23 rd , 2012 (Figures 8a
328	and 8b). The first parcel collected water vapour over the Norwegian Sea, east of
329	Iceland and collided with the northern British Isles along a north-westerly trajectory
330	(Figure 8a). The second air mass collected moisture from the central Atlantic Ocean
331	and collided with the British Isles along a south-westerly trajectory. On the 24 th
332	January 2012, HYSPLIT modelling indicates that the majority of moisture was
333	derived from the central Atlantic, impacting the British Isles along a westerly/south-
334	westerly trajectory (Figure 8d). On the 25 th January 2012, HYSPLIT modelling
335	indicates moisture arriving from a south-westerly trajectory, having markedly
336	changed direction beforehand above the Bay of Biscay to the south (Figure 8g).
337	
338	The relationship between the spatial distribution of $\delta^{18}O_P$ and synoptic weather types,
339	as classified through the Lamb Weather Type scheme (LWT; Jones et al. 1993; Lamb
340	1950) is explored in Figures 9 and 10. The majority of rainfall events during the
341	studied period were associated with two principle weather types: Cyclonic (LWT =
342	20) and South Westerly (LWT = 15) (Figure S5). Under the influence of Cyclonic
343	weather types, $\delta^{18}O_P$ exhibits a pattern of higher values in southern and south-west
344	England contrasting with a frequently occurring region of markedly lower $\delta^{18}O_P$ over
345	northern England and southern Scotland (Figure 9). Occasionally, those lower $\delta^{18}O_P$
346	values extend southwards along the eastern coast of England, e.g. on 8 th July 2011 and

347	17^{m} July 2011 (Figure 9). Higher $\delta^{18}O_{\text{P}}$ values can also be observed to the far north
348	during these weather events, e.g. on 3 rd October 2010, 8 th July 2011 and 18 th July
349	2011 (Figure 9). Under the influence of South Westerly weather, a consistent spatial
350	pattern in $\delta^{18}O_P$ is not evident and many of these days exhibit a narrow range of $\delta^{18}O_P$
351	(<5‰). On the 24 th January 2012, $\delta^{18}O_P$ values largely exhibit a SW-NE gradient
352	across Great Britain, except for low $\delta^{18}O_P$ at Llansadwryn (LLAN), North Wales
353	(Figure 10). By contrast, on 10^{th} October 2010, the lowest $\delta^{18}O_P$ values were recorded
354	in the south west of England and Wales. On other days (e.g. 18 th March 2010; 23 rd
355	March 2010), the lowest or highest $\delta^{18}O_P$ values were recorded in northern-central
356	England.

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358 Discussion

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Daily monitoring of isotopes in rainfall, on the basis of 57 days and 17 sites, support 360 previous observations that square root transformed daily precipitation amount $(P^{0.5})$ is 361 the most consistent predictor of daily $\delta^{18}O_P$ in maritime, mid-latitude regions (Baldini 362 363 et al. 2010; Fischer and Baldini 2011; Fischer and Treble 2008). All data combined (including January 2012) define a daily function $\delta^{I8}O_{P-day} = (-0.9)P^{0.5}_{day} - 4.7$, $r^2 =$ 364 365 0.1 (Figure 2a) which is very similar to the model derived by Baldini et al. (2010) based on two years monitoring of event based $\delta^{18}O_P$ at Dublin, Ireland (Baldini et al. 366 2010; Fischer and Baldini 2011) and with the relationship between daily $\delta^{18}O_P$ and 367 $P^{0.5}$ at Wallingford, England, between November 1979-October 1980 (data reported 368 by Darling and Talbot 2003). The $P^{0.5}$ regression coefficients derived when data are 369 370 subset according to month (Figure 3) and by site (Figure 4) vary compared to those 371 based on the whole dataset combined. Firstly, the regression slope varies between the

