Measurement reports

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Spatial variability of northern Iberian rainfall stable isotope values: Investigating <u>elimatic atmospheric</u> controls on daily and monthly timescales

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Abstract. This article presents for the first time a large dataset of rainfall isotopic measurements $(\delta^{18}O_p \text{ and } \delta^2H_p)_{-}$ -sampled every day or every-two days from seven sites <u>oin</u> a west-to-east transect across northern Spain for 2010-2017. The main aim of this study is to: (1) characterize <u>the</u> rainfall isotopic variability in northern Spain at daily and monthly time scales, and (2)

assess the principal influencing factors determining theinfluencing rainfall isotopic variability. This comprehensive spatiotemporal approach allows exploring the role of air mass source in determining the isotopic composition of rainfall in northern Iberia by using back trajectories; Atlantic fronts are found to be the dominant source of northern Iberia rain events

- 35 studied. The relative role of air temperature and rainfall amount in determining shaping determining the stable isotope composition of precipitation changes along the west-to-east transect, being and is presented overlapping with other processes. Aair temperature appears to be the most significant influence on highly correlated with $\delta^{18}O_0$ at daily and monthly time scales, with the highest air temperature 8⁴⁸O, dependency found for a the Pyrenean station located in the Pyrenees while a few sites in-along the transect show a significant negative correlation with precipitation. The highest air temperature δ^{18} Or
- dependency is found for a station located in the Pyrenees, amount, Distance from the coast, Frontal systems associated with 40 North Atlantic cyclones are the dominant mechanism inducing precipitation in this region, in particularly in winter. This study allows an exploringation of the role of air mass source and trajectory in determining the isotopic composition of rainfall in northern Iberia by characterizing the moisture uptake for three of the seven stations. It is evident the The importance of continental versus marine moisture sources is evident, which clearly change its relevance depending owith
- 45 clear seasonal and spatial variationsn the analysed station and the season site elevation, and moisture source region (Atlantic versus Mediterranean) also significantly modulate the $\delta^{18}\Theta_p$ values and ranges In addition, but the type of precipitation (convective versuss frontal rainfall) plays a key control-role, with convective rainfall associated with higher $\delta^{18}O_n$ values. This comprehensive spatio-temporal approach This-to dataset-analyzseing of-the rainfall isotopic composition represents another step forward towards developing a more detailed, mechanistic framework for interpreting stable isotopes in rainfall 50 as a palaeoclimate palaeoclimate and hydrological tracer.

1 Introduction

The oxygen isotopic composition of rainfall ($\delta^{18}O_p$) is often considered as the dominant influence on the isotopic composition of terrestrial archives (ice cores, speleothems or authigenic lacustrine carbonates) used to reconstruct past climate (e.g., Leng, 2006). However, few palaeoclimate reconstructions are supported by an in-depth understanding of the regional climatic controls on modern precipitation $\delta^{18}\Theta_{0}$ (e.g. Treble et al., 2005). As a consequence, palaeoclimate 55 proxies are often interpreted without a clear knowledge of the processes involved in modulating $\delta^{18}O_n$ at in a particular region (López-Blanco et al., 2016; Moreno et al., 2017). It has long been established that $\delta^{18}O_n$ is an integrated product of air masses history-, modulated by specific prevailing meteorological conditions, for examplein particular, -(air temperature and amount of precipitation for example) (Craig, 1961; Dansgaard, 1964). This results in implies that several different dominant factors may controlling $\delta^{18}O_n$ variability depending on the site location, i.e., latitude, continentality, elevation, seasonal 60 distribution, local air temperature, and the amount and source of precipitation (Rozanski et al., 1993). A detailed study of

current $\delta^{18}O_p$ values and their variability in a given region is mandatory if one wishes<u>essential</u> to reconstruct<u>ing</u> past climate changes using $\delta^{18}O$ in regional climate archives (Lachniet, 2009).

Long rainfall isotopic time series allow for comparison of the $\delta^{18}O_p$ signal with meteorological variables and calibration of proxy records. Unfortunately such long-term observational studies are scarce, and thus, only a few, although-albeit

outstanding, examples of studies examining factors controlling $\delta^{18}O_p$ are available for continental Europe (Field, 2010; Genty et al., 2014; Tyler et al., 2016). The Yet, application of results obtained in for other European regions mostly under the influence of rainfall with Atlantic origin (e.g., Baldini et al., 2010) are not valid<u>cannot be directly applied to for the</u> Iberian Peninsula (IP), where three major precipitation weather regimes of precipitation regimes coexist (Millán et al., 2005)

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- 70 and where a potential for palaeoclimate reconstructions exists through via speleothems derived proxiesanalyses. Previous studies have shown that the spatial distribution of $\delta^{18}O_p$ and δ^2H_p at aon monthly time scales in Spain can be explained approached by a simple multiple regression model, based only on two geographic factors: latitude and elevation (Díaz et al., 2007; Díaz-Tejeiro et al., 2013). However, this models do not reproduce the observed distribution of stable isotope precipitation composition of precipitation with a detailed spatial resolution. -The well-known complex topography and varied
- 75 weather regimes of Spainof the Iberian Peninsula (AEMET and Instituto de Meteorologia de Portugal, 2011; Martin-Vide and Olcina-Cantos, 2001) require further detailed more targeted studies that take into account for the and highly spatiallyresolved studies high spatial variability of $\delta^{18}O_p$ in the regions Iberia and the multiple (and sometimes overlapping)se processes determining rainfall isotopic composition its temporal variation.

A major advance in understanding the controls on $\delta^{18}O_p$ has been the proliferation of studies using daily-scale monitoring to

80 address the mechanisms behind isotopic signatures at daily timescales (Baldini et al., 2010; Fischer and Baldini, 2011), which incorporating incorporate the complexity associated with the different types of rainfall (eg. frontal or convective system) associated to the type of rainfall, for example (Aggarwal et al., 2016). Regrettably, the scarcity of Global Network of Isotopes in Precipitation (hereafter GNIP) sites in Iberia, particularly those recovering obtaining using data at a-daily time scales, prevents a broader regional study of climate controls on $\delta^{18}O_n$ values. In the IP, only one study has analysed $\delta^{18}O_n$ 85 variability at-on a daily basis covering-over a short 3-year period (2000-2002) (Araguás-Araguás and Diaz Teijeiro, 2005) and, more recently, a 3-year monitoring survey focused on the Iberian Range (Molinos Cave, Teruel, NE Spain) (Moreno et al., 2014). That study revealed the importance of the *source effect* on $\delta^{18}O_p$ values, due to the alternating influence of two air masses with different trajectories origins with and different isotopic ranges. the basically, Atlantic fronts, which are associated with more negative $\delta^{18}O_p$ values (from the west), and Mediterranean convective storms, with more positive values (from the 90 eastern sources) air trajectories (Moreno et al., 2014). Additionally, another recent study based on back trajectories emphasized the role of recycled moisture uptake within the IP in the final values of $\delta^{18}O_p$ in Central Spain (Eagle Cave) (Krklec and Domínguez-Villar, 2014). Besides those studies based on-the $\delta^{18}O_{n}$, another recent work focused on trace elements on precipitation at two Pyrenean sites reveals the importance of seasonality in the role played by continental vs. marine sources of moisture (Suess et al., 2019). In addition, to date, the majority of empirical studies of the-meteorological

- 95 controls over δ¹⁸O_P-δ¹⁸O_P, rely uponare based on event, scale, daily, or monthly time series from individual locations (Moreno et al., 2014; Smith et al., 2016); The scarce studies dealing with multiple sampling locations span areas under the influence of the -the influence of same climatic regime (Baldini et al., 2010; Jeelani et al., 2018), which This approach raises concerns about the spatial representativeness of the resulting statistical models and the mechanisms behind those-the identified relationships in areas so complex as the IP. (Baldini et al., 2010; Jeelani et al., 2018)
- In this paper, we present a comprehensive analysis one f daily and monthly propose an alternative approach by analysing daily and monthly patterns of $\delta^{18}O_P$ from *multiple* stations across northern IP, and all the way to the Balearic Islands, following an 850-km long west-to-east transect (of 850_km in straight line)spherical_distance_that extends from an area dominated by a a-typical Atlantic climate elimate to one dominated by a fully-Mediterranean sitesclimate. The overall aim is to characterize and quantify the dominant factors modulating $\delta^{18}O_P$ variations in time (daily and monthly) and space, in order
- to determine the causes of <u>regional</u> precipitation <u>isotopic-isotope</u> variations<u>-regionally</u>. <u>The role of geographic factors</u> (continental and elevation effects) and atmospheric processes (moisture origin and type of rainfall) is evaluated. Additionally, this study will serve to improve the interpretation of oxygen isotope paleo-records from <u>the regionnorthern IP</u> that depend on $\delta^{18}O_{p}$ Bartolomé et al., 2015; Domínguez-Villar et al., 2017; López-Blanco et al., 2016; Pérez-Mejías et al., 2019; Sancho et al., 2018, 2015).

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2 Prevailing climate regime Weather regime, climate and site description

Our study compares for the first time, rainfall isotopic values and meteorological variables (temperature, precipitation, moisture sources and type of rainfall) at seven sites in northern Iberia and Balearic Islands, covering an 850-km long west-to-east transect from an area under typical Atlantic <u>climate</u> (Oviedo and El Pindal) to <u>a</u> fully Mediterranean <u>climate</u> (Mallorca Island and Barcelona)-elimate. The west-to-east transect is completed with three additional sites in a transitional zone: two from the Iberian Range (Molinos and Ortigosa de Cameros) and one from the Pyrenees (Borrastre) (Figure 1a). At those seven locations, rainfall was sampled <u>on</u> daily <u>basis</u> covering different time periods except at El Pindal where rain-it was collected every 48h (Table 1). <u>The</u> Borrastre record is, to our knowledge, the most comprehensive dataset of daily δ¹⁸O_p for Spain in terms of both the-time span covered (2011-2016) and number of samples (380 days) (Table S1).

- In north-western and north-central Iberia, precipitation is mainly controlled by the presence of westerly winds and the passage of Atlantic fronts, mainly especially during November-April (Martín-Vide and Olcina Cantos, J., 2001; Rüdisühli et al., 2020). During the rest of the year, the subtropical Azores high-pressure system shifts northward, favouring stable conditions by which blocking blocks the westerly circulation and moisture inflow inflow from Atlantic sources (Archer and Cantos, C
- 125 Caldeira, 2008), thus <u>favouring stable atmospheric conditions and</u> reducing precipitation. This wet winter/dry summer regime is quite different from that in the north-eastern Mediterranean region<u>of Iberia</u>, where winters are generally dry (foehn effect) whereas <u>warm seasonin the warm season precipitation</u> (from late spring to early autumn) <u>precipitation is more</u> <u>abundant and is</u>-dominated by convective storms and also <u>easterly advections over the Mediterranean Seafronts that</u>

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approach the IP from the east (backdoor cold fronts) (Millán et al., 2005). These local to-mesoscale storms are circulation is 130 primarily associated with frequent and persistent sea breezes (Azorin-Molina et al., 2011), -which bring warm and moist air masses from the Mediterranean sea inland (Azorin-Molina et al., 2009). During the summer season, this is typically the only source of precipitation in the north-eastern areaIP, bringing an average of 100-125 mm yearly (Millán et al., 2005). Backdoor cold fronts from the Mediterranean Sea are sporadic events occurring mainly in autumn (secondarily and to a lesser extent in winter-spring), but they-that can cause heavy precipitation and flooding (Llasat et al., 2007). Figure 1B summarizes these 135 three major precipitation regimes defined by Millán et al. (2005): (i) Atlantic frontal systems (westerly winds), (ii)

convective-orographic storms, and (iii) Backdoor cold fronts from the Mediterranean Sea (easterly winds). Winter precipitation in large parts of the IP is strongly influenced by the North Atlantic Oscillation (NAO) at annual and interannual scales: higher precipitation occurs when the NAO index (NAOI) is negative and the storm tracks are shifted southwards, more directly influencing the IP (Trigo et al., 2002). Lower correlation values (r = 0.1, 0.4) between the NAOI

and winter rainfall, however, are observed in our region of interest, the northern IP (Goodess and Jones, 2002), which 140 encompasses both the wet western regions and the dry Mediterranean in the northeast. For the latter region, a significant relationship with the Western Mediterranean Oscillation (WeMO) in spring and autumn is attributed to fluctuations of warm moist inflow air from the east and its influence on Mediterranean cyclogenesis (Martin-Vide and Lopez-Bustins, 2006).

The rainfall influencing the seven stations included in the studied transect originates in two dominant source regions: the tropical subtropical North Atlantic and the Western Mediterranean (Gimeno et al., 2010). Below, the seven studied stations 145 are grouped into four regions across which the seven stations are distributed areand described in terms of their climatology. Regional meteorological data are provided in Figure 4A.

The Cantabrian coast. The sites of El Pindal and Oviedo in the Cantabrian coast (Figure 1A) is-are characterized by a typical oceanic climate with mild summers and winters (Cfb, following Köppen and Geiger - KGC- classification) due to the

proximity to the coast. Rainfall occurs along the whole yearmainly occurs in late autumn and early winter with a minimum 150 in summer-(Figure 4A), and are is associated with Atlantic frontal systems (westerly winds). Additionally, rainfall samples from Oviedo (climate characteristics similar to those at El Pindal) were collected and are also included in this study.

