#### Isotopic composition of convective rainfall in the inland tropics of 1 **Brazil** 2

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20 Abstract. Strong convective systems characterize the tropical central-southern region of Brazil. These systems provide 21 abundant water supply for agro-industrial activities and pose flood risks to large cities. Here, we present high-frequency (2-10 22 min; inter and intra-event) rainfall isotopic compositions (n=90 samples) to reveal regional and local atmospheric processes 23 controlling the isotopic variability of convective systems between 2019-2021. Inter-events rainfall weighted-average ( $\delta_{wed}$ ) values were low ( $\delta^{18}O_{wed} \leq -10.0$  ‰) due to the influence of evapotranspiration from the Amazon forest during the summer 24 25 and associated with higher rainfall along Hysplit trajectories. In contrast, during autumn and spring seasons Hysplit trajectories from the Atlantic Ocean and South Brazil exhibited lower rainfall amounts, resulting in high  $\delta^{18}O_{wed} \ge -4.2$  ‰. This strong 26 27 regional  $\delta_{wed}$  pattern often masks intra-event isotopic variability. Therefore, we analyzed the vertical structure of local rainfall 28 using reflectivity (Z) from micro radar data. Variations in Z indicate that microphysical processes within falling raindrops led 29 to changes in  $\delta^{18}$ O and *d*-excess. Our findings establish a novel framework for evaluating the meteorological controls on the 30 isotopic variability of convective precipitation in tropical South America, fill the gap of high-frequency studies in this region,

31 and generate an isotopic dataset for convective model evaluations.

## 32 1 Introduction

33 The tropical central-southern region of Brazil (CSB) is the primary contributor to the country's economy, with 34 agriculture and agroindustry as the main sectors (Zilli et al., 2017). These economic activities are highly dependent on seasonal 35 rainfall for irrigation and hydropower supply (Luiz Silva et al., 2019). Projected changes in the frequency of heavy and extreme 36 rainfall events (Marengo et al., 2020; Donat et al., 2013; IPCC, 2021; Marengo et al., 2021) pose a significant threat to regional 37 economic growth and energy generation. Similarly, according to Marengo et al. (2021), simulations with pre-CMIP6 models 38 suggest that the intensification of heavy rainfall events could exacerbate the prevalence of floods and landslides in susceptible 39 regions. Such occurrences have resulted in a total cost of US\$41.7 billion over the past half-century (Marengo et al., 2020; 40 World Meteorological Organization, 2021).

41 Extreme precipitation events are linked to convective systems (CS). These systems significantly contribute to the annual 42 rainfall budget and account for a large number of extreme rainfall events (Roca and Fiolleau, 2020). Across the tropics, diurnal 43 surface heating amplifies convection, generating short-lived events that can occur in consecutive days. Rapid upward 44 movement of air results in large condensation and precipitation rates (Breugem et al., 2020; Kastman et al., 2017; Lima et al., 45 2010; Machado et al., 1998). This is identified by vigorous vertical development in the form of *cumulus-nimbus* and *cumulus* 46 congestus (convective clouds) and low-level divergence (stratiform clouds) (Sigueira et al., 2005; Machado and Rossow, 1993; 47 Zilli et al., 2017; Houze, 1989, 2004). Precipitation associated with these systems are commonly referred as convective and 48 stratiform rainfall, and account for 45% and 46% of the total rainfall in South America, respectively (Romatschke and Houze, 2013). 49