372 three months studied, suggestive of a seasonally modulated relationship between δ^{18} O_P and $P^{0.5}$ as described by Fischer and Baldini (2011). Indeed, the $P^{0.5}$ coefficients 373 374 obtained here (Figure 3) are consistent with those predicted by Equation 8 in Fischer 375 and Baldini (2011) for March and October 2010 (-1.45 and -0.97 respectively) but 376 not for July 2011, where our data indicate a slope of -0.85 compared to a predicted -0.41. Secondly, the $P^{0.5}$ coefficients vary spatially: steeper negative slope coefficients 377 378 and higher r^2 values are generally observed in northern England and Scotland 379 compared to larger slope coefficients and less frequent significant relationships at 380 sites in southern England (Figure 7). One exception to this rule - Acton (ACT) - is 381 located in the English west midlands in the rain shadow of the Welsh mountains (Figure 7a). We will discuss potential reasons for the variable $\delta^{18}O_{P-P}$ relationship 382 383 at the end of this section.

384

The relationship between air temperature and daily $\delta^{18}O_P$ is less convincing. A weak 385 yet significant correlation is observed between daily $\delta^{18}O_P$ and daily maximum air 386 387 temperature (T_{max}) based on all samples (Figure 2b). At first glance, this apparent 388 temperature effect appears to support previous observations, based on global 389 compilations of monthly data (e.g. Araguas-Araguas et al. 2000; Dansgaard 1964; 390 Rozanski et al. 1993; Rozanski et al. 1992). Furthermore, a temperature effect has 391 some theoretical grounding, since changes in temperature affect the vapour-liquid 392 fractionation factor during condensation (Dansgaard 1964; Merlivat 1978; Merlivat and Nief 1967). However the relationship between T_{max} and $\delta^{18}O_P$ does not 393 394 consistently hold when samples are subset according to month or site, with only the 395 July 2011 monthly subset and 5 out of 17 site-specific analyses producing a significant T_{max} regression (Figures 3, 5 and 7). In addition, as is the case with $P^{0.5}$, the 396

397	occurrence of significant relationships with T_{max} varies spatially, with predominantly
398	southern and central sites exhibiting T_{max} effects (Figure 7c and d). One of those sites
399	is Wallingford (WALL), for which Darling and Talbot (2003) observed significant
400	positive correlations between daily average air temperature and daily $\delta^{18}O_P$ for winter
401	(DJF) and autumn (OSN) precipitation (sampled between November 1979 - October
402	1980). The coefficients of those 1979-1980 models are similar to those derived for
403	July 2011 (Figure 3f). However, Darling and Talbot (2003) did not observe a
404	significant relationship with temperature during summer precipitation at Wallingford,
405	further highlighting the inconsistency of temperature-based regression with daily
406	$\delta^{18}O_P$. Three sites along the southern English coast exhibit no significant relationship
407	between $\delta^{18}O_P$ and T_{max} , whilst another site, Glenmore Lodge in Scotland, does
408	(Figures 7c and 7d). It is therefore not possible to make generalisations concerning the
409	spatial patterns in T_{max} effects upon δ^{18} O _P . One potential explanation for the
410	correlation between T_{max} and $\delta^{18}O_P$ when all data are combined (Figure 2), but the
411	absence of such a correlation when data are subset according to month (Figure 3), is
412	that both T_{max} and δ^{18} O _P exhibit strong seasonal components which do not affect
413	regressions on sub-monthly timescales. However, although both vary seasonally, a
414	casual relationship between T_{max} and $\delta^{18}O_P$ is not certain and their correlation may
415	instead relate to seasonal changes in a variety of conditions, including moisture
416	source, trajectory, weather type and land surface feedbacks (Baldini et al. 2010;
417	Fischer and Treble 2008; Treble et al. 2005).
418	
419	Changes in the North Atlantic Oscillation (NAO) (Hurrell 1995) would be expected to

420 influence $\delta^{18}O_P$ as it reflects the position of the westerly jet as a function of air

421 pressure differentials across the North Atlantic, resulting in changes to the source and