The Iberian Range. Ortigosa de Cameros is located in the Encinedo Mountain area in the westernmost sector of the Cameros Range (Iberian Range, Figure 1A) and is dominated by a continental Mediterranean climate (Dsb, following KGC

classification). Rainfall mostly-occurs mostly in autumn and spring, with some convective-orographic storms in summer 155 (climograph in Figure 4A). The Molinos site is aAlso located in the Iberian Range and at similar elevation but further east, in the Maestrazgo basin, It -the Molinos site is characterized by a similar climate (Dsb in KGC classification), with a highly pronounced seasonality;-and pprecipitation occurring-occurs mainly in spring and in-autumn (Figure 4A).

The Pyrenees. Borrastre village is located in the Central Pyrenees (Figure 1A) and is influenced by has a transitional climate 160 Mediterranean-Oceanic (Csb in KGC classification), with precipitation occurring mainly in spring and, to a lesser extent, in autumn-(Figure 4A), exhibiting a mix of the three Atlantic, Mediterranean and convective precipitation regimes.

The Mediterranean. The typical Mediterranean climate (Csa in KGC classification) is represented by the Manacor and Porto Cristo localities in the Mallorca island and by Barcelona (Figure 1A). Precipitation is mostly distributed from October to April (Figure 4A) mostly typically associated with backdoor cold fronts from the Mediterranean Sea (easterly winds) as the influence of Atlantic precipitation is weak ened-over this area.

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3 Analytical and statistical methods

3.1 Sampling

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Rainwater was collected using a similar procedure to that recommended by the International Atomic Energy Agency (IAEA) for daily sampling (http://www-naweb.iaea.org/napc/ih/IHS resources gnip.html) for six of the seven stations (Oviedo, Ortigosa de Cameros, Molinos, Borrastre, Mallorca and Barcelona). Thus, Pprecipitation events greater than or equal to 1 mm were sampled manually from a the water accumulated in the rain gauge which allows measuring the amount of rain

- fallen and sampleusing -it manually taking out the water from the rain gauge with a syringe. The collected water was then 175 homogenized and filtered at the time of sampling; later a 5 ml aliquot was stored in polypropylene tubes sealed with screwcup without air inside and kept cold in a refrigerator until isotopie be analysis analyzed. Rainfall samples were collected at the end of each precipitation event, immediately afterwards whenever possible or after a few hours, with the total event precipitation homogenized. At El Pindal site the procedure was different: rainfall was collected every 48h for several months
- 180 (November 2006 to April 2009, a total of 101 samples) using an automated self-built revolver-type sampler-(Table 1)(Kennedy et al., 1979)- which that contained twenty-four 1L Nalgene bottles, thus avoiding any mixing of subsequent samples (see Fischer et al., 2019). A film of paraffin oil was used to prevent evaporation. It was-located on the roof of the San Emeterio lighthouse located <10 m from the modern sea cliff (Table 1). and 200 m from the cave. Thus, since the samples were automatically collected and remained in the lighthouse for several days, a film of paraffin oil was used to

185 prevent evaporation.

The observation staff in charge of each location collected a sample directly following every rainfall event, except in El Pindal that the system washas an automatic system and in Mallorca, where several events were missed during the first two years of the collection period, preventing the calculation of monthly averages for some intervals (monthly and annual averages and standard deviations in Table 2). In addition, seven rainfall events were collected at two different localities in

- Mallorca (Manacor and Porto Cristo) obtaining similar S¹⁰O₂ results. For those events, a weighted-average value using the 190 ealculated (see Table S1). Thus, 47 rainfall samples were collected from Oviedo manually in 2015. In Ortigosa de Cameros, rainfall was manually collected daily between September 2010 and December 2014 by the staff (guides) of the La Viña and La Paz show caves, with an interruption from December 2012 – January 2014 (total of 193 samples). In Molinos, rainfall was manually collected by the staff of the Grutas de Cristal cave every day for just over five 195 years (March 2010-May 2015, 268 samples). The first 2.53 years rainfall data from that survey was previously published

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(Moreno et al., 2014; Pérez-Mejías et al., 2018). In Borrastre, rainfall was manually collected daily using a Hellman rain gauge daily from April 2011 to May 2016 (380 events). In Barcelona, rainfall samples were manually obtained from the weather station on the roof of the School of Physics of the University of Barcelona using a standard rain gauge (53 samples). In addition, 98 seven-rainfall events were collected in Mallorca, 7 were replicated at two different localities in Mallorea (Manacor and Porto Cristo) obtaining similar $\delta^{18}O_2$ results. For those 7 events, a weighted-average value using the two

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localities was calculated (see Table S1).

3.2 Analytical methods

The isotopic composition of oxygen and hydrogen in rainfall samples are, expressed as δ^{18} O and δ^{2} H, reported in % relative to Vienna Standard Mean Ocean Water (VSMOW). Molinos, Borrastre and most of Ortigosa de Cameros samples (143 samples) were analysed using a Finningan Delta Plus XL mass spectrometer at the IACT-CSIC in Granada. Water samples were equilibrated with CO₂ for the analysis of δ^{18} O values (Epstein and Mayeda, 1953), while the hydrogen isotopic ratios were measured on H₂ produced by the reaction of 10 µL of water with metallic zinc at 500°C, following the analytical method of Coleman et al. (1982). The analytical error for δ^{18} O and δ^{2} H was ±0.1 and ±1 ‰, respectively. The Mallorca and Barcelona samples and the remaining samples from Ortigosa de Cameros (50 samples) were analysed at the Scientific and

- Technological Centre from the University of Barcelona, δ²H via TCEA pyrolysis coupled to Thermo Delta Plus XP mass spectrometer and δ¹⁸O with a MAT 253 Thermofisher spectrometer coupled with a gas bench. The analytical error for δ¹⁸O and δ²H was ±0.2 and ±1.5 ‰ respectively. El Pindal samples were measured at three different laboratories (see Stoll et al., 2015, for more details). Rainfall collected from November 2006 through the end of February 2007 was analysed at the University of Barcelona using the procedure described above. Rainfall collected from June 2007 to May 2008 was analysed
- 215 in the Marine Biological Laboratories of the University of Oviedo, using equilibration with CO₂ on GV Multiflow-Bio unit coupled to a GV ISOPRIME CF mass spectrometer. Rainfall collected from June 2008 to April 2009 and samples from 2015 were analysed using equilibration with CO₂ on Gas Prep unit coupled to a Nu Instruments Horizon mass spectrometer at the University of Oviedo. Uncertainties are $\pm 0.1\%$ (1s) for δ^{18} O and ± 1 %.for δ^{2} H, based on replicate analyses. Unfortunately, no comparison was made between the different involved laboratories and thus the study does not account for possible offsets
- 220 between them.

Additionally, 18 samples of potentially evaporated water with abnormally high values in $\delta^{18}O_p$ - and that occurred in summer months when maximum daily air temperatures exceeded 30°C - were classified as outliers and removed from the database. These 18 samples (Table S1) were from Ortigosa de Cameros (4 samples), Borrastre (6 samples) and Molinos (8 samples). Partial evaporation of falling rain-droplets is an alternative interpretation of tfor the high $\delta^{18}O_p$ values of these samples.

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3.3 Meteorological data

Air temperature and precipitation Meteorological data to investigate the statistical relationship between isotopic values (at on daily and monthly time scales) and main climate variables (air temperature and precipitation) were obtained from the closest 230 meteorological stations over the sampling periods, as indicated in Table 1, to investigate the statistical relationship with isotopic values. For Oviedo, meteorological data are obtained from Oviedo AEMET station. For -and for-El Pindal (120 km from Oviedo; 70 km from Santander), since there was-were not good data from nearby stations, we decided to use ERA-Interim re-analysis of from the European Center for Medium-range Weather Forecasts (ECMWF), which that provides gridded weather data (Berrisford et al., 2009; Dee et al., 2011). For Ortigosa site, meteorological data are were obtained from 235 the Villoslada de Cameros meteorological station (http://www.larioja.org/ emergencias-112/es/meteorologia), at-6.5 km far from the rainfall collection site. The Borrastre sampling site has its own meteorological station (http://borrastre.dyndns.org/MeteoBorrastre) (Table 1), except for the but for the first 22 events data were obtained-that-were derived-from ERA-Interim since the station was not yet operative. Finally, for Mallorca we used data from thefrom Sant Llorenç station (8 km), --while Barcelona meteorological data are obtained from Zona Universitaria station (www.meteo4u.com). 240

3.4 Statistical analyses

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Prior to conducting correlation analysis at daily time scales, we removed the seasonal component cycle of the variables by subtracting their monthly averages to avoid sympathetic seasonal correlations (e.g. {Kawale et al., 2011; Rozanski et al., 1993) (Table 3A). To establish correlations on theat monthly scale with meteorological variables (Table 3B), $\delta^{18}O_n$ monthly averages weighted by the amount of precipitation were calculated using the following formula (Figure 4B):

 $\delta^{18}O_{\text{monthly}} = ((Q1 \times \delta^{18}O_1) + (Q2 \times \delta^{18}O_2) \dots (Q_i \times \delta^{18}O_i)) / (Q_1 + Q_2 + \dots Q_i) [1]$

with where Q = rainfall quantities quantity for the day i (in mm). Daily values were not averaged since there was only one 250 rainfall sample per day resulting from the homogenization of all the event samples of from that day. Spearman's rank correlation analysis, a non-parametric measure alternative to Pearson correlation analysis, was preferred to account for nonlinear relationships, with r as-indicating the a-the correlation coefficient (PAST software, Hammer et al, 2001) (Table 3). The analyses were conducted at-on daily (Table 3A) and monthly (Table 3B) time scales. Bonferroni The Bonferroni test was applied to prevent data from incorrectly spuriously appearing to beas statistically significant by making an adjustment during comparison testing. Additionally, to integrate the both temperature effect and the rainfall amount effects, a multiple 255

3.5. Backward-trajectory and moisture uptake analysis

260 Backward-trajectory analysis was performed using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) Model (Version 4.8) (Draxler and Rolph, 2010) and following a similar methodology to Baldini et al. (2010) over a 120-24

regression model for δ^{18} O was carried out using PAST software for every studied site (Table 3C).

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	hours lifetime (Figure 5) (24-120 hours trajectories were also calculated, Fig. S1) for three of the seven stations: Oviedo,
265	Borrastre and Mallorca. Global Data Assimilation System (GDAS) have been used in Hysplit simulations with 0.5°x0.5°
	spatial resolution. All points produced by the HYSPLIT model every hour (120 points) were used to generate a vector
	representing the origin and mean trajectory of the rainfall collected. Once all the 120 vectors were produced, they were
	averaged, and one unique vector was assigned to each rainfall event. After that, all the averaged vectors associated with each
	different location studied, are presented in a compass rose using 10° intervals, together with δ^{18} O values and rainfall amount
	of each event (mm) provided by the closest weather stations to each analyzed location (Figure 5). Thus, to facilitate the
	statistical comparison of the-mean trajectory paths and moisture uptake regions with the oxygen isotope signature of sampled
270	rain events, the vector angle between every site (Oviedo-Borrastre-Mallorca) and each hourly position along 12024-h back
	trajectories (at 700 and 850 hPa) for each event was estimated, following the methodology presented in Baldini et al. (2010)
	(Figure 5). Once all the vectors that represent the mean trajectory of the air mass transport associated with the-precipitation
	were produced for each sampled event, they were averaged, and presented in a compass rose using 10° intervals, together
	with $\delta^{18}O_p$ values and rainfall amount of each daily sample (mm) provided by weather stations closed to each analyzed
275	location analyzed (Figure 5). To explore the moisture uptake along the backwards trajectories, we have performed a new
	analysis in all events (850hpa trajectories) using Baldini's method (Baldini et al., 2010) in a more restrictive way (see also
	Iglesias González, 2019), rto identify the locations where moisture uptake processes have been produced taken place during
	the 48h before the rainfall samples were collected. Taking into account that the Iberian Peninsula is surrounded by ocean,
	together with the fact that most of the analyzed rainfall events analyzed in the investigation-were produced by frontal
280	systems and convection events (see synoptic analysis), only 850-hPa air mass moisture uptake events have been considered
	as relevant togin our new analysis. In addition, while Baldini et al, (2010) considered moisture uptake processes with an
	increase in 1h-of 0.1 gH2Ov/kgeig in one hour as significant, in our analysis we only took into account events where in which
	moisture uptake process where higher than 0.25 g H ₂ O _x /kg _{air} ; therefore, if there, so if exists any influence of the rainfall
	isotopical signal, it would be easier to identify than in other-previous studies. With this restricted method, and considering all
285	the events analyzed amined, more than 3000 moisture uptake events have been identified. These events were analyzed
	considering seasonal variability and the different locations where the rainfall samples were collected (Oviedo, Borrastre,
	Mallorca)

3.6. Precipitation types

- Lastly, to better explore the role of the type of precipitation in controlling the isotopic composition of rainfall across northern
 Iberia, we applied a disaggregation procedure of <u>the precipitation series on the basis of their meteorological origin</u>, following the same <u>methodology-subjective criteria</u> described in Millan et al. (2005) (see disaggregation criteria in his Table 1). This novel-method classifies each precipitation event on the basis of its characteristics and moisture source region, distinguishing between three categories (Figure 1B, Table 4): (i) frontal systems associated with passing cold fronts from the west, (ii)
- 295 convective-orographic storms driven by differential heating, sea breezes and local winds (Azorin-Molina et al., 2009) and

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(iii) easterly advection from the Mediterranean Sea (backdoor cold fronts). The Kruskal-Wallis H test (sometimes also called the "one-way ANOVA on ranks") is a rank-based nonparametric test (Hammer et al., 2001) that was applied to the three rainfall categories to determine if there were statistically significant differences on in their δ^{18} distributions (Table 5).