50 Whether rainfall is convective or stratiform has been suggested to determine variations in stable isotope composition of 51 precipitation across the tropics (Zwart et al., 2018; Sánchez-Murillo et al., 2019; Sun et al., 2019; Han et al., 2021; Aggarwal 52 et al., 2016; Munksgaard et al., 2019). Processes driving the variations in the isotopic composition in CS are more complex 53 and less understood compared to other precipitation producing systems. Studies using the isotopic composition of rain and 54 water vapor have quantified and modelled physical processes related to convection (Bony et al., 2008; Kurita, 2013). Previous 55 studies have suggested that the isotopic composition of CS is connected to the integrated history of convective activity (Risi 56 et al., 2008; Moerman et al., 2013), depth of organized convection and aggregation (Lawrence et al., 2004; Lekshmy et al., 57 2014; Lacour et al., 2018; Galewsky et al., 2023), microphysical processes within clouds (Aggarwal et al., 2016; Lawrence et 58 al., 2004; Zwart et al., 2018), and cold pool dynamics (Torri, 2021). These interpretations simplified and lumped the effects of 59 multiple rainfall timescales (e.g. monthly, daily and sub-hourly), providing different perspectives on convective processes, 60 such as the regional (synoptic forcings) and local factors (e.g. microphysical processes occurring both within and below the cloud) (Kurita et al., 2009; Muller et al., 2015; Graf et al., 2019; Lee and Fung, 2008)... 61

High-frequency rainfall sampling and analyses of stable isotope ratios has been used to better understand the evolution of
large weather systems such as tropical cyclones and typhoons (Sun et al., 2022; Sánchez-Murillo et al., 2019; Han et al., 2021),
squall lines (Taupin et al., 1997; Risi et al., 2010; Tremoy et al., 2014; de Vries et al., 2022), mid-latitude cyclones, and cold

fronts (Barras and Simmonds, 2009; Celle-Jeanton et al., 2004; Aemisegger et al., 2015; Thurnherr and Aemisegger, 2022;
Muller et al., 2015; Landais et al., 2023). High-resolution isotope information can provide a better insight into the isotopic
variability during the life cycle of rainfall events (Coplen et al., 2008; Muller et al., 2015; Celle-Jeanton et al., 2004).

In this study, we used high-frequency rainfall sampling to disentangle regional (moisture origin/transport, regional atmospheric circulation) from local (below-cloud processes, vertical structure of rainfall, cloud top temperature) processes controlling the isotopic composition of convective rainfall. High-frequency rainfall was integrated with ground-based observational data (Micro Rain Radar and automatic weather station), satellite imagery (GOES-16), ERA-5 reanalysis products, and HYSPLIT trajectories to better characterize convective rainfall over the inland tropics of Brazil.

### 73 2 Data and Methods

## 74 2.1 Sampling site and weather systems

75 The rainfall sampling site was located in Rio Claro city, São Paulo State (Fig. 1a). The station (-22.39°S, -47.54°W, 670 m 76 a.s.l.) is part of the Global Network of Isotopes in Precipitation network (GNIP) and is influenced by weather systems 77 responsible for rainfall variations and seasonality linked to the regional atmospheric circulations across the CSB region. The 78 rainfall seasonality over CSB is associated with: (i) frontal systems (FS), represented mainly by cold fronts from southern 79 South America acting throughout the year, and (ii) the activity of the South Atlantic Convergence Zone (SACZ) during austral 80 summer (December to March) (Kodama, 1992; Garreaud, 2000) (Fig. 1b). These features are mostly responsible for CS 81 development (Romatschke and Houze, 2013; Siqueira et al., 2005; Machado and Rossow, 1993) (Fig. 1c), and were captured 82 during their passage over the Rio Claro station.

### 83 2.2 Rainfall sampling and isotope analyses

84 High-frequency rainfall sampling was conducted using a passive collector (2 to 10 minutes intervals) from September 2019 to 85 February 2021, except for April, July, and August (during winter 2020), when no rainfall was observed in the study area. The pandemic Covid-19 disrupted access to the university campus, thereby reducing the number of rainfall events sampled during 86 87 the spring of 2020, particularly at night (e.g., lockdowns). In this study, the rainfall samples collected do not consist of consecutive day-night pairs during the same day. In total, 90 samples representing eight convective events (3 night-time and 5 88 89 day-time events) were collected. Samples were transferred to the laboratory and stored in 20 mL HDPE bottles at 4°C. In 90 parallel to high-frequency sampling, monthly cumulative rainfall samples were also collected using the methodology 91 recommended by the International Atomic Energy Agency (IAEA, 2014).