422 trajectory of weather systems and water vapour travelling to the British Isles. However, we observe no significant regression between $\delta^{18}O_P$ and NAO, either when 423 424 all data are combined or when data are subset according to month (Figures 2c and 3g-425 i). It should be noted that our data do not comprehensively represent winter (DJF) 426 conditions in the British Isles, the season when NAO is considered to have it's largest effect upon $\delta^{18}O_P$ (Baldini et al. 2008; Fischer and Baldini 2011). Except for January 427 2012, for which three days sampling is insufficient to examine the role of temporal 428 429 changes in the NAO, the closest month to winter sampled in this study is March 2010, whereby three sites exhibit significant correlations between NAO and $\delta^{18}O_P$: 430 431 Horsham, West Moors and Edenbridge (for which March 2010 are the only samples 432 collected) (Figure 6 and Figure S1). Otherwise, none of the other sites, or months examined, exhibit significant effects of the NAO upon British δ^{18} O_P. A generally 433 434 weak effect of the NAO would contrast with the conclusions of previous studies that the NAO has a significant, positive relationship with winter $\delta^{18}O_P$ (Baldini et al. 435 2008: Fischer and Baldini 2011). However, the limited coverage of winter rainfall 436 through this study precludes further comment on the effect of NAO upon $\delta^{18}O_P$ and 437 further sampling of daily winter rainfall across a spatial gradient is required to fully 438 439 address this uncertainty. 440

The spatial and temporal variability in $P^{0.5}$ and T_{max} coefficients indicates that models derived from individual sites cannot be unilaterally applied, even within relatively small geographical areas such as the British Isles. However, those variable coefficients do provide potential insights into the mechanisms behind the relationship between δ^{18} O_P, $P^{0.5}$ and T_{max} . Rayleigh distillation is a commonly cited simple model for isotope fractionation during water condensation within a cloud (Dansgaard 1964).

447	As a finite parcel of moisture condenses, Rayleigh fractionation predicts that the
448	initial stages of condensation will be associated with relatively little change in $\delta^{18}O_P$.
449	For example, condensation of the initial 50% of vapour is predicted to equate to an \sim
450	-8% decrease in $\delta^{18}O_P$. By contrast, condensation of the final 20% of vapour within a
451	parcel is predicted to impart an isotopic depletion of >30‰ (Dansgaard 1964). The
452	degree of rainout from a vapour parcel is therefore likely to be a simple, first order
453	mechanism which results in an inverse correlation between $\delta^{18}O_P$ and rainfall amount.
454	In reality, a pure Rayleigh distillation is unlikely to occur within a cloud, due to the
455	resupply of moisture from evapotranspiration and mixing between vapour parcels.
456	Furthermore, a wide range of factors introduce complexity, from changes in the
457	isotopic composition of the initial vapour parcel (reflecting the conditions at it's
458	origin and subsequent mixing and phase changes during transit) to the subsequent
459	modification of raindrop δ^{18} O due to evaporation and equilibrium exchange with
460	ambient vapour (Celle-Jeanton et al. 2004; Gedzelman and Arnold 1994).
461	Nevertheless, some degree of Rayleigh fractionation of atmospheric vapour remains a
462	viable explanation for some of the observed relationships between $\delta^{18}O_P$ and
463	precipitation amount, whereby weather events associated with a high amount of
464	precipitation become progressively isotopically depleted. The rain out effect is non-
465	linear and likely to result in heterogeneous spatial and temporal patterns. In particular,
466	low altitude, coastal sites more frequently encounter vapour parcels in their initial
467	stages of moisture depletion, and consequently precipitation at those locations is less
468	likely to exhibit a marked sensitivity to rainfall amount. By contrast, high altitude and
469	inland sites encounter vapour parcels that have undergone a larger degree of prior
470	rainout, meaning that subsequent condensation and precipitation should exhibit
471	steeper isotope effects. For the maritime climate of the British Isles, this pattern of

472 progressive rainout may provide one explanation for the spatial and temporal variability in the relationship between $\delta^{18}O_P$ and rainfall amount, whereby $\delta^{18}O_P$ is 473 474 more sensitive to rainout effects in northern Britain, downstream of the direction of the prevailing weather (Figure 7). By contrast, southern, low elevation sites are less 475 476 likely to be susceptible to rainout effects and therefore may exhibit correlations with 477 other variables, including temperature and changes in oceanic moisture source. It is 478 important to recognise that due to the temporal migration of weather trajectories, spatial patterns in $\delta^{18}O_P$ and associated correlations with climate variables are 479 unlikely to remain constant in time. A more detailed elucidation of the mechanisms 480 481 behind isotope fractionation in daily British rainfall could be achieved using isotope 482 enabled climate models, validated or trained against spatially resolved data such as 483 those presented here (Langebroek et al. 2011; Risi et al. 2010). Such an analysis is 484 beyond the scope of this paper but would represent a valuable direction for future 485 research.