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4 Results and discussion 300

This section is focused on characterizing $\delta^{18}O_n$ in the studied transect in northern Spain atom daily and monthly time scales and on analysing the main factors behing the observed patterns. It is important to highlight here the high complexity of the hydrological cycle, with many processes playing a role in the formation of the isotope signals in precipitation, ranging from 305 source processes, transport processes, as well as cloud and rainfall formation at the sampling site. Besides, tThose factors may play an overlapping role also overlap, thus making it difficult to disentangle their effects. The following sections are presented as follows: Sect. 4.1 and 4.2 are dedicated to daily and monthly data respectively. Sect 4.3 is dedicated to the influence of geographical parameters, such as distance to from coast or elevation of the studied sites. Sect. 4.4 deals with the role of meteorological parameters, in particular, local air temperature and precipitation amount. Sect. 4.5 investigates the role of moisture origin on δ^{18} O_n variability while Sect. 4.6 explores the role of rainfall type (convective, frontal) in determining $\delta^{18}O_{p}$.

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4.1 Daily scale rainfall isotopic variability in northern Iberia

The rainfall samples for the studied stations at on a daily scale_-define local meteoric water lines (LMWL) that are roughly 315 parallel for all sites with similar offset from the Global Meteoric Water Line (GMWL, $\delta^2 H = 8 \times \delta^{18} O + 10$) (Figure 2). All the slopes and the intercepts are lower than the GMWL, with slopes ranging from 6.9 to 7.2 and intercepts from 1.05 to 6.4 (Figure 2). Thus, the LMWLs determined from with-daily data for each of the studied sites reveal a broadly similar regional signal and are consistent with previous studies using GNIP data from southern France (Genty et al., 2014), even 320 considering even though that study is made with based on monthly $\delta^{18}O_n$ data. The slopes obtained in our study are slightly lower in our study compared to a previous analysis from in the IP (Araguás-Araguás and Diaz Teijeiro, 2005), in which t where the sampling period only covered the rainy season (October to March).

Despite the differential time coverage of samples among the studied fact that the data were not collected for the same time period of time at each stations, the different daily time series of $\delta^{18}O_p$ at all stations are presented together versus time 325 (Figure 3). Figures with $\delta^{18}O_p$, d-excess and total precipitation for every site separately site are included as Supplementary material (Figs. S2 to S8). From 2010 to 2017, daily $\delta^{18}O_n$ clearly show lower values in winter and higher (sometimes positive) values in summer at all stations (Figure 3). Yet some summer rainy episodes (e.g., the 25 June 2014 event in

Borrastre or the 18 June 2016 one in Barcelona) exhibit values typical of winter after raining-several days of rain or after an intense rainfall event (41.6 mm in Borrastre, rainfall amount from 23rd to 25th June, or 17.8 mm in Barcelona form the 17th 330 and to 18th June).

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Also evident in Figure 3 is the synchronicity among stations for specific events. A good example is the episode of 16-18th November 2013 (inset in Figure 3) when very negative values were reached at Molinos (black line), Borrastre (green line) and Mallorca (red line). This period was characterized by intense widespread rain - eg. 43 mm in Mallorca and 36 mm in

Molinos (Table S1). At the three sites, this period was among the rainiest of-in our record with some of the lowest $\delta^{18}O_{\rm p}$ 335 values recorded.

From thilt is evident from this s-large dataset, it becomes evident a very that there is significant day-to-day variability, as large as the seasonal cycle, thus emphasising the need offor high-resolution measurements as the ones presented in this study to characterize rainfall isotopic variability in northern Iberia.

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4.2 Monthly scale-rainfall isotopic variability in northern Iberia

Seasonality in $\delta^{18}O_n$ in northern Iberia is further explored in Figure 4B (data in Table 2, Table S2). All stations exhibit a clear seasonal pattern in temperature, with a peak in July/August and minimum values in December/January- and a similar 345 The seasonal signal in $\delta^{18}O_n$ roughly follows this pattern with peak $\delta^{18}O_n$ values in summer and minimum $\delta^{18}O_n$ in winter. It is worth noting that precipitation exhibits a bi-modal-modael pattern with peaks in Sspring and Aautumn, which is not reflected in $\delta^{18}O_p$. The average seasonal differences between the $\delta^{18}O_p$ maximum values in July-August and minimum $\delta^{18}O_p$ values in January-February are quite-large: 5.8‰ at Borrastre, 4.6‰ at Ortigosa de Cameros, 6.2‰ at Molinos and about 4‰ at Mallorca-Barcelona. Interestingly, the Oviedo-El Pindal samples reveal a very different pattern, with a marked reduction in seasonality compared to the other sites (2 $\% \delta^{18}O_n$ difference between winter and summer) (Figure 4B). The 350 seasonal difference from winter to summer in Oviedo-El Pindal is similar to the values published by Genty et al., (2014) for stations in southern France (e. g., 2.1 ‰ in Villars with only Atlantic influence, and 3.6 ‰ in Orgnac with Atlantic and Mediterranean influence). The explanation for the weak seasonality in the Oviedo-El Pindal $\delta^{18}O_{n}$ signal and the similarity withto the Villars station could be related to the origin and type of precipitation (Sect. 4.5 and 4.6). In spite. Even though there are certainly much less fewer precipitation events associated with fronts in summer than in winter, still-Oviedo and 355 Villars stations are characterized by a relatively constant source of precipitation throughout the year derived from associated with Atlantic fronts and no-an absence of a dry season (Figure 4A). This is in clear contrast to the other stations, which are characterized by a more hybrid Atlantic/Mediterranean climate (e.g., Orgnac, Genty et al., 2014). In pParticularly, in Barcelona and Mallorca the seasonal difference in $\delta^{18}O_n$ monthly values is high (6%) (Figure 4B). At these two stations, the influence of different air masses histories may be important to explain the observed high variability. These influences are further evaluated using back trajectory and moisture uptake analysis in (Sect. 4.5.).

5 Discussion

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This discussion section is focused on analysing the main factors controlling 5¹⁸O_n in the studied transect in northern Spain at daily and monthly time scales. Sect. 5.1 is dedicated to the influence of geographical parameters, such as distance to coast or elevation of the studied sites. Seet. 5.2 deals with the role of meteorological parameters, in particular, local air temperature and precipitation amount. Sect. 5.3 investigates the role of moisture origin on $\delta^{18}\Omega_{2}$ variability while Sect. 5.4 explores the role of rainfall type (convective, frontal) in determining $\delta^{18}\Theta_{n}$.

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5.14.3 Geographical controls on rainfall isotopic variability

The combination of the various isotope effects results in consistent and spatially coherent variations in $\delta^{18}O_p$ values, that are primarily related to geographical location and regional orography parameters, such as Jatitude, , longitude and plevation, 375 moisture source and air masses history (Rozanski et al., 1993; Bowen, 2008), parameters that - Then, those parameters, i.e. the latitudinal location and the regional orography. later influence the circulation and therefore the air mass history. The LMWLs determined with daily data for each of the studied sites reveal a broadly similar regional signal and are consistent vious studies using GNIP data from southern France (Genty et al., 2014), even considering that study is made monthly $\delta^{12}\Theta_{a}$ data. The slopes obtained are slightly lower in our study compared to a previous analysis from the (Araguás Araguás and Diaz Teijeiro, 2005) where the sampling period only covered the rainy season (October to March). 380 Initial Preliminary observation of the monthly averaged $\delta^{18}O_n$ data (Table 2 and Figure 4B) reveals similar values The comparison of monthly averaged $\delta^{13}\Theta_{a}$ values in the studied stations allows an assessment of the relative importance of geographical factors in the observed patterns (Table 2, Figure 4B). Ortigosa de Cameros, Molinos and Borrastre stations show monthly 8¹⁸O_a values guite similar and, normally, more negative than Oviedo, El Pindal, Barcelona and Mallorca sites. This pattern is particularly clear for autumn values (see monthly averaged or setting a setting to December in 385 Table2 along the west to east transect). The similarity found among the sites located at opposite ends of the transect, that is, (Oviedo and El Pindal compared to Barcelona and Mallorca). This similarity and, presumably, influenced by different air masses with different isotopic composition in the initial water vapor (i.e., Atlantic vs Mediterranean) as previously described

390 expected since differences in moisture source conditions at the location of the moisture uptake were supposed anticipated to be markedly different (see also Sect. 4.5). However, their is similarity This pattern may be explained by two processes, partly associated to the geographical location of the studied sites.

First, the fact that Oviedo and El Pindal rainfall samples show enriched $\delta^{18}O_p$ values (Table 2, Figure 4B) is consistent with their location in the Cantabrian coast, very close to the Atlantic Ocean, with climatological oceanic climatological conditions

by LeGrande and Schmidt (2006) in their global study of oxygen isotopic composition in seawater, is not what w was une

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- characterized by high-relatively mild mean-temperatures (Table 1). Thus, Oviedo (and El Pindal) are the stations that receive the first precipitation produced by-via contact with the Atlantic-air masses; therefore they are the stations in the transect least affected by the "continental effect"; which occurs -when moist air and clouds move inland from the Atlantic Ocean and become gradually isotopically depleted due to progressive rainout (Dansgaard, 1964). Thus Therefore, as we follow the typical movement-displacement of an Atlantic front on its way to the IP, from west to east, we find progressively more negative winter $\delta^{18}O_p$ values (see values when considering an average of for January-February-March; in (Table 2), -going
- from El Pindal (-6.0‰) to Ortigosa de Cameros (-8.1‰), to Borrastre (-9.8‰) and, finally, to Molinos (-10.0‰). This pattern is not so-as evident in other seasons where the entrance of Atlantic fronts is not the only maindominant synoptic pattern-situation that generates rainfall in the transect_(Rüdisühli et al., 2020). However, these large observed differences cannot be explained only by In addition to this "continental effect" that, which accounts for only a very smallsmall gradient
- 405 variation in δ¹⁸O_p-(about 0.002‰ per km in Europe as described in Rozanski et al., 1993)<u>,-Additionally, the higher annual</u> mean air temperature at those stations (in both Mallorca and -and-Barcelona, on the one hand, together withand Oviedo and El Pindal, on the other.) compared to the other onesstations (Table 1), may help to explain their similar A second factor to explain why Mallorca and Barcelona rainfall samples display the least negative δ¹⁸O_p values. -in the
- transect, is the influence of air masses derived from the Mediterranean Sea. Initial water vapour 8¹⁸O values in the
 Mediterranean Sea are typically more positive (0.5 1 ‰) than Atlantic Ocean water vapour due to enhanced evaporation in within the semi-enclosed Mediterranean basin (LeGrande and Schmidt, 2006). An additional effect, probably more important than 0.5 1 ‰ of difference, is the higher annual mean air temperature at those stations (Mallorca and Barcelona together with Oviedo and El Pindal) compare to the other ones (Table 1). The effect of of warmer temperature to onproduce the less negative 8¹⁸O_p values recorded will be explained-discussed with more detail below (Sect. 54.24).
- 415 Second, another geographical factor tothat could -account for the similarities found among Oviedo El Pindal and Barcelona-Malloreain stations at opposed ends of the transect is related to the elevation of those sites. In addition, the three stations with more negative monthly $\delta^{18}O_p$ values (Ortigosa de Cameros, Borrastre and Molinos) are at higher elevation than the other stations that are all located close to sea level. Ortigosa de Cameros, Molinos and Borrastre stations, the three stations located at a higher elevation, show monthly $\delta^{18}O_p$ values quite similar and, normally, more negative than Oviedo, El Pindal,
- 420 Barcelona and Mallorca sites, the siteswhich are at low elevation, close to sea-level. This pattern is particularly clear for autumn values (see monthly averaged $\delta^{18}O_p$ values from September to December in Table2 along the west-to-east transect). Therefore, the "elevation effect" (Siegenthaler and Oeschger, 1980) likely also plays a role in explaining the more negative $\delta^{18}O_p$ values at those stations. Considering the $\delta^{18}O_p$ annual averages (Table 2), there is a difference of 2.3% between Molinos (1040 m asl) and Mallorca (90 m asl). Based on the difference of elevation, the vertical isotopic gradient observed is
- -0.24‰ per 100 m of elevation. This result is consistent with previous studies in other mountain ranges such as the Alps, where an altitudinal gradient of -0.2 to 0.3‰ per 100 m of elevation was observed (Ambach et al., 1968; Siegenthaler and Oeschger, 1980). However, in spite the difference in elevation, we need to consider that the sites are very distant and

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separated by the Mediterranean Sea. Therefore, the altitude cannot be the only parameter controlling the differences between the studied sites.