Rainfall samples were analyzed for stable isotope composition using Off-Axis Integrated Cavity Output Spectroscopy (Los
 Gatos Research Inc.) at the Hydrogeology and Hydrochemistry laboratory of UNESP's Department of Applied Geology and

94 at the Chemistry School of the National University (UNA, Heredia, Costa Rica). All results are expressed in per mil relative 95 to Vienna Standard Mean Ocean Water (V-SMOW). The certified calibration standards used in UNESP were USGS-45 (δ<sup>2</sup>H = -10.3,  $\delta^{18}O = -2.24 \text{ }$ , USGS-46 ( $\delta^2H = -236.0 \text{ }$ ,  $\delta^{18}O = -29.80 \text{ }$ ), including one internal standard (Cachoeira de 96 97 Emas - CE -  $\delta^2$ H = -36.1 ‰,  $\delta^{18}$ O = -5.36 ‰). USGS standards were used to calibrate the results on the V-SMOW2-SLAP2 98 scale, whereas CE was used for memory and drift corrections. At UNA, the certified standards MTW ( $\delta^2 H = -130.3 \%$ ,  $\delta^{18}O$ = -16.7 ‰), USGS45 ( $\delta^2$ H = -10.3 ‰,  $\delta^{18}$ O = -2.2 ‰), and CAS ( $\delta^2$ H = -64.3 ‰,  $\delta^{18}$ O = -8.3 ‰) were used to correct the 99 100 measurement results for memory and drift effects and to calibrate them on the V-SMOW2-SLAP2 scale (García-Santos et al., 101 2022). The analytical uncertainty (1 $\sigma$ ) was 1.2 % for  $\delta^{2}$ H and 0.2 % for  $\delta^{18}$ O for UNESP analysis and 0.38 % for  $\delta^{2}$ H and 0.07 ‰ for  $\delta^{18}$ O for UNA analysis. Deuterium excess (*d*-excess) was calculated as: *d*-excess =  $\delta^2$ H -  $8*\delta^{18}$ O (Dansgaard, 102 103 1964), with uncertainties  $(1\sigma)$  of 1.33 and 0.43 %, respectively. This secondary isotope parameter was used to interpret the 104 influence of moisture origin/transport (Sánchez-Murillo et al., 2017; Froehlich et al., 2002) and local processes (Aemisegger 105 et al., 2015; Muller et al., 2015; Celle-Jeanton et al., 2004).

### 106 2.3 Meteorological data

107 An Automatic Weather Station (AWS) Decagon Em50 (METER) was installed next to the Micro Rain Radar (MRR) (METEK) 108 at 670 m.a.s.l, within immediate vicinity of the rainfall collection site. Meteorological data were recorded at 1 min intervals 109 for rain rate (RR, mm min<sup>-1</sup>), air temperature (T, °C) and relative humidity (RH, %). The MRR data for reflectivity (Z, in dBZ), and fall velocity (w, m s<sup>-1</sup>) were also recorded at 1 min intervals. MRR parameters correspond to the mean values measured at 110 111 the elevation between 150 and 350 meters above surface. MRR operated at a frequency of 24.230 GHz, modulation of 0.5 -112 15 MHz according to the height resolution mode. For this work, different height resolutions (31 range bin) were tested, 150 113 m, 200 m, 300 m and 350 m, resulting in vertical profiles of 4650 m, 6200 m, 930 0m and 10.850 m, respectively (Endries et 114 al., 2018). The MRR data used in the following discussion are the near-surface data (first measurement from 150 m to 350 m). 115 Lifting Condensation Level (LCL, meters) was computed from AWS RH and T, using expression proposed by Soderberg et 116 al. (2013). The rainfall amount (R, mm) was also calculated during the sampling interval. GOES-16 imagery was used to identify the convective nuclei of the cloud-top (10.35-µm, Band-13) and brightness temperature (BT, °C), at 10 min intervals 117 118 during the sampling period (Ribeiro et al., 2019; Schmit et al., 2017). The 10.35- $\mu$ m BT is often used to estimate the convective 119 cloud depth, since the lower BT is linked to deeper cloud tops (Adler and Fenn, 1979; Roberts and Rutledge, 2003; Adler and 120 Mack, 1986; Ribeiro et al., 2019; Machado et al., 1998). The weather systems (fronts, instabilities, and low pressure) were 121 defined according to the synoptic chart and meteorological technical bulletin of the Center for Weather Forecast and Climatic 122 Studies of the National Institute of Space Research (CPTEC/INPE) that used information of numerical models, automatic 123 weather stations, satellite and radar images, reanalysis data and regional atmospheric models, such as the Brazilian Global 124 Atmospheric Model and ETA model.