486

487 Spatial patterns in daily δ^{18} O relate to weather types

488

Analysis of $\delta^{18}O_P$ from a highly resolved spatial context provides a means of further 489 deciphering the controls over $\delta^{18}O_P$. This is particularly apparent for the three days in 490 January 2012 for which 70 sites provided $\delta^{18}O_P$ data (Figure 8). On 23rd January 491 492 2012, the passage of an occluded front, with low pressure centres to the east and north 493 west of the British Isles resulted in a complex weather pattern (Figure 8b). The 494 majority of atmospheric flow approached the British Isles from the southwest, accounting for the gradient of decreasing $\delta^{18}O_P$ along that trajectory in southern 495 England and Wales (Figure 8a. However, the markedly low $\delta^{18}O_P$ values measured 496

497 from central Scotland relate to northerly winds travelling along a trough which 498 developed to the north west of Scotland and brought air masses from near Iceland to the British Isles (Figure 8b). On the 24th January, 2012, the previous day's complex 499 500 weather had passed, and eastern England and Scotland experienced South Westerly 501 weather characterised by a warm front passing perpendicular to the east coast, leading to a characteristic east-west $\delta^{18}O_P$ gradient suggestive of progressive rainout (Figures 502 8d-f). On 25th January 2012, a further cold front arrived in north western Scotland and 503 504 England and Northern Ireland resulting in moderate isotopic depletion of rainfall in 505 the west, and limited precipitation in the south east (Figures 8g-i).

506

The coherency of daily $\delta^{18}O_P$ patterns for the British Isles under different weather-507 508 types highlights significant potential for developing an isotope-based synoptic 509 typology, which could then be applied to palaeoclimate research. To do so rigorously 510 would require a much more detailed study, however the data obtained to date 511 highlight some encouraging patterns. The rainfall events sampled through this study 512 predominantly occurred during two common weather types: Cyclonic and South 513 Westerly weather types (according to the Lamb Weather Type scheme). Although it is 514 not possible to make conclusive statements on the way these weather types are manifest in daily $\delta^{18}O_P$ over the British Isles, there is evidence to suggest that the 515 516 rotational flow associated with Cyclonic weather types is manifest in a progressive south-north $\delta^{18}O_P$ gradient, which curves around a region of maximum isotopic 517 depletion (lowest $\delta^{18}O_P$), representative of the epicentre of the cyclonic vortex (Figure 518 519 9). The patterns associated with Cyclonic weather types in the British Isles are 520 similar, if smaller, to those related to cyclonic precipitation in the eastern United 521 States, including storm precipitation (Good et al. 2014; Lawrence and Gedzelman

522 1996). By contrast we are not yet able to make generalisations concerning the spatial $\delta^{18}O_P$ patterns associated with South Westerly weather types, for which the data to 523 524 date exhibit less coherent patterns in space (Figure 10). These varying isotopic spatial 525 gradients associated with South Westerly weather types may reflect complex or 526 heterogeneous rainfall patterns across the country and associated fractionation 527 processes. Within this context, complexities arise owing to variability in the direction 528 of flow and the interaction between numerous air parcels. For example, our data to date are insufficient to identify and evaluate the effect of frontal rainfall upon $\delta^{18}O_P$, 529 530 however it is expected that the passage of, and interaction between, warm and cold fronts would cause significant intra- and inter-daily variability in $\delta^{18}O_P$ due to 531 localised changes in air pressure, the altitude of precipitation formation and air 532 533 temperature (Celle-Jeanton et al. 2004; Celle-Jeanton et al. 2001). Furthermore, a 534 number of weather types are only sporadically captured in our dataset (Figure S5) and 535 future research should therefore attempt to undertake a more detailed and prolonged monitoring project in order to develop a synoptic typology of δ^{18} O_P for the British 536 537 Isles.