- 430 In the next sections we explore how Finally, the geographical factors reviewed in this section (distance to the coast or continental effect and $\frac{1}{2}$ elevation effect. and $\frac{1}{2}$ composition of the sea waters) exert a small direct influence on the observed spatial distribution of rainfall $\delta^{18}O_n$ at the studied sites by , specially by their contribution but contribute to the effects of influence on other, controlling factors: air temperature, rainfall amount, air mass trajectory origin and rainfall type. , which will be described in following Sect. 5.24.4, 54.5.3 and 5.4and 4.6.
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5.24.4 The influence of air temperature and rain amount on the spatial distribution of rainfall $\delta^{18}O_{D}$ values at daily and monthly time scales

Spearman's rank correlation analysis (Table 3) reveals that $\delta^{18}O_p$ does not correlate with air temperature or amount of precipitation in a similar way at each station, neither at daily nor monthly time scales, thus reinforcing the need for 440 conducting such calibrating studies on a local basis, particularly when conducting palaeoelimate palaeoelimate reconstructions (Leng, 2006). Air temperature appears as the most robust influence variable better correlated with $\delta^{18}O_n$ across the west-to-east transect, with low-modest but statistically significant correlations (daily scale) with $\delta^{18}\Theta_{p}$ at all sites (red numbers in Table 3A) except Oviedo and Barcelona, most likely due to the low number of daily samples (n=39 and 445 n=53, respectively). The coefficient of correlation among $\delta^{18}O_p$ daily values and air temperature is highly variably from west

- to east: El Pindal ($r_s = 0.34$; p = 0.0012), Ortigosa de Cameros ($r_s = 0.25$; p = 0.001), Molinos ($r_s = 0.42$; p = < 2.00E110.001), Borrastre ($r_s = 0.29$; $p = \frac{6.33E \ 0.80.01}{2}$ and Mallorca ($r_s = 0.35$; p = 0.0013) (Table 3A). Regarding monthly values, air temperature is significantly correlated with $\delta^{18}O_p$ values at eastern stations, with the highest coefficients associated with higher altitude sites (e.g., in Molinos with $r_s = 0.76$ and p=3.36E-10<0.001 or in Borrastre with $r_s = 0.61$ and 450 $p < 0.001 = 1.44 E \cdot 05$) (Table 3B).

The dependence of ⁵¹⁸O_n on air temperature has been extensively studied, yielding an *average* slope for mid latitude continental stations of 0.58%/°C (Rozanski et al., 1993). HoweverIn our data, that value is highly variable in time and space The strongest air temperature $\delta^{13}\Theta_{a}$ relationship, based on daily data, is found at the Pyrenean station. Borrastre site, with 0.4%/°C and the weakest at Oviedo+El Pindal (0.2%/°C). The other three stations. Ortigosa de Cameros (0.3%/°C). Molinos (0.4%/°C) and Mallorea+Barcelona (0.3%/°C), show similar intermediate values. Compared to other areas, such as the Alps with temperature δ^{48} Q₄-gradients of 0.5 to 0.7 ‰ per °C, the results presented above indicate that, although important, air temperature only explains between 20 and 40 % of the observed 8¹⁸O_e variability and is, therefore, not the only

control. The influence of rainfall amount on $\delta^{18}O_p$ effect is dominant is strong in tropical regions where deep vertical convection is

common-frequent, although it may also occur in the extratropics in summer (Bar-Matthews et al., 2003; Treble et al., 2005b). 460

Correlation among rainfall amount and $\delta^{18}O_p$ is negative, associated to the raindrop evaporation during periods of sparse rains (Dansgaard, 1964; Risi et al., 2008) when the relative humidity is low and is thus not a purely equilibrium process. In the studied transect, at the daily scale, the strongest correlation with amount of precipitation is observed in Barcelona ($r_s = -0.35$; p=0.029) (Table 3A). Besides, there There is also is a significant correlation at the two sites of the Iberian Range ($r_s = -0.35$; p=0.029) (Table 3A).

- 465 0.32; p=1.05E-05 in Ortigosa and $r_s = -0.19$; p=0.005 in Molinos). Interestingly, the westernmost stations (El Pindal and Oviedo) do not show a significant $\delta^{18}O_p$ -precipitation correlation at theon daily nor monthly scales. This lack of a correlation in the Atlantic sites (El Pindal and Oviedo) contrasts with a previous study carried out in northern Spain and at a site also characterized by an Atlantic climate (Matienzo depression), where there is found a significant $\delta^{18}O_p$ -precipitation monthly correlation $\frac{1}{18}$ was found (r = -0.51; p < 0.01) (Smith et al., 2016). In our study, the $\delta^{18}O_p$ -precipitation correlation at
- 470 monthly scale is only significant in Molinos, in the Iberian Range ($r_s = -0.4$; p=0.018), -while no correlation is observed in-<u>at</u> the other sites (Table 3B).

To further assess the relative role of temperature and <u>rainfall</u> amount effects, a multiple regression model for δ¹⁸O_p was carried out for the seven studied sites in which the temperature effect exerted a clear dominant control (Table 3C). Still, b The dependence of δ¹⁸O_p on air temperature has been extensively studied, yielding an *average* slope for mid-latitude continental stations of 0.58‰/°C (Rozanski et al., 1993). In our data that value is highly variable in time and space. The strongest air temperature-δ¹⁸O_p relationship, based on daily data, are found at Borrastre (0.4‰/°C), Molinos (0.4‰/°C) and Barcelona (0.37‰/°C) while the weakest at Oviedo/El Pindal (0.2‰/°C). The other three stations, Ortigosa de Cameros (0.25‰/°C) and Mallorca (0.317‰/°C), show intermediate values. Compared to other areas, such as the Alps with temperature-δ¹⁸O_p gradients of 0.5 to 0.7 ‰ per °C, the results presented above indicate that, although important, air temperature only explains part of the observed δ¹⁸O_p variability and is, therefore, not the only control. Temperature, together

with amount of precipitation (Table 3C) Both influences together account for less than 20-40 % of the variability of $\delta^{18}O_p$ in the study transect. Since the origin of air masses producing of rainfall and the type of rainfall (i.e., convective vs. frontal) is also spatially dependent in northern Iberia, these variables and their influence on the observed $\delta^{18}O_p$ variability are investigated in Sect. 5.34.5 and 5.44.6 below.

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$\frac{5.34.5}{5.34.5}$ The role of the *source effect* in modulating northern Iberian Peninsula $\delta^{18}O_p$

The source effect describes how air masses derived from different moisture sources have distinct δ¹⁸O_p values (e.g., Friedman, 2002). The source effect results from varying air mass histories, different conditions of the moisture source (temperature, relative humidity and wind speed) and regional differences in the δ¹⁸O of the surface ocean (LeGrande and Schmidt, 2006). In the case of northern IP, it is necessary to consider the effect of both the Atlantic Ocean and Mediterranean Sea as important sources of atmospheric moisture (Gimeno et al., 2010). Their -whose-relative influence on regional IP δ¹⁸O_p may exhibit highcould be very difference spatial variability-different because of the complex regional

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- topography of the arealP. General Typical present-day δ^{18} O values of seawater reconstructions (LeGrande and Schmidt, 2006) indicate slightly different values for the Atlantic Ocean and the Mediterranean Sea, due to temperature and salinity differences. Source δ^{18} O values range from 1 to 1.5% in the subtropical Atlantic to 2% in the Mediterranean (Schmidt et al., 1999), Although t These differences in the source (about 0.5 - 1 %) are small, since but they are further modulated by the air mass history, we expect to see thus resulting in a change in the relative influence of moisture sources on $\delta^{18}O_n$ along the westto-east transect.
- Evaluation of monthly 1400, patterns represented in Figure 4B reveals more muted seasonality (2%) at Oviedo El Pindal 500 sites compared to other stations in the transect (> 4‰). The seasonal difference from winter to summer in Oviedo is similar alues published by Genty et al., (2014) for stations in southern France (e. g., 2.1 ‰ in Villars with only Atlantic influence and 3.6 % in Orgnac with Atlantic and Mediterranean influence). The explanation for the weak seasonality in the Θ_{a} signal and the similarity with Villars station could be related to the precipitation type (Sect. 5.4) and the
- 505 aphic origin. Oviedo and Villars stations are characterized by a relatively constant source of precipitation through the year derived from Atlantic fronts and no dry season (Figure 4A). This is in clear contrast to the other stations which are characterized by a more hybrid Atlantic/Mediterranean climate (e.g., Orgnac, Genty et al., 2014), Particularly, in Barcelona wasonal difference in ⊟¹⁸Q, monthly values is high (6‰) (Figure 4B). At these two stations, the influence of different rainfall sources (Atlantic and Mediterranean) with distinct isotopic values as demonstrated by a global study of 510 in surface oceans (LeGrande and Schmidt, 2006) and different air masses histories may be important to explain
- the high variability. These influences are further evaluated using back trajectory analysis.

To evaluate the role of moisture sourceair mass origin in determining $\delta_{i}^{18}O_{p}$ values at a daily scale in northern Iberia and Balearic islands, back trajectories were calculated for all the rainy days and subsequently averaged into wind rose diagrams, following the representation applied-used in previous studies (Smith et al., 2016), for three stations along our northern Iberia

515 transect: Oviedo and Mallorca, the two extreme locations-of the studied-transect, and Borrastre, situated-located at an intermediate location, representing comprising, a total number of 519 events (Figure 5). To facilitate statistical comparison of the mean trajectory paths and moisture uptake regions with the oxygen isotope signature of sampled rain events angle between every site (Oviedo Borrastre Mallorea) and each hourly position along 250 hPa) for wing the methodology presented in Baldini et al.

values and rainfall amount of each daily together with sample (mm) provided by location analyzed (Figure 5).

event, they were averaged, and presented

This analysis reveals the dominance of western trajectories atim the three studied sites considering 24h life-time, with very few episodes associated with other a different directions (Figure 5, see also Fig. S1 for trajectories calculated over a 120h life

time). In fact, comparison among the study with the analysis, carried out over 24h and over 120h shows Oonly some few, 525 episodes from-with trajectories from the SW (e.g., Borrastre) or SE (e.g., Mallorca) trajectories are found-and, interestingly, Con formato: Fuente: (Predeterminado) +Títulos (Times New Roman)

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they all have distinct-different $\delta_{l}^{18}O_{p}$ values (see below). This low, almost negligible, presence of trajectories associated with Mediterranean air mass advections, does not inhibit the possibility of a moisture uptake over the Mediterranean or moisture recycling with altitude in the mountain region surrounding Borrastre since meteorological processes connected to convection

- 530 (e.g., orographic, dynamic, thermal) can produce moisture uptake in less than 6h (Romero et al., 2000, 1997; Tudurí and Ramis, 1997) and may not be well captured in the back trajectory analyses, which are computed for the previous 120 hours (see Methods). Therefore, convection processes, that may be associated with casterly trajectories, are under represented in this methodology (see 24 hours analyses in Figure S1 where more trajectories with different origin appear more frequently). Therefore, it is important to note here that this method provides information on the air mass origin (source effect) but not in
- 535 the moisture uptake regions. In that way, it is clear the dominant WNW trajectory for the three studied stations. Despite the three sites sharing a common dominant WNW <u>direction of Atlantic trajectoryair mass origin</u>, <u>that is Atlantic</u>, they behave quite differently in terms of the associated amount of rainfall and $\delta^{18}O_p$ values for every event. Oviedo (with a temperate oceanic climate - Cfb, Table 1) presents a narrower range of <u>in both</u> rainfall amounts and $\delta^{18}O_p$ values thanin comparison to at the other two sites (clearly seen at 120h, Fig. S1), as shown in Figure 5A by the negligible frequency of
- 540 rainfall amounts above 32 mm (orange) or below 2 mm (purple), while "extreme" events are much more common in Borrastre or Mallorca sites. Similarly, in figure 5B, where the isotopic values for the different trajectories are plotted, Oviedo appears as the station with more stable<u>more homogenous</u>uniform $\delta^{18}O_p$ values ($\delta^{48}O_p$ values <u>among(between</u> -10 and -2‰) compared to the other two<u>stations</u>. Thus, in Borrastre and Mallorca, $\delta^{18}O_p$ values between -8 and -12‰ (red – green – yellow - dark blue) are only present in northwestern trajectories, while less negative values (- 6 to 2‰) appear in events with
- 545a_SW and SE directions (see also Fig. S1). These results confirm the homogeneity of the Atlantic sites in terms of $\delta^{18}O_p$ amplitude (Cantabrian coastal sites: Oviedo, El Pindal) compared to the intermediate (Iberian range and Pyrenean sites:
Ortigosa de Cameros, Molinos and Borrastre) stations already described by monthly data in Figure 4.