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# 126 2.4 Hysplit modeling and Reanalysis data

127 The origin of air masses and moisture transport to the Rio Claro site were evaluated using the HYSPLIT (Hybrid-Single 128 Particle Langragian Integrated Trajectory) modeling framework (Stein et al., 2015; Soderberg et al., 2013). The trajectories of 129 the air masses were estimated for 240 hours prior to rainfall onset, considering the estimated time of residence of the water 130 vapor (Gimeno et al., 2010, 2020; van der Ent and Tuinenburg, 2017). Start time of trajectories was the same as the start time 131 of rainfall events. The trajectories were computed using NOAA's meteorological data (global data assimilation system, GDAS: 132 1 degree, global, 2006-present), with ending elevations of the trajectories at 1500 m above the surface, taking into account the 133 climatological height of the Low Level Jet, within 1000-2000 m (Marengo et al., 2004). Ten-day trajectories representing 134 convective events were calculated as trajectory ensembles, each consisting of twenty-seven ensemble members released at 135 start hour of convective rainfall sample collection. Ensembles were produced by varying the initial trajectory wind speeds and pressures, according to the HYSPLIT ensemble algorithm, in order to account for the uncertainties involved in the simulation 136 137 of individual backward trajectories (Jeelani et al., 2018). A sum of the rainfall intensity (mm  $hr^{-1}$ ) along the trajectories was 138 used to analyse rainout of the moist air masses according to the Jeelani et al. (2018).

Reanalysis data were used to better understand the influence of atmospheric circulation on isotopic composition of rainfall at the study area. ERA-5 information was used to evaluate hourly vertical integrals of eastward water vapor flux (kg m<sup>-1</sup> s<sup>-1</sup>) during convective events sampled. The Global Modeling and Assimilation Office (GMAO) data (MERRA-2, 1 hour, 0.5 x 0.625 degree, V5.12.4 were used for calculations of latent heat flux (LHF). Aqua/AIRS L3 Daily Standard Physical Retrieval (AIRS-only) data (1-degree x 1-degree V7.0, Greenbelt, MD, USA, Goddard Earth Sciences Data and Information Services Center) (known as GES DISC) were used for average outgoing longwave radiation (OLR). OLR values below 240 W m<sup>-2</sup> indicate organized deep convection (Gadgil, 2003).

### 146 **2.5 Identification of convective rainfall events and vertical variations of reflectivity**

147 In general, the identification of CS was based on the vertical structure of the given precipitation system (lack of the 148 melting layer and bright band - BB) in the radar profiles featuring high reflectivity values (Z > 38 dBZ) (Houze, 1993, 1997; 149 Steiner and Smith, 1998; Rao et al., 2008; Mehta et al., 2020; Endries et al., 2018) and satellite imagery (Vila et al., 2008; 150 Ribeiro et al., 2019; Sigueira et al., 2005; Machado et al., 1998). Consequently, convective rainfall was defined in this study 151 by (i) convective cloud nuclei observed in GOES-16 imagery, (ii) no BB detected, (iii) Z > 38 dBZ near to the surface and (iv) rainfall intensity (AWS) of at least 10 mm h<sup>-1</sup> (Klaassen, 1988) (Fig. 1c,d). The convective nuclei were identified using GOES-152 153 16 imagery, determined as a contiguous area of at least 40 pixels with BT lower than 235K ( $\leq$  -38 °C) over Rio Claro station, 154 according to previous studies (Ribeiro et al., 2019).