538

539 Conclusion

540

541 Daily monitoring of the oxygen isotope composition of rainfall from multiple sites 542 across the British Isles reveals a relationship between rainfall amount (square root 543 transformed; $P^{0.5}$) and δ^{18} O_P, which emerges when all data are combined and when 544 the data are subset according to month or site. By contrast, daily maximum air 545 temperature (T_{max}) exerts a weaker and less consistent relationship and daily NAO 546 exhibits limited influence, although winter conditions are not extensively sampled in

our data. The $P^{0.5}$ and T_{max} regression coefficients and r^2 vary seasonally, in support of 547 548 previous observations from Dublin, Ireland (Fischer and Baldini 2011). They also 549 vary spatially, with a greater influence of temperature in southern sites, and greater 550 influence of precipitation amount at northern sites. We propose a simple explanation 551 that these spatio-temporal patterns in regression coefficients reflect the non-linear influence of Rayleigh fractionation and rainout upon $\delta^{18}O_P$ as a vapour parcel 552 553 becomes progressively depleted. Future research involving the integration of climate 554 models with highly resolved data such as ours should be directed at testing this interpretation. By mapping the distribution of daily $\delta^{18}O_P$ across the British Isles, we 555 556 are able to observe patterns that may be characteristic of some weather types, namely 557 Cyclonic weather types under the Lamb classification scheme. These observations are 558 a step towards an improved mechanistic understanding of the climate controls over $\delta^{18}O_P$ and a synoptic typology which will aid attempts to reconstruct past changes in 559 560 dominant weather patterns.

561

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563

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579

- 581 **Table 1**
- 582 Details of sites sampled during March 2010, October 2010 and July 2011. Site numbers relate to those mapped in Figure 1. Values within the
- 583 'Sample months' columns indicate the number of samples collected in each month, at each site. Details of nearby Met Office stations used to
- 584 complement data where no local temperature measurements were taken are given in the 'Notes' column.
- 585
- 586

					Sa	mple month	s	
Site	Site No.	Latitude (°N)	Longitude (°E)	Height (m.a.s.l.)	March 2010	October 2010	July 2011	Notes
Aboyne	1	57.07	-2.79	126		15	11	
Acton	2	53.07	-2.55	44	10		10	Temperature data from MIDAS (site 1132; Reaseheath Hall; 2km NE)
Carlton-in-Cleveland	3	54.43	-1.22	88	11	15	14	
Darvel	4	55.60	-4.23	217	10	14	15	
Eastry	5	51.25	1.31	32	10	15	11	
Ebbw Vale	6	51.74	-3.18	303	10	15	14	
Edenbridge	7	51.20	0.07	47	10			
Glenmore Lodge	8	57.17	-3.68	344		13	17	Temperature data from MIDAS (site 118; Cairngorm Chairlift; 4km S)
Horsham	9	51.07	-0.34	37	10	13	8	
Lawkland	10	54.09	-2.34	176	10			Temperature data from MIDAS (site 513; Bingley; 44km SW)
Llansadwrn	11	53.26	-4.17	107	11	19	15	
Lunan Vale	12	56.66	-2.51	19		15	15	Temperature data from MIDAS (site 15045; Crombie Country Park; 19km SW)
Malham Tarn	13	54.10	-2.16	384		15	13	
Marlborough	14	51.43	-1.73	142	10	15	2	
Okehampton	15	50.74	-4.00	170		15	14	Temperature data from MIDAS (site 1345; East Okement Farm; 4km S)
Wallingford	16	51.60	-1.11	50		10	9	
West Moors	17	50.82	-1.88	17	10	13	12	
Total samples					102	202	180	



Fig. 1

Topographic map of the British Isles (dark shaded areas = higher elevation), including
the location of the sampling sites used in this study. Open circles are those sites only
sampled in January 2012, whereas open boxes indicate the sites sampled during all
months. The numbers in the squares indicates the site number, as detailed in Table 1.