These results highlight the importance of moisture source in generating the observed $\delta^{18}\Theta_{p}$ differences along the west to east transect in this study. At Borrastre (our mid transect site) two mean trajectories are distinguished in terms of $\delta^{18}\Theta_{p}$ values:

- 550 northwesterly trajectory associated with more negative $\delta^{18}\Theta_{p}$ -values and southwesterly trajectory associated with less negative values (Figure 5B). Borrastre station is chosen to further evaluate back trajectories for all rainfall events over one whole year (2014, n=126 rainfall events) since the presence of rainfall events where moisture comes from the SW, with usually less negative $\delta^{18}\Theta_{p}$ -values, is significant compared to, for example, Oviedo station. Thus, one example from every trajectory is presented in Figure 6. This study of the source origin indicates a low, almost negligible, presence of trajectories
- 555 associated with Mediterranean air mass advections. However, it is well-known that, since meteorological processes connected to convection (e.g., orographic, dynamic and; thermodynamical) can produce moisture uptake in less than 6h (Romero et al., 2000, 1997; Tudurí and Ramis, 1997) they won'twill not be well-captured in this-back trajectory analyses which that are computed for the previous 24 hours (and and even less for 120h, Fig.S1). Therefore, it is important to note here

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that this method provides information on the air mass origin (source effect) but not oin the moisture uptake regions. In that way, it is clear the dominant WNW air mass origin for the three studied stations is clear.

- To account for the different moisture uptake processes along the studied trajectories, we followed Baldini et al. (2010) methodology in a more restricted way (see Methods) for Oviedo-Borrastre-Mallorca stations (Figure 6). These results Our findings highlight the importance of moisture uptake in generating the observed $\delta^{18}O_{n}$ differences along the our west-to-east transect-in this study, with Oviedo and Mallorca showing a clearly dominant marine signal (Atlantic and Mediterranean, 565 respectively), -while in Borrastre- the moisture uptake occurs over the whole Iberian Peninsula with less importance of thethe marine uptake being less important. The Oviedo station appears very homogenous in terms of the moisture uptake regions, clearly focused concentrated on the Portuguese margin and with very few events characterized by recycled continental moisture. Interestingly, we observe seasonal differences in the dominant regions for moisture uptake, particularly relevant at Borrastre station. Thus, the contribution of continental moisture recycling, although observed along the whole year, is more 570 frequent in summer, as deduced from the red dots located very close to the station, pointing -which point to local convective processes (Figure 6). The oceanic contribution to Borrastre rainfall is reduced-small and focused present mostly in winter (WNW oceanic source) and, with-to a lesserless extent, in spring and autumn (Mediterranean source). A recent study analyzing the trace element composition of precipitation also shows this seasonal tendency for the Pyrenees (Suess et al., 2020). This seasonality in land vs. ocean moisture source contribution to Pyrenean precipitation is most certainly an 575 important driver of the seasonal cycle of the isotope signals in precipitation, as was previously highlighted in many stations in Figure 4B. The moisture uptake regions identified observed for Mallorca rainfall events are also quite heterogeneous, highlighting the Mediterranean as the dominant source for moisture uptake, while in winter-spring some events are observed from theto originate in the WNW sector. Additionally, some southern events arriving from NorthN Africa are also detected, indicating the importance of that area to account for the Balearic islands rainfall composition. In addition, we suggest that Borrastre station as offersis a good representation of the Iberian rainfall in terms of $\delta^{18}O_n$ composition since it receives 580 moisture from a largewide area, thus being of utility for further palaeoelimatic paleoelimatic studies in the Pyrenean region. Similarly, Oviedo site appears as at be a good exponent-prototypeof for the Atlantic region, allowing closer-nearby palaeoclimate palaeoclimate sites to be compared with well-known marine cores from the Portuguese margin. F-and, finally, Mallorca site would represent the Western Mediterranean in terms of preferred moisture uptake region.
- 585 Above 80% of winter trajectories recorded in Borrastre rainfall events originate in the North Atlantic, Artic or inland USA or Canada. They cross the Atlantic Ocean north of Madeira Island and usually enter the IP by the west, next to the Galicia and Portugal border. Those trajectories arriving from the N NW reach Borrastre site at the Pyrenees almost without crossing the IP, thus providing the more negative $\delta^{18}O_p$ values (e.g., 7th February, Figure 6A, with $\delta^{18}O_p = 6.5\%$). On the contrary, those arriving from the W SW enter via Lisbon and cross central IP providing less negative $\delta^{18}O_p$ values (e.g., 16th January, Figure
- 590 6B, with $\delta^{18}O_p = 1.2\%$). If the trajectory of the air mass travels larger distances over the continent, the contribution of reevaporated land moisture to the water vapour travelling inland may be significant and thus $\delta^{18}O_p$ values may appear higher,

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as it has been shown to occur in other regions (Krklec and Domínguez Villar, 2014). In that case, the progressive rainout effect may be compensated by the moisture uptake of evaporated (high δ¹⁸O) surface water.

During spring, the typical situation of air masses entering from the W alternates with those arriving from the SW, entering at the latitude of the Cape San Vicente and crossing the IP from south to north (e.g., 20th April, Figure 6C; with δ¹⁸O_p = 2.1‰). Some spring trajectories are subject to Mediterranean influence (eg. 20th May; Figure 6D) and are characterized by higher δ¹⁸O_p, values (δ¹⁸O_p = 1.3‰). In general, the penetration of subtropical Atlantic air masses, which becomes a very common situation in summer, results in higher δ¹⁸O_p, values (e.g., 6th July, Figure 6E, with δ¹⁸O_p = 2.2‰). Therefore, the less negative δ¹⁸O_p, values usually associated with SW trajectories in Borrastre can be explained by (1) the origin in the subtropical Atlantic Ocean with higher δ¹⁸O_p, values (1.5‰) compared to North Atlantic (0.5‰) (LeGrande and Schmidt, 2006) and, (2) the recycling of surface moisture over land incorporating enriched δ¹⁸O_p, values from surface waters that have been subject to evanoration over time (Krklec and Domínguez Villar, 2014).

5.4<u>4.6</u> The influence of rainfall type on isotopes.

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The influence of rainfall type on the $\delta^{18}O_p$ is well documented globally, with different $\delta^{18}O_p$ observed depending <u>on</u> the type of precipitation: ___(convective showers, frontal, continuous stratiform precipitation, etc.) (Aggarwal et al., 2012). This relationship-dependency is observed in previous studies both <u>at-on</u> daily or monthly timescales (Aggarwal et al., 2016), with few examples <u>in-of frontal precipitation in Europe-(Aemisegger et al., 2015) or tropical convective processes (Risi et al., 2016)</u>.

- 2008) in Europethe Equatorial Indian Ocean (Gat, 1996) and California, USA (Coplen et al., 2015), _both indicatingTheose previous studies indicated that δ¹⁸O_p values were lower when precipitation was dominantly stratiform and higher when it was mostly convective; ____TtfThe main reason to explain this difference lies_lyingies_on the processes of condensation and riming associated with boundary layer moisture which produced_higher isotope ratios in convective rain (Aggarwal et al., 2016).
 Additionally, Some-some studies in the Mediterranean region (Celle-Jeanton et al., 2001; Lee et al., 2019) also directly link
 the isotopic signature of the precipitation to the prevailing weather conditions during the rainfall event. (Celle-Jeanton et al., 2016)
- 2001), helping to further understand the role of water vapour transport and the moisture cycling during convective events (Lee et al., 2019).

Here we explore how the specific synoptic situation, i.e., rainfall types or rainfall components, influence $\delta^{18}O_p$ values across the studied transect. Table 4 shows the percentage of rain events associated with each type of precipitation, which that were

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the studied transect. Table 4 shows the percentage of rain events associated with each type of precipitation, which that were previously defined following (Millán et al., 2005), and represented in Figure 1B: (i) Atlantic frontal systems (westerly winds), (ii) convective–orographic storms, and (iii) backdoor cold fronts from the Mediterranean Sea (easterly winds). Backdoor cold fronts from the Mediterranean Sea are sporadic events occurring in autumn (secondarily in winter spring), but they eause heavy precipitation and flooding (Llasat et al., 2007). Con formato: Fuente: (Predeterminado) +Títulos (Times New Roman)

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The prominence of rainfall associated with Atlantic fronts is evident (above 40% in the seven studied stations). This 625 percentage decreases eastward, from 68-09/71.29-% in Oviedo/El Pindal to 58-49/40.8241 % in Barcelona/Mallorca. A previous study at a north Iberian site (Matienzo, Cantabria) indicates that approximately 80% of air masses originate in the North Atlantic, and that their movement is associated with westerly frontal systems (Smith et al., 2016). This situation appears to be true along the studied transectfor the Cantabrian coastal sites; - however for the Mediterranean and Iberian Range sites, however, the Atlantic and Mediterranean sources are balanced of comparable importance (including backdoor 630 cold fronts-as Mediterranean) (Table 4). Distance to the Mediterranean and elevation are also important factors in determining the frequency of rainfall associated with backdoor cold fronts. Thus, backdoor cold fronts are associated with $\frac{38.7839\%}{38.7839\%}$ of Mallorca rain events and are still frequent situations at the two sites from the Iberian Range ($\frac{20.621\%}{20.621\%}$ in Ortigosa de Cameros and 23.924% in Molinos). The frequency of convective precipitation is higher at the three mountain sites ($\frac{20.621\%}{100}$ in Ortigosa de Cameros, $24\frac{3}{2}\%$ in Molinos and 23% in Borrastre), compared to those sites at lower elevation 635 (17% in Oviedo; 11.912% in El Pindal, 17% in Barcelona and 20.4% in Mallorca).

The Kruskal-Wallis test was applied to investigate if there were significant differences in the $\delta^{18}O_p$ values of the three rainfall types analysed (Atlantic, backdoor frontal precipitation, and convective) in the studied stations at the daily scale. Test values shown in Table 5 (p values < 0.05) indicate the $\delta^{18}O_n$ values of at least two of the three rainfall types are significantly different (this does not apply for-to Oviedo and Barcelona since the number of degrees of freedom freedom

- 640 degrees isare too low-small to yield a significant result). Thus We conclude that, this means that the type of rainfall (frontal versus convective) is an important factor controlling $\delta^{18}O_p$ values in the studied transect at <u>the</u> daily scale. This result is also evident where the three rainfall types are represented according to their $\delta^{18}O_p$ composition (Figure 7). Thus, regarding $\delta^{18}O_p$ composition, convective precipitation (in green in Figure 7) is associated with the highest $\delta^{18}O_p$ values, while events related to Atlantic and backdoor cold fronts display more negative $\delta^{18}O_p$ values (albeit with a large spread), consistent with previous
- studies (Aggarwal et al., 2016). The highest $\delta^{18}O_p$ values associated with convective precipitation may be related to the 645 critical role played by the re-evaporation of droplets, a circumstance-process that usually takes place during convective rainfall (Bony et al., 2008). In any case, what is relevant here, is the similarity among between $\delta^{18}O_p$ values of associated with the two types of frontal rains (Atlantic fronts and Mediterranean backdoor cold fronts), -while there is a significant difference when considering the type of precipitation, i.e. convective versus frontal.
- Besides 8¹⁸O₄ values associated with the three rainfall types, variations of air temperature and precipitation have an effect in 650 separating the three rainfall types (Figure 7). Regarding air temperature, backdoor cold front events are the ones occurring with colder temperatures while convective rains are more associated with the warm season. Thus, air temperature (and its variation along a vertical profile) is another variable clearly associated with the type of rainfall, with higher temperature during convective rains and lower for the Atlantic and backdoor types. This is a clear reflection of the seasonal pattern of
- 655 eonvective rains, which are more abundant in summer months (Table S1) thus preventing an isolation of the effect of the of rainfall which appears mixed with the temperature effect. In contrast, the high number of outliers in the box plots of

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the amount of precipitation when organized by rainfall type (Figure 7) indicates that this parameter is determined more by local factors (e.g., topography) than by the specific synoptic situation.

660 5 Conclusion

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The major findings in this study are summarized as follows:

- The analysis of δ¹⁸O_p and δ²H_p at seven stations along a west-to-east transect in northern Iberia and Balearic Islands yields similar LMWLs₂-but all with lower slope and intercept values than the GMWL.
- Oviedo/El Pindal and Mallorca/Barcelona rainfall samples display the least negative δ¹⁸O_p and δ²H_p values in the transect. Our results suggest that this similarity in offbetween the two stations located at the far-most-western and eastern sides-ends of the northern IP the two opposite stations (the westernmost ones and the easternmost ones) are is due to,due, in the first case,firstly, to the initial condensate of water vapour generated over the North Atlantic and, in the second casesecondly, to the influence of air masses originating in the Mediterranean Sea, together with the much warmer temperatures there than in the other three sites. Besides those effects, the "elevation effect" must be taken into account to explain the more negative average values at the three mid-transect stations (Ortigosa de Cameros, Borrastre and Molinos).
 - The seasonal variability is larger at Ortigosa de Cameros, Borrastre and Molinos, while in Oviedo El Pindal it is is reduced in Oviedo-El Pindal due to the single origin of rainfall in that area and the concentrated regions of moisture uptake along the Portuguese margin.
 - Air temperature appears to be the most significant influence onbest correlated variable with δ¹⁸O_p onat daily and monthly time_scales_a with the highest air temperature-δ¹⁸O_p dependency found for at the Pyrenean station (slope of 0.38‰/°C)., while oOnly a few sites in the transect show a significant negative correlation with precipitation amount (monthly in Molinos; daily in Ortigosa de Cameros, Molinos, Barcelona and Mallorca)-with precipitation amount.
 - The dominance of rainfall with an Atlantic origin is clear in the study of rainfall back trajectories associated with each <u>rainy</u> event <u>analysed_in Oviedo</u>, Borrastre and Mallorca sites, but the <u>regions where in which moisture uptake</u> takes place are highly heterogeneous in space and time. At Borrastre station, moisture comes from all over the wholethe Iberian Peninsula, with a dominance oft recycled continental moisture <u>in summer</u>, and less effectinfluence of oceanic moisture in summer (Atlantic influence in winter, Mediterranean influence in spring and autumn). Additionally, the distance travelled inland in a quite dry region also conditions the recycling of re-evaporated moisture providing final enriched 5¹⁸O_n values.
 - Convective rainfall yields higher $\delta^{18}O_p$ values, while rainfall events related to Atlantic and backdoor fronts display exhibit more negative $\delta^{18}O_p$ values.

690 In conclusion, the northern Iberian region, is under the influence of two climatic regimes (Atlantic and Mediterranean) and affected by different moisture sources origins and uptake regions. Therefore, the synoptic-scale atmospheric circulation is playing a key role in determining the ranges, values and seasonal distribution of δ^{18} O_n variability. Future detailed studies focusing on particular events that can be traced along the whole west to east transect will be conducted to further understand the air masses trajectories over northern Spain and their influence on $\delta^{18}\Theta_{a}$ variability.

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Data availability

All data are included in the Supplementary Tables S1 and S2.

700 Author contribution

The paper was conceived by AM, CPM, MB, CS, HS and IC. MI carried out the back trajectory and moisture uptake study studies and CAM provided the synoptic patterns during rainfall days. JF, CO, ArM and ADH contributed to rainfall sampling and/or isotopic analyses. IB and FV helped with data interpretation. All authors contributed to the writing of the paper.

Competing interests

The authors declare that they have no conflict of interest

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- 720 www.meteo4u.com. www.meteoclimatic.net. http://www.larioja.org/emergencias-112/es/meteorologia

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2020 respectively.

References

AEMET, Instituto de Meteorologia de Portugal, 2011. Atlas climático Ibérico. Temperatura del aire y precipitación (1971-2000).

- 730 Aemisegger, F., Spiegel, J.K., Pfahl, S., Sodemann, H., Eugster, W., Wernli, H., 2015. Isotope meteorology of cold front passages: A case study combining observations and modeling. Geophysical Research Letters 42, 5652–5660. https://doi.org/10.1002/2015GL063988
 - Aggarwal, Alduchov Oleg A., Froehlich Klaus O., Araguas-Araguas Luis J., Sturchio Neil C., Kurita Naoyuki, 2012. Stable isotopes in global precipitation: A unified interpretation based on atmospheric moisture residence time. Geophysical Research Letters 39. https://doi.org/10.1029/2012GL051937
- Aggarwal, P.K., Romatschke, U., Araguas-Araguas, L., Belachew, D., Longstaffe, F.J., Berg, P., Schumacher, C., Funk, A., 2016. Proportions of convective and stratiform precipitation revealed in water isotope ratios. Nature Geoscience 9, 624–629. https://doi.org/10.1038/ngeo2739
- Ambach, W., Dansgaard, W., Eisner, H., Møller, J., 1968. The altitude effect on the isotopic composition of precipitation and glacier ice in the Alps. Tellus 20, 595–600. https://doi.org/10.3402/tellusa.v20i4.10040
- Araguás-Araguás, L.J., Diaz Teijeiro, M.F., 2005. Isotope composition of precipitation and water vapour in the Iberian Peninsula, in: Isotopic Composition of Precipitation in the Mediterranean Basin in Relation to Air Circulation Patterns and Climate. Vienna, Austria, pp. 173–191.
- Archer, C.L., Caldeira, K., 2008. Historical trends in the jet streams. Geophys. Res. Lett. 35, L08803.
- 745 https://doi.org/10.1029/2008GL033614
 - Azorin-Molina, C., Chen, D., Tijm, S., Baldi, M., 2011. A multi-year study of sea breezes in a Mediterranean coastal site: Alicante (Spain). International Journal of Climatology 31, 468–486. https://doi.org/10.1002/joc.2064
- Azorin-Molina, C., Connell, B.H., Baena-Calatrava, R., 2009. Sea-Breeze Convergence Zones from AVHRR over the Iberian Mediterranean Area and the Isle of Mallorca, Spain. J. Appl. Meteor. Climatol. 48, 2069–2085. https://doi.org/10.1175/2009JAMC2141.1
 - Baldini, L.M., McDermott, F., Baldini, J.U.L., Fischer, M.J., Möllhoff, M., 2010. An investigation of the controls on Irish precipitation δ18O values on monthly and event timescales. Clim Dyn 35, 977–993. https://doi.org/10.1007/s00382-010-0774-6
- 755 Bar-Matthews, M., Ayalon, A., Gilmour, M.A., Matthews, A., Hawkesworth, C.J., 2003. Sea-land oxygen isotopic relationships from planktonic foraminifera and speleothems in the Eastern Mediterranean region and their implication for paleorainfall during interglacial intervals. Geochimica et Cosmochimica Acta 67, 3181–3199.
- Bartolomé, M., Moreno, A., Sancho, C., Stoll, H.M., Cacho, I., Spötl, C., Belmonte, Á., Edwards, R.L., Cheng, H., Hellstrom, J.C., 2015. Hydrological change in Southern Europe responding to increasing North

Con formato: Inglés (Estados Unidos)

Código de campo cambiado Con formato: Inglés (Estados Unidos)

	Atlantic overturning during Greenland Stadial 1. PNAS 112, 6568–6572.		
	https://doi.org/10.1073/pnas.1503990112 Berrisford, P., Dee, D.P., Fielding, M., Fuentes, M., Kallberg, P.W., Kobayashi, S., Uppala, S., 2009. The ERA-		
	Interim archive (No. 1), ERA Report Series, ECMWF.		
765	Bony, S., Risi, C., Vimeux, F., 2008. Influence of convective processes on the isotopic composition (δ 180 and		
100	δD) of precipitation and water vapor in the tropics: 1. Radiative-convective equilibrium and Tropical		
	Ocean–Global Atmosphere–Coupled Ocean-Atmosphere Response Experiment (TOGA-COARE)		
	simulations. Journal of Geophysical Research: Atmospheres 113. https://doi.org/10.1029/2008JD009942		
	Bowen, G.J., 2008. Spatial analysis of the intra-annual variation of precipitation isotope ratios and its		
770	climatological corollaries. Journal of Geophysical Research 113. https://doi.org/10.1029/2007JD009295		
	Coleman, M.L., Shepherd, T.J., Durham, J.J., Rouse, J.E., Moore, G.R., 1982. Reduction of water with zinc for		
	hydrogen isotope analysis. Anal. Chem. 54, 993-995. https://doi.org/10.1021/ac00243a035		
	Craig, H., 1961. Isotopic Variations in Meteoric Waters. Science 133, 1702–1703.		
	https://doi.org/10.1126/science.133.3465.1702		
775	Dansgaard, W., 1964. Stable isotopes in precipitation. Tellus 16, 436–468. https://doi.org/10.1111/j.2153-		
	3490.1964.tb00181.x		
	Dee, D.P., Uppala, S.M., Simmons, A.J., Berrisford, P., Poli, P., Kobayashi, S., Andrae, U., Balmaseda, M.A.,		
	Balsamo, G., Bauer, P., Bechtold, P., Beljaars, A.C.M., Berg, L. van de, Bidlot, J., Bormann, N., Delsol,		
	C., Dragani, R., Fuentes, M., Geer, A.J., Haimberger, L., Healy, S.B., Hersbach, H., Hólm, E.V., Isaksen,		
780	L., Kållberg, P., Köhler, M., Matricardi, M., McNally, A.P., Monge-Sanz, B.M., Morcrette, JJ., Park,		
	BK., Peubey, C., Rosnay, P. de, Tavolato, C., Thépaut, JN., Vitart, F., 2011. The ERA-Interim		
	reanalysis: configuration and performance of the data assimilation system. Quarterly Journal of the Royal		
i	Meteorological Society 137, 553–597. https://doi.org/10.1002/qj.828		(
	Díaz, M.F., Rodríguez, J., Pérez, E., Castaño, S., Araguás, L., 2007. Factors controlling the stable isotopic		Con formato: Español (alfab. internacional)
785	composition of recent precipitation in Spain, in: Advances in Isotope Hydrology and Its Role in		Internacionary
	Sustainable Water Resources Management (IHS-2007). Proceedings Series. Proceedings of an		
	International Symposium on Advances in Isotope Hydrology and Its Role in Sustainable Water Resources Management (IHS-2007). pp. 239–249.		
1	Díaz-Tejeiro, M.F., Pérez-Zabaleta, E., Prado-Pérez, A., Bardasano-Picazo, L., Muñoz-Delavarga, D., Rodríguez,	_	Con formato: Español (alfab.
790	J., 2013. La Red Española de Vigilancia de Isótopos en la Precipitación (REVIP). Presented at the XI		internacional)
170	CONGRESO NACIONAL DE GEOQUÍMICA, Soria.		
	Domínguez-Villar, D., Wang, X., Krklec, K., Cheng, H., Edwards, R.L., 2017. The control of the tropical North		
1	Atlantic on Holocene millennial climate oscillations. Geology 45, 303–306.		
	https://doi.org/10.1130/G38573.1		
795	Draxler, R.R., Rolph, G.D., 2010. HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model		
	access via NOAA ARL READY Website. http://ready.arl.noaa.gov/HYSPLIT.php.		
	Epstein, S., Mayeda, T., 1953. Variation of O18 content of waters from natural sources. Geochimica et		Con formato: Español (alfab.
	Cosmochimica Acta 4, 213–224. https://doi.org/10.1016/0016-7037(53)90051-9		internacional)
	Field, R.D., 2010. Observed and modeled controls on precipitation δ^{18} O over Europe: From local temperature to		
800	the Northern Annular Mode. Journal of Geophysical Research 115.		
	https://doi.org/10.1029/2009JD013370		
	Fischer, B.M.C., Aemisegger, F., Graf, P., Sodemann, H., Seibert, J., 2019. Assessing the Sampling Quality of a		
	Low-Tech Low-Budget Volume-Based Rainfall Sampler for Stable Isotope Analysis. Front. Earth Sci. 7.		
805	https://doi.org/10.3389/feart.2019.00244		
805	Fischer, M., Baldini, L., 2011. A climate-isotope regression model with seasonally-varying and time-integrated relationships. Climate Dynamics 37, 2235–2251. https://doi.org/10.1007/s00382-011-1009-1		
	relationships. Chinate Dynamics 57, 2255–2251. https://doi.org/10.1007/500562-011-1009-1		
	24		

	Friedman, I., 2002. Stable isotope composition of waters in the Great Basin, United States 1. Air-mass		
	trajectories. Journal of Geophysical Research 107. https://doi.org/10.1029/2001JD000565		
010	G. A. Schmidt, G. R. Bigg, E. J. Rohling, 1999. Global Seawater Oxygen-18 Database - v1.22. Global Seawater		
810	Oxygen-18 Database - v1.22. URL https://data.giss.nasa.gov/o18data/		
	Genty, D., Labuhn, I., Hoffmann, G., Danis, P.A., Mestre, O., Bourges, F., Wainer, K., Massault, M., Van Exter,		
1	S., Régnier, E., Orengo, Ph., Falourd, S., Minster, B., 2014. Rainfall and cave water isotopic relationships	(
	in two South-France sites. Geochimica et Cosmochimica Acta 131, 323–343.		Con formato: Español (alfab. internacional)
015	https://doi.org/10.1016/j.gca.2014.01.043	(
815	Gimeno, L., Nieto, R., Trigo, R.M., Vicente-Serrano, S.M., Lopez-Moreno, J.I., 2010. Where does the Iberian		
	Peninsula moisture come from? An answer based on a Lagrangian approach. Journal of		
	Hydrometeorology.		
1	Hammer, O., Harper, D. A. T., Ryan, P. D., 2001. PAST: Paleontological statistics software package for	(
000	education and data analysis. 4(1): 9pp. Palaeontologia Electronica 4 (1), 9.		Con formato: Español (alfab. internacional)
820	Iglesias González, M.I., 2019. Variabilidad climática del noroeste de la península ibérica durante los últimos	(
	1500 años, descrita por espeleotemas de diversas cuevas del principado de Asturias		
	(http://purl.org/dc/dcmitype/Text). Universidad de Oviedo.		
	Jeelani, G., Deshpande, R.D., Galkowski, M., Rozanski, K., 2018. Isotopic composition of daily precipitation		
025	along the southern foothills of the Himalayas: impact of marine and continental sources of atmospheric		
825	moisture. Atmospheric Chemistry and Physics 18, 8789–8805. https://doi.org/10.5194/acp-18-8789-2018 Kawale, J., Chatterjee, S., Kumar, A., Liess, S., Steinbach, M., Kumar, V., 2011. Anomaly construction in		
	climate data: issues and challenges. Proceedings of the Conference on Intelligent Data Understanding		
	Mountain View, California, USA, 189–203.		
1	Krklec, K., Domínguez-Villar, D., 2014. Quantification of the impact of moisture source regions on the oxygen	ſ	Con formator Fanañal (alfah
830	isotope composition of precipitation over Eagle Cave, central Spain. Geochimica et Cosmochimica Acta.		Con formato: Español (alfab. internacional)
850	https://doi.org/10.1016/j.gca.2014.03.011	-	Con formato: Español (alfab.
	Lachniet, M.S., 2009. Climatic and environmental controls on speleothem oxygen-isotope values. Quaternary		internacional)
	Science Reviews 28, 412–432.	, v	
	Lee, KO., Aemisegger, F., Pfahl, S., Flamant, C., Lacour, JL., Chaboureau, JP., 2019. Contrasting stable		
835	water isotope signals from convective and large-scale precipitation phases of a heavy precipitation event		
000	in southern Italy during HyMeX IOP 13: a modelling perspective. Atmospheric Chemistry and Physics		
	19, 7487–7506. https://doi.org/10.5194/acp-19-7487-2019		
	LeGrande, A.N., Schmidt, G.A., 2006. Global gridded data set of the oxygen isotopic composition in seawater.		
	Geophys. Res. Lett. 33, 5 PP. https://doi.org/200610.1029/2006GL026011		
840	Leng, M.J., 2006. Isotopes in palaeoenvironmental research. Springer.		
	Llasat, MC., Martín, F., Barrera, A., 2007. From the concept of "Kaltlufttropfen" (cold air pool) to the cut-off		Con formato: Español (alfab.
	low. The case of September 1971 in Spain as an example of their role in heavy rainfalls. Meteorology		internacional)
	and Atmospheric Physics 96, 43-60. https://doi.org/10.1007/s00703-006-0220-9		
	López-Blanco, C., Andrews, J., Dennis, P., Miracle, M.R., Vicente, E., 2016. Sedimentary response of lake El		
845	Tobar, Spain, to climate: lake level changes after the Maunder Minimum. J. Quaternary Sci. 31, 905–918.		
	https://doi.org/10.1002/jqs.2915		
	Martín-Vide, J., Olcina Cantos, J., 2001. Climas y tiempos de España. Alianza Editorial, Madrid.		Con formato: Español (alfab.
	Martin-Vide, J., Olcina-Cantos, J., 2001. Tiempos y climas de España.	l	internacional)
	Millán, M.M., Estrela, M.J., Miró, J., 2005. Rainfall Components: Variability and Spatial Distribution in a		
850	Mediterranean Area (Valencia Region). Journal of Climate 18, 2682–2705.		
	https://doi.org/10.1175/JCLI3426.1		

	Moreno, A., Pérez-Mejías, C., Bartolomé, M., Sancho, C., Cacho, I., Stoll, H., Delgado-Huertas, A., Hellstrom, J., Edwards, R.L., Cheng, H., 2017. New speleothem data from Molinos and Ejulve caves reveal		
1	Holocene hydrological variability in northeast Iberia. Quaternary Research 1–11.	- (Con formato: Español (alfab.
855	https://doi.org/10.1017/qua.2017.39		nternacional)
	Moreno, A., Sancho, C., Bartolomé, M., Oliva-Urcia, B., Delgado-Huertas, A., Estrela, M.J., Corell, D., López-		
	Moreno, J.I., Cacho, I., 2014. Climate controls on rainfall isotopes and their effects on cave drip water		
	and speleothem growth: the case of Molinos cave (Teruel, NE Spain). Clim Dyn 43, 221–241.		Con formato: Español (alfab.
	https://doi.org/10.1007/s00382-014-2140-6		nternacional)
860	Pérez-Mejías, C., Moreno, A., Sancho, C., Bartolomé, M., Stoll, H., Osácar, M.C., Cacho, I., Delgado-Huertas,	_	
	A., 2018. Transference of isotopic signal from rainfall to dripwaters and farmed calcite in Mediterranean		
	semi-arid karst. Geochimica et Cosmochimica Acta 243, 66–98.		Con formato: Español (alfab.
	https://doi.org/10.1016/j.gca.2018.09.014		nternacional)
	Pérez-Mejías, C., Moreno, A., Sancho, C., Martín-García, R., Spötl, C., Cacho, I., Cheng, H., Edwards, R.L.,	_	
865	2019. Orbital-to-millennial scale climate variability during Marine Isotope Stages 5 to 3 in northeast		
000	Iberia. Quaternary Science Reviews 224, 105946. https://doi.org/10.1016/j.quascirev.2019.105946		
	Risi, C., Bony, S., Vimeux, F., 2008. Influence of convective processes on the isotopic composition (δ18O and		
	δD) of precipitation and water vapor in the tropics: 2. Physical interpretation of the amount effect.		
	Journal of Geophysical Research: Atmospheres 113. https://doi.org/10.1029/2008JD009943		
870		_	Con formato: Español (alfab.
070	Stationary Convective Systems over Eastern Spain. Mon. Wea. Rev. 128, 3731–3751.		nternacional)
	https://doi.org/10.1175/1520-0493(2001)129<3731:MNSOTC>2.0.CO;2	_	
	Romero, R., Ramis, C., Alonso, S., 1997. Numerical simulation of an extreme rainfall event in Catalonia: Role of		
	orography and evaporation from the sea. Quarterly Journal of the Royal Meteorological Society 123,		
875	537–559. https://doi.org/10.1002/qj.49712353902		
015	Rozanski, K., Araguás-Araguás, L., Gonfiantini, R., 1993. Isotopic patterns in modern global precipitation.		Con formato: Español (alfab.
I	Geophysical Monograph Series 78, PP. 1-36.		nternacional)
	Rüdisühli, S., Sprenger, M., Leutwyler, D., Schär, C., Wernli, H., 2020. Attribution of precipitation to cyclones	_	
	and fronts over Europe in a kilometer-scale regional climate simulation. Weather and Climate Dynamics		
880	1, 675–699. https://doi.org/10.5194/wcd-1-675-2020		
000	Sancho, C., Arenas, C., Vázquez-Urbez, M., Pardo, G., Lozano, M.V., Peña-Monné, J.L., Hellstrom, J., Ortiz,		
	J.E., Osácar, M.C., Auqué, L., Torres, T., 2015. Climatic implications of the Quaternary fluvial tufa		
ĺ	record in the NE Iberian Peninsula over the last 500 ka. Quaternary Research 84, 398–414.		Con formato: Español (alfab.
	https://doi.org/10.1016/j.yqres.2015.08.003		nternacional)
885	Sancho, C., Belmonte, Á., Bartolomé, M., Moreno, A., Leunda, M., López-Martínez, J., 2018. Middle-to-late	_	
	Holocene palaeoenvironmental reconstruction from the A294 ice-cave record (Central Pyrenees, northern		
	Spain). Earth and Planetary Science Letters 484, 135–144. https://doi.org/10.1016/j.epsl.2017.12.027		
	Siegenthaler, U., Oeschger, H., 1980. Correlation of 180 in precipitation with temperature and altitude. Nature		
	285, 314–317. https://doi.org/10.1038/285314a0		
890	Smith, A., British Geological Survey, Wynn, P., Lancaster University, Barker, P., Lancaster University, Leng,		
	M., British Geological Survey, Noble, S., British Geological Survey, Stott, A., Centre for Ecology and		
	Hydrology, 2016. Cave monitoring and the potential for palaeoclimate reconstruction from Cueva de		
	Asiul, Cantabria (N. Spain). International Journal of Speleology 45, 1–9. https://doi.org/10.5038/1827-		
	806X.45.1.1928		
895	Stoll, H., Mendez-Vicente, A., Gonzalez-Lemos, S., Moreno, A., Cacho, I., Cheng, H., Edwards, R.L., 2015.		
	Interpretation of orbital scale variability in mid-latitude speleothem $\delta 180$: Significance of growth rate		
	controlled kinetic fractionation effects. Quaternary Science Reviews, Novel approaches to and new		

insights from speleothem-based climate reconstructions 127, 215–228. https://doi.org/10.1016/j.quascirev.2015.08.025

- 900 Suess, E., Aemisegger, F., Sonke, J.E., Sprenger, M., Wernli, H., 2019. Marine versus continental sources of iodine and selenium in rainfall at two European high-altitude locations. Environmental Science & Technology 53, 4, 1905–1917.
 - Treble, P.C., Budd, W.F., Hope, P.K., Rustomji, P.K., 2005a. Synoptic-scale climate patterns associated with rainfall δ18O in southern Australia. Journal of Hydrology 302, 270–282.
- 905 https://doi.org/10.1016/j.jhydrol.2004.07.003
 - Treble, P.C., Chappell, J., Gagan, M.K., McKeegan, K.D., Harrison, T.M., 2005b. In situ measurement of seasonal δ18O variations and analysis of isotopic trends in a modern speleothem from southwest Australia. Earth and Planetary Science Letters 233, 17–32. https://doi.org/10.1016/j.epsl.2005.02.013
- Tudurí, E., Ramis, C., 1997. The Environments of Significant Convective Events in the Western Mediterranean. 910 Wea. Forecasting 12, 294–306. https://doi.org/10.1175/1520-0434(1997)012<0294:TEOSCE>2.0.CO;2
- Tyler, J.J., Jones, M., Arrowsmith, C., Allott, T., Leng, M.J., 2016. Spatial patterns in the oxygen isotope composition of daily rainfall in the British Isles. Clim Dyn 47, 1971–1987. https://doi.org/10.1007/s00382-015-2945-y



Figure 1. (A) Location of the studied stations in northern Spain where rainfall was collected. Map source: Average annual precipitation (mm) for 1980-2005 provided by the Spanish Ministry of Agriculture and Fisheries, Food and Environment (MAPAMA); (B) weather maps showing the three precipitation regimes of the IP defined by Millán et al. (2005): (i) Atlantic frontal systems, (ii) convective-orographic storms, and (iii) Backdoor cold fronts from the Mediterranean Sea. In the maps, the sea level pressure and the 500 hPa geopotential height (gpdam in German) are indicated by the different colors; the scale represents the height- from 4600 to 6000 m - where the pressure of 500hPa is reached. White lines are the isobars (bodendruck in German). Source: CFS Reanalysis (CFSR) and Wetterzentrale.



Figure 2. Scatter plots of $\delta^2 H_p$ versus $\delta^{18}O_p$ in precipitation and Local Meteoric Water Lines (LMWL), including equations, for El Pindal, Ortigosa de Cameros, Borrastre, Molinos and Mallorca with Barcelona stations. Note that El Pindal plot includes only 36 samples since $\delta^2 H$ was not measured in the remaining <u>onessamples</u>. The difference in the other graphs in sample number (n) respect to those indicated in Table S1 is due to the removal of some samples that have been subject to evaporation effects (see text for more information). Global Meteoric Water Line (GMWL) and Western Mediterranean Meteoric Water Line (WMMWL) are plotted in black and gray, respectively, in every graph.



935 Figure 3. Event $\delta^{18}O_p$ daily time series for the studied stations presented versus time (2010-2017). Note that El Pindal samples (2006-2009) are not represented since they do not overlap with the time period of the other stations (2006-2009). See text for more explanation.



Figure 4. (A) Climographs showing monthly mean temperature (line with dots) and monthly mean rainfall (bars) obtained for the longest AEMET meteorological stations available next to the study sites (Oviedo, Logroño, Teruel, Huesca, Barcelona and Palma de Mallorca). Note that these stations are not at the same elevation or microclimate as the ones where rainfall was collected. For this reason, the climographs are indicated here to account for broad regional climates while the correlations (Table3) with meteorological data were performed using more proximal (although shorter in the recorded time interval) stations. (B) Variability of monthly weighted $\delta^{18}O_p$ at the studied sites. Dots represent monthly precipitation-amount weighted averages and lines are the mean of these monthly precipitation-amount weighted averages (see also Table 2 and Table S2).





Figure 5. Wind roses showing the averaged back trajectories directions <u>over 24 h life time</u> whose air masses produced precipitation in three stations in northern Iberia: Oviedo (northern Spain), Borrastre (central Pyrenees) and Mallorca (Balearic Islands). (A) Amount of precipitation (measured at the nearest meteorological station) during the intervals of sample collection and (B) $\delta^{18}O_p$ indicated by colors (see legends). Source regions of each air mass, generated by averaging the direction of each point of the back trajectory (20 points), are <u>broken-divided</u> into 10° sectors. The percentages of back trajectories, whose averaged directions are associated with each 10° sector, are shown as dashed circles (from 0 to 12%).



960 trajectory over Iberia in every case. The three lines represent the air masses at different elevation (red: 850 hPa, blue: 700 hPa and green: 500hPa) (see text for more explanation).

Con formato: Centrado



Figure 6. Maps of moisture uptake locations along rainfall backward trajectories represented for Oviedo, Borrastre and Mallorca stations. See legend for colors (indicating seasons) and dot sizes (indicating amount of moisture).





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Figure 7. Box plots of δ¹⁸O_{pr} air temperature and precipitation amount_for the three identified rainfall types in northern Iberia: Atlantic fronts (in dark blue), backdoor cold fronts (light blue) and convective precipitation (in green) for the studied stations. The central rectangle spans the first quartile to the third quartile (the *likely range of variation*, the *IQR*). A segment inside the rectangle shows the median and "whiskers" above and below the box show the locations of the minimum and maximum. <u>Values of δ¹⁸O_p</u>

appear normalized to better compare among the seven stations. The Kruskal-Wallis test indicates that at least two of the three rainfall types are significantly different in terms of their $\delta^{18}O_p$ values.

975 Table captions

Table 1. Data collection details for the seven studied stations. KGC: Köppen and Geiger climate classification; AEMET: Agencia Española de Meteorología; SAIH: Automatic Hydrologic Information System. See Table S1 for all the isotopic and meteorological data. The AEMET stations with long series represented in Figure 4A are indicated.

			Ra	ainfall colle	ction site	•				Meteo	orological da	ıta	
1	Location	Coordi Lat	nates and d	altitude Altitude m asl	KGC	I Collection period	Data descrip Nº samples	tion Laboratory	Statio Name	n Type	Annual mean Temp (°C)	mean rainfall Temp (mm)	
151	Oviedo	43°21N	5°51W	245	Cfb	Feb 2015- Jan 2016	47	Universities. of Oviedo and	Oviedo (120 km from El Pindal)	AEMET	13.3	960	Oviedo
Cantabrian coast	El Pindal	43°23N	4°31W	24	Cfb	Nov 2006- Feb 2007 July 2007- May 2008 Jan 2009- April2009	101	Barcelona (UB)	Data from reanalysis (ECWMF ERA interim data)				
Range	Ortigosa de Cameros	42°10N	2°42W	1060	Dsb	Sep 2010– Dec 2014	189	IACT-CSIC and UB	Villoslada de Cameros (6.5 km)	La Rioja govern	9.6	650	Logroño
Ibe rian Range	Molinos	40°47N	0°26W	1040	Dsb	March 2010-May 2015	254	IACT-CSIC	Gallipuén (7 km)	SAIH Ebro	12	500	Teruel
Pyrenees	Borrastre	42°29N	0°06W	770	Csb	Since April 2011	374		Borrastre (in situ)	Meteo- climatic	13.5	900	Huesca
ean	Barcelona	41°21N	2°06E	20	Csa	Since Oct 2015	53	UB	Barcelona- Zona Universitaria (in situ)	meteocat	17.2	430	Barcelona
Medite weanean	Mallorca (Manacor and Porto Cristo)	39°33N	3°12E	90	Csa	Since May 2013	98		Sant Llorenç (8 km)	AEMET	18.8	590	Palma de Mallorca

Table 2. Mean values of $\delta^{18}O_p$ data for the seven sites every station in the study transect at a monthly and annual scale. Only months and years with all events collected are averaged. Note that the number of months or years averaged (a, in the tablenumber between brackets after the $\delta^{18}O_p$ values) are not the same for all the stations, neither the time period considered (check Table 1 for the sampling period in every station). For the complete monthly dataset with all the monthly values indicated, please refer to see Table S2.

δ ¹⁸ O _p (‰)	Cantabr	ian coast	Iberian	Range	Pyrenees	Mediterranean		
	Oviedo	El Pindal	Ortigosa de Cameros	Molinos	Borrastre	Barcelona	Mallorca	
January		-7.46 (3)	-12.29 (2)	-10.12 (4)	-8.64 (4)	-5.15 (1)	-8.91 (4)	
February		-5.49 (2)	-7.28 (3)	-9.71 (5)	-11.25 (4)	-6.71 (2)	-4.57 (4)	
March	-6.29 (1)	-5.19 (2)	-7.74 (3)	-10.25 (6)	-9.49 (4)	-8.00 (2)	-7.15 (3)	
April	-5.12 (1)	-4.27 (2)	-6.25 (4)	-7.68 (6)	-6.35 (4)	-7.38 (2)	-4.86 (3)	
May		-5.25 (1)	-3.66 (3)	-6.13 (5)	-5.19 (6)	-2.21 (2)	-3.36 (3)	
June	-3.73 (1)		-5.21 (4)	-5.12 (4)	-4.32 (5)	-8.01 (1)	-3.06 (2)	
July	-7.50 (1)	-2.04 (1)	-6.39 (4)	-2.22 (4)	-4.44 (5)	-1.64 (1)	-1.13 (1)	
August	-4.80 (1)	-3.94 (1)	-3.64 (4)	-5.00 (4)	-4.65 (5)		-3.15 (3)	
September	-5.83 (1)	-3.17 (1)	-7.09 (3)	-5.93 (3)	-5.83 (5)	-6.13 (1)	-4.14 (5)	
October	-5.47 (1)	-4.12 (1)	-6.17 (4)	-7.18 (3)	-6.46 (5)	-5.53 (1)	-6.38 (4)	
November	-5.87 (1)	-4.31 (1)	-9.40 (4)	-11.26 (4)	-7.24 (5)	-6.95 (2)	-6.34 (4)	
December	-7.16 (1)	-5.23 (2)	-7.91 (3)	-10.41 (3)	-8.00 (4)	-7.27 (1)	-5.28 (4)	
Annual			-7.09 (1)	-7.18 (2)	-6.37 (3)			

δ ¹⁸ O _p (‰)	Cantabrian coast					Iberian Range				Pyrenees		Mediterranean			
	Oviedo		El Pindal		Ortigosa de Cameros		Molinos		Borrastre		Barcelona		Mallorca		
January	n=0		n=3	-7.46	n=2	-12.29	n=4	-10.12	n=4	-8.64	n=1	-5.15	n=4	-8.9	
February	n=0		n=2	-5.49	n=3	-7.28	n=5	-9.71	n=4	-11.25	n=2	-6.71	n=4	-4.5	
March	n=1	-6.29	n=2	-5.19	n=3	-7.74	n=6	-10.25	n=4	-9.49	n=2	-8.00	n=3	-7.1	
April	n=1	-5.12	n=2	-4.27	n=4	-6.25	n=6	-7.68	n=4	-6.35	n=2	-7.38	n=3	-4.8	
May	n=0		n=1	-5.25	n=3	-3.66	n=5	-6.13	n=6	-5.19	n=2	-2.21	n=3	-3.3	
June	n=1	-3.73	n=0		n=4	-5.21	n=4	-5.12	n=5	-4.32	n=1	-8.01	n=2	-3.0	
July	n=1	-7.50	n=1	-2.04	n=4	-6.39	n=4	-2.22	n=5	-4.44	n=1	-1.64	n=1	-1.1	
August	n=1	-4.80	n=1	-3.94	n=4	-3.64	n=4	-5.00	n=5	-4.65	n=0		n=3	-3.1	
September	n=1	-5.83	n=1	-3.17	n=3	-7.09	n=3	-5.93	n=5	-5.83	n=1	-6.13	n=5	-4.1	
October	n=1	-5.47	n=1	-4.12	n=4	-6.17	n=3	-7.18	n=5	-6.46	n=1	-5.53	n=4	-6.3	
November	n=1	-5.87	n=1	-4.31	n=4	-9.40	n=4	-11.26	n=5	-7.24	n=2	-6.95	n=4	-6.3	
December	n=1	-7.16	n=2	-5.23	n=3	-7.91	n=3	-10.41	n=4	-8.00	n=1	-7.27	n=4	-5.2	
Annual	n=0		n=0		n=1	-7.09	n=2	-7.18	n=3	-6.37	n=0		n=0		

Table 3. Spearman's rank correlation coefficients between $\delta^{18}O_p$ and air temperature and precipitation amount for every sampling station at daily scale (A) and monthly scale (B) using deseasonalised deseasonalized data (removing the seasonal component by 995 subtracting their monthly averages). Significant correlations with p value < 0.05 after application of Bonferroni test are in reditalics. Note that the relatively small size of Oviedo and Barcelona rain events likely precludes statistically significant correlations. (C) Multiple regression coefficient (r) and *p-value* for every site is included, indicating the coefficient and the standard error for the constant, the precipitation and the temperature variables. As an example, the equation for Molinos should be read as follows: $\delta^{18}O_p = -0.05(\pm 0.019) A + 0.40(\pm 0.05) T + 0.43$, with A as the amount of precipitation, T as air temperature and

		Oviedo	El Pindal	Ortigosa de Cameros	Molinos	Borrastre	Barcelona	Mallorca
(A) Daily correlations		n = 39	n = 109	n=189	n=248	n=352	n=53	n=98
$\delta^{18}O_p$ - temperature	rs	0.23	0.34	0.25	0.41	0.31	0.24	0.35
	p value	0.328	0.0012	0.001	2.00E-11	1.17E-09	0.21	0.0013
$\delta^{18}O_p$ - precipitation amount	rs	-0.22	-0.06	-0.32	-0.19	-0.11	-0.35	-0.28
	p value	0.368	1	1.05E-05	0.005	0.119	0.029	0.013
(B) Monthly correlations	(B) Monthly correlations		n = 17	n=41	n=51	n=49	n=16	n=40
$\delta^{18}O_p$ - temperature	rs	0.3	0.33	0.46	0.76	0.61	0.39	0.41
	p value	1	1	0.013	3.36E-10	1.44E-05	0.804	0.05
$\delta^{18}O_p$ - precipitation amount	rs	0.066	-0.44	-0.34	-0.4	-0.11	-0.30	-0.12
	p value	0.843	0.4	0.176	0.018	1	1	0.436
(C) Multiple regression	r	0.30	0.40	0.40	0.43	0.30	0.32	0.41
(with daily data)	p value	0.118	0.0001	3.36E-08	4.68E-13	8.13E-09	0.004	0.008
Constant	Coeff	0.14	0.32	-1.6	0.43	-2.83E-11	-0.49	0.23
	Std err	0.43	0.24	0.22	0.18	0.16	0.38	0.26
Precipitation	Coeff	-0.015	-0.013	-0.11	-0.05	-0.018	-0.05	-0.02
	Std err	0.05	0.04	0.02	0.019	0.014	0.02	0.017
Temperature	Coeff	0.21	0.25	0.25	0.40	0.40	0.37	0.31
	Std err	0.11	0.05	0.06	0.05	0.06	0.19	0.11

1000 0.43 as a constant value. 1005 Table 4. Relative frequency (in %) of the three rainfall types in every studied station.

	Cantabrian coast		Iberian Range		Pyrenees	Mediterranean	
	Oviedo	El Pindal	Ortigosa de Cameros	Molinos	Borrastre	Barcelona	Mallorca
Atlantic fronts	68.09	71.29	58.7	51.8	65.2	58.49	40.82
Backdoor cold fronts	14.89	16.83	20.6	23.9	11.8	24.53	38.78
Convective	17.02	11.88	20.6	24.3	23.0	16.98	20.41

Tabla 5. Kruskal-Wallis test performed on $\delta^{18}O_p$ data to discriminate if the three synoptic patterns are statistically different in 1010 terms of their isotopic composition. High values of the test (Kruskal-Wallis H) and low *p*-values indicate that at least two of the three synoptic patterns are statistically different in terms of $\delta^{18}O_p$ data.

Cantabrian coast Iberian Range Pyrenees Mediterranean Oviedo El Ortigosa Molinos Borrastre Barcelona Mallorca Pindal de Cameros 3.017 23.3 22.23 Kruskal-10.86 48.38 47.84 4.109 Wallis H 0.221 0.004 8.7E-06 3.12E-11 4.09E-11 0.1282 1.49E-05 p value

Con formato: Inglés (Estados Unidos)

Supplementary

Figure S1. Wind roses represent the averaged back trajectories of air masses that produced precipitation at three stations in northern Iberia: Oviedo (northern Spain), Borrastre (central Pyrenees) and Mallorca (Balearic Islands). Trajectories shown were 1020 computed for only 24120 hours.

As supplementary

I	Figure S2. Oviedo station. From top to bottom: daily precipitation (mm), $\delta^{18}O$ (‰) and daily temperature average (°C).
1025	Figure S3. El Pindal station. From top to bottom: daily precipitation (mm), <i>d-excess</i> , δ ¹⁸ O (‰) and daily temperature average (°C).
	Figure S4. Ortigosa station. From top to bottom: daily precipitation (mm), <i>d-excess</i> , δ^{18} O (‰) and daily temperature average (°C).
1030	Figure S5. Molinos station. From top to bottom: daily precipitation (mm), <i>d-excess</i> , δ^{18} O (‰) and daily temperature average (°C).
	Figure S6. Borrastre station. From top to bottom: daily precipitation (mm), d-excess, δ^{18} O (‰) and daily temperature average
	<u>(°C).</u>
	Figure S7. Barcelona station. From top to bottom: daily precipitation (mm), <i>d-excess</i> , δ^{18} O (‰) and daily temperature average
1035	<u>(°C).</u>
	Figure S8. Mallorca station. From top to bottom: daily precipitation (mm), <i>d-excess</i> , δ^{18} O (‰) and daily temperature average (°C).
	Table S1. Event $\delta^{18}O_p$ and δ^2H_p data for the stations considered in this study. Meteorological data from nearby stations (Table 1)

1040 are also included.

As supplementary

Table S2. Monthly $\delta^{18}O_p$ data for the stations considered in this study

As supplementary

1045

Con formato: Fuente: Symbol
Con formato: Superíndice