The Z is defined as the mean number of raindrops within a specific diameter interval per unit volume of air. Therefore, Z represents the concentration of a particular raindrop property (in this case, the 6<sup>th</sup> power of their diameter, proportional to the

square of their volume) (Houze, 1993; Mehta et al., 2020; Uijlenhoet, 2001). A high Z value indicates a high concentration of 157 158 raindrops. A modification in the formation mechanism for precipitation particles results in a change in Z of the vertical profile 159 (Houze, 1997). Descriptive statistics were conducted on the Z values at different heights to comprehend and quantify the 160 dynamics of rain particle formation during intra-events. The resulting parameters from considering the entire vertical profile 161 of the MRR are: Z<sub>max</sub>: is the maximum reflectivity value in the vertical profile indicating the maximum concentration of 162 raindrops; Z<sub>median</sub>: refers to the median reflectivity in the vertical profile and was used to synthetize the change in vertical Z values; and Z<sub>amplitude</sub> (Z<sub>ampl</sub>) is defined as the difference between the maximum and minimum reflectivity values in the vertical 163 164 profile. In other words, a larger Z<sub>ampl</sub> indicates that raindrops undergo more microphysical transformations as they fall to the 165 surface.

### 166 2.6 Statistical tests

The Shapiro-Wilk test was applied to verify that the data distribution was normal (parametric) or non-normal (nonparametric) (Shapiro, S. S.; Wilk, 1965). A significant difference (p-value < 0.05) indicates a non-parametric distribution. A Spearman rank correlation test was used for nonparametric distribution data, whereas Pearson's linear correlation test was applied for parametric data. Correlation tests were conducted between isotopes ( $\delta^{18}O$  and *d*-excess) and meteorological data (AWS and MRR variables) during the same time interval and from individual events. Correlation tests were not applied to GOES-16 BT and reanalysis data due to their temporal resolution, which reduced the number of samples. All tests were performed with significance levels defined by a p-value < 0.05, using the R statistical package (R Core Team, 2023).

174 Rainfall weighted averages were calculated for each event to evaluate large-scale processes using the equation:

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$$\delta_{wgd} = \frac{\sum_{i=1}^{n} Ri\delta i}{\sum_{i=1}^{n} Ri}$$
(1)

where  $\delta_{wgd}$  is the rainfall weighted average of the isotopic composition, R<sub>i</sub> is the rainfall of the event (mm),  $\delta_i$  is the isotopic composition of an individual sample (‰), and n is the number of samples from each event. Rainfall weighted averages refers to the  $\delta^{18}O_{wgd}$ ,  $\delta^2H_{wgd}$  and  $d_{wgd}$ , and median of the  $\delta^{18}O_{med}$ ,  $\delta^2H_{med}$  and  $d_{med}$ .

## 179 3 Results

## 180 3.1 Inter-event variability of meteorological and isotopic parameters

# 181 3.3.1. Seasonal-mean climatic and isotopic conditions

The isotopic composition of monthly rainfall exhibits clear seasonal variations between September 2019 and February 2021 (Fig. 2a). Seasonal variability was characterized by wet (low  $\delta^{18}$ O) and dry (high  $\delta^{18}$ O) seasons (austral summer and autumnspring, respectively). Summer months were characterized by the influence of convective activity, reflected in high latent heat flux and lower OLR (Fig. 2 b-d). During autumn and spring, significant lower latent heat flux and higher OLR were associated with less convective development (Houze, 1997, 1989). The formation of convective rainfall may not be primarily controlled by diurnal thermal convection, as rainfall is more likely to be associated with frontal systems (Siqueira and Machado, 2004), as observed in the rainfall episodes during autumn and spring.

189 A significant influence of the cold fronts was observed before, during, and after their passage over the study area (Fig. 2a). 190 During autumn and spring, the convective events of 2019/11/05, 2020/11/18, and 2020/05/23 were associated with cold fronts 191 in the study area. On 2020/06/09, changes in the regional atmosphere over the state of São Paulo caused convective rainfall 192 due to an instability (frontal) system resulting from a cold front settling over the southern region of Brazil. During the summer 193 season, convective rainfall also occurred on 2020/02/01 and 2021/02/24 due to cold fronts and instability (frontal), 194 respectively. In addition, the thermal convection over land, lead to convective rainfall event on 2020/01/30. As a result of the 195 interaction between thermal convection and the incursion of the frontal system, a low-pressure system (frontal) was responsible 196 for the convective rainfall event on 2020/02/10.

### 197 3.3.2. Isotopic and local meteorological variations

Table 1 presents an overview of the sampling, isotope compositions ( $\delta_{med}$  and  $\delta_{wgd}$ ) and median values of meteorological variables from individual events. The duration of sampled events ranged from 141 min to 18 min. The T and T<sub>wd</sub> exhibited small differences among the events. In contrast, RR, RH, LCL, Z, w, and BT varied considerably between events. The maximum recorded values for these parameters were 97%, 489 m, 46 dBZ, 8 m s<sup>-1</sup> and -63 °C, respectively. Isotope values varied among convective events, with a range of -11.0 ‰, -91.2 ‰ and +15.7 ‰ for  $\delta^{18}O_{med}$ ,  $\delta^{2}H_{med}$  and  $d_{med}$ , respectively (Table 1).

#### 204 **3.1.3. Moisture origin**

The sourcing of moisture for rainfall over Rio Claro varies seasonally and spatially, suggesting complex interactions in moisture transport and mixing that strongly influence the  $\delta_{wgd}$  isotopic composition of rainfall throughout the year (Table 1). Hysplit air mass back-trajectories revealed three main domains as moisture origin during the presence of convective rainfall: Amazon forest, Atlantic Ocean, and southern Brazil (Fig. 3).

Summer rainfall events were characterized by the trajectory and length of moist air masses arriving from the Amazon forest
 (2020/02/10, 2020/02/01, and 2020/01/30) (Fig. 3a). As a result, there was a large amount of rainfall along Hysplit trajectories.

211 Rainfall amounts were 177 mm, 126 mm and 78 mm, respectively. Remarkably, these events exhibited similar isotope 212 characteristics ( $\delta^2 H_{wgd}$ ,  $\delta^{18} O_{wgd}$ ) (Table 1). In contrast, the event on 2021/02/24 presented higher  $\delta_{wgd}$  values, reflecting the 213 oceanic moisture influence (Fig. 3a), with a lowest amount of rainfall (53 mm) along Hysplit trajectory.

Based on ERA-5, the vertically integrated eastward vapor flux corroborates the influence of a distinct mechanism for moisture transport and  $\delta_{wgd}$  values. Negative values for vertical vapor fluxes over the Amazon forest during sampled convective events in summer (Fig. 4a, b, d) clearly illustrate a westward moisture flux from the Atlantic Ocean to the Amazon forest. Positive values in the central-southern region of Brazil indicate moisture being transported eastward from the Amazon forest. However, these moisture fluxes were not observed on 2021/02/24 when the eastward vapor flux was positive with high values over the Atlantic Ocean (250 ~ 750 kg m<sup>-1</sup> s<sup>-1</sup>).

220 The autumn convective events on 2020/05/23 and 2020/06/09 revealed a significant continental origin of moist air masses 221 (from south-western Brazil). In addition, during the second event, the Amazon-type trajectory started in the southern Atlantic 222 and did not reach the boundary of the rainforest (Fig. 3b). Both autumn events reported the lowest rainfall amounts (4 mm) 223 along Hysplit trajectories. On 2020/05/23 negative vertical flux values (-500 ~ -250 kg m<sup>-1</sup>s<sup>-1</sup>) were observed in south-western Brazil, indicating moisture transport from the Atlantic Ocean to the continent. This favored a vapor flux (500 ~ 750 kg m<sup>-1</sup>s<sup>-1</sup>) 224 from western Brazil to the study area (Figure 4f). On 2020/06/09, there were slightly negative values (-250 ~ 0 kg m<sup>-1</sup>s<sup>-1</sup>) of 225 226 eastward vapor flux in the Amazon forest, indicating less influence from rainforest moisture. Conversely, positive vapor flux values  $(250 \sim 500 \text{ kg m}^{-1}\text{s}^{-1})$  were observed in the western part of continental Brazil. 227

Two events in the spring season (Fig. 3c) also showed contrasting origin of moisture and  $d_{wgd}$  values, despite only slight 228 229 differences in  $\delta^{18}O_{wed}$  (Table 1). The mean trajectory on 2020/11/18 clearly belongs to the Amazon category, although it only 230 passed over the south-eastern boundary of the Amazon rainforest and had a much shorter length and lower rainfall along 231 Hysplit trajectory (23 mm) compared to the Amazon trajectories observed during the summer season. Thus, positive values of the eastward vapor flux (250 ~ 750 kg m<sup>-1</sup> s<sup>-1</sup>) were not distributed along the Amazon forest to the Atlantic Ocean as typically 232 observed (Fig. 4h). The mean trajectory on 2019/11/05 the eastward vapor flux (> 500 kg m<sup>-1</sup> s<sup>-1</sup>, Fig. 4g) were circling around 233 234 Rio Claro, indicating the continental moisture origin (from southern Brazil), and low amount of rainfall along Hysplit trajectory 235 of 8 mm.

#### 236 **3.2 Intra-event variability of the isotope and meteorological parameters**

The temporal evolution of isotope characteristics and selected meteorological parameters of convective rainfall are shown in Fig. 5-6 (summer), Fig. 7 (autumn) and Fig. 8 (spring). The vertical Z variation of the MRR in all events shows a pattern of values ranging from 0 to 10 dBZ at the top, a wide band of lowest values and noise attenuating the reflectivity producing white horizontal and vertical bands, and an increase in Z values closer to the surface where Zmax occurs (highest values ranging from 44 to 51 dBZ). During intra-events, Z, isotopic parameters, and GOES-16 BT display distinct temporal 242 patterns across events and seasons. There are large variations in Z values and inverse patterns between  $\delta^{18}$ O and *d*-excess

(more variable), and between T and RH. Different decreasing, increasing or stable trends were observed in BT values. Thefollowing sections described the main seasonal results for the intra-event analysis.

### 245 **3.2.1. During summer**

246 Low variability patterns were observed on 2020/02/01 and 2020/01/30 (Fig.5) for  $\delta^{18}$ O, T, RH, and BT. Both events 247 were shorter in duration ( $\leq 25$  minutes) and had a higher R ( $\leq 5.4$  mm) value at the beginning, which decreased over the course 248 of the event (0.2 mm). Similar MRR vertical profiles were observed between the events, illustrated by similar Z values, with low variability of Zmedian (7 ~ 17 dBZ and 8 ~ 15 dBZ), Zmax (23 ~ 48 dBZ and 19 ~ 46 dBZ) and Z amplitude decreasing 249 250 along the event (17 ~ 45 dBZ and 19 ~ 42 dBZ), respectively. Strong and significant (p < 0.0001) correlations were observed between isotopic composition and MRR parameters for 2020/02/01;  $\delta^{18}$ O-Z (r = -0.9),  $\delta^{18}$ O-W (r = -0.9),  $\delta^{18}$ O-Zmáx (r = -0.9), 251  $\delta^{18}$ O-Zampl (r = -0.8), d-excess-Z (r = 0.9), d-excess-W (r = 0.9), d-excess-Zmax (r = -.9) and d-excess-Zampl (r = 0.9). No 252 253 significant correlations between isotopic composition and meteorological parameters for 2020/01/30, except for a moderate correlation between  $\delta^2$ H and Zmedian (r = -0.5). 254

255 Large isotopic and meteorological variations were observed for 2021/02/24 ( $\delta^{18}$ O: -7.9 ~ -4.4 ‰, d-excess: +1.2 to 256 +18.4 ‰) and 2020/02/10 ( $\delta^{18}$ O: -15.2 ~ -7.9 ‰, d-excess: +4.8 ~ +21.4 ‰.) (Fig. 6). On 2021/02/24 a strong and significant (p < 0.05) correlation was observed between  $\delta^{18}$ O and R (r = -0.8), Z (r = -0.6), Zmax (r = -0.6), Zampl (r = -0.6), Zmedian (r 257 = 0.7), and between d-excess and R (r = -0.6), Z (r = -0.5), Zmax (r = 0.5), Zampl (r = 0.5) and Zmedian (r = -0.7). For 258 259 2020/02/10, significant correlations were reported between  $\delta^{18}$ O-RH (r = -0.5), d-excess-RH (r = 0.5) and d-excess and Zmedian (r = 0.5). In addition, d-excess values lower than  $\pm 10\%$  were observed at the end of the events (2020/02/01, 260 261 2020/02/10 and 2021/02/24), corresponding to low values of the R and Z parameters and high RH (black dotted cycle in Figs. 262 5 and 6).

## 263 3.2.2 During autumn and spring

Autumn events show distinct isotopic patterns. The 2020/05/23 event exhibited a small isotopic ( $\delta^{18}$ O: -2.6 ~ -2.7 ‰, d-excess: +16.7 ~ +19.0 ‰) and meteorological (declining T, R and Z parameters along the event) variation (Fig. 7). On 2020/06/09 (Fig. 7) two isotopic distribution patterns were recorded, with minimal ( $\delta^{18}$ O: -3.6 ~ -3.4 ‰, *d*-excess: +26.4 ~ +17.7 ‰) and large ( $\delta^{18}$ O: -1.5 ~ -2.9 ‰; *d*-excess: +15.3 ~ +6.3 ‰) variations, corresponding to high RH, R, Zamplt and Zmax (grey bands in Fig. 8). Strong and significant (p < 0.05) correlations were observed between  $\delta^{18}$ O-RH (r = 0.5),  $\delta^{18}$ O-T (r = -0.6), *d*-excess-RH (r = -0.6), *d*-excess-T (r = 0.7) and *d*-excess-Zamplt (r = -0.5) on 2020/06/09. However, no significant correlations were found during the event on 2020/05/23. 271 Distinctive isotopic patterns were also found during spring events. On 2019/11/05, a change in the vertical profile 272 and Z parameters was observed (grev bands in Fig. 8), with a shift in  $\delta^{18}$ O from maximum depletion (-4.1 ‰) to enrichment at the end of the event (-3.2 ~ -1.7 %). On 2020/11/18, there was a gradual decrease observed in  $\delta^{18}$ O (-2.7 ~ -5.4 %) and an 273 increase in d-excess ( $\pm 10.2 \sim \pm 23.1$  %). The latter was accompanied by a progressive increase in RH, decrease in T, and 274 275 constant Zmedian values (Fig. 8). On 2019/11/05 a strong and significant (p < 0.005) correlations were observed between  $\delta^{18}$ O 276 and Z (r = -0.7), w (r = -0.7), Zmax (r = -0.7) and Zampl (r = -0.6), and between d-excess and RH (r = -0.7), T (r = 0.8), w (r = 0.6), Zampl (r = 0.5) and Zmedian (r = -0.5). For 2020/11/18, significant correlations were obtained between  $\delta^{18}$ O-RH (r = 277 -0.5),  $\delta^{18}$ O-T (r = 0.7), *d*-excess-Zampl (r = 0.7). 278

### 279 4. Discussion

280 Detailed evaluations of the isotopic variability in