Scatter plots of daily $\delta^{18}O_P$ vs. (a) precipitation amount (square root transformed; $P^{0.5}$), (b) daily maximum temperature (T_{max}) and (c) North Atlantic Oscillation index (NAO) for all samples collected during March 2010, October 2010 and July 2011. Regression coefficients are given inset. Solid blue lines = significant linear model fit, dashed green lines = 95% confidence intervals. Non-significant linear models are illustrated with dotted red lines. Blue text relates to significant regression models, whereas red text indicates non-significant models.





Scatter plots of daily $\delta^{18}O_P$ vs. (a, d, g) precipitation amount (square root transformed, 609 $P^{0.5}$), (b, e, h) daily maximum temperature (T_{max}) and (c, f, i) North Atlantic 610 611 Oscillation index (NAO) for all samples subset according to month of sampling 612 collected during March 2010, October 2010 and July 2011. Regression coefficients 613 are given inset. Solid blue lines = linear model fit and dashed green lines = 95%614 confidence intervals for significant regressions. Non-significant linear models are 615 illustrated with dotted red lines. Blue text relates to significant regression models, 616 whereas red text indicates non-significant models.





618 **Fig. 4**

619 Scatter plots of daily $\delta^{18}O_P$ vs precipitation amount (square root transformed, $P^{0.5}$) 620 subset by site for all samples collected during March 2010 (open circles), October 621 2010 (open triangles) and July 2011 (crosses). Regression coefficients are given inset. 622 Solid blue lines = linear model fit and dashed green lines = 95% confidence intervals 623 for significant regressions. Non-significant linear models are illustrated with dotted 624 red lines. Blue text relates to significant regression models, whereas red text indicates 625 non-significant models.





629 Scatter plots of daily $\delta^{18}O_P$ vs. daily maximum temperature (T_{max}) subset by site for 630 all samples collected during March 2010 (open circles), October 2010 (open triangles) 631 and July 2011 (crosses). Regression coefficients are given inset. Solid blue lines = 632 linear model fit and dashed green lines = 95% confidence intervals for significant 633 regressions. Non-significant linear models are illustrated with dotted red lines. Blue 634 text relates to significant regression models, whereas red text indicates non-significant 635 models.







639 Scatter plots of daily $\delta^{18}O_P$ vs. North Atlantic Oscillation index (NAO) subset by site 640 for all samples collected during March 2010 (open circles), October 2010 (open 641 triangles) and July 2011 (crosses). Regression coefficients are given inset. Solid blue 642 lines = linear model fit and dashed green lines = 95% confidence intervals for 643 significant regressions. Non-significant linear models are illustrated with dotted red 644 lines. Blue text relates to significant regression models, whereas red text indicates 645 non-significant models.



648 Fig. 7

649 Map of regression coefficients against daily rainfall amount (square root transformed, 650 $P^{0.5}$) and daily maximum temperature (T_{max}) for all sites in Table 1, except Edenbridge 651 and Lawkland, for which only one month's data were collected. (a) r^2 statistic for $P^{0.5}$ 652 vs. $\delta^{18}O_P$; (b) slope for $P^{0.5}$ vs. $\delta^{18}O_P$; (c) r^2 statistic for T_{max} vs. $\delta^{18}O_P$; (d) slope for 653 T_{max} vs. $\delta^{18}O_P$. Open circles indicate the location of sites for which no significant 654 regression was observed for the variable mapped.



655

656 Fig. 8

Weather and isotope maps for 23rd, 24th and 25th January 2012. (a, d, g) HYSPLIT 96 657 658 hour back trajectories for air parcels arriving at a matrix of 63 points across the 659 British Isles. All simulations start at 1500 m.a.s.l. Grey lines indicate all back 660 trajectories, red lines are those whereby significant moisture uptake is estimated. The 661 length of the red lines indicates the distance from the initial site of moisture uptake. 662 (b, e, h) Daily weather maps, as reported by the U.K. Met Office. (c, f, i) The spatial distribution of δ^{18} O_P based on upto 70 monitoring stations, collecting precipitation 663 664 water at 9 am the following day.







668 Spatial pattern of δ^{18} O_P occurring on days with a Cyclonic weather pattern (Lamb

669 Weather Type =
$$20$$
)



Fig. 10

674 Spatial pattern of δ^{18} O_P occurring on days with a South Westerly weather pattern

675 (Lamb Weather Type = 15)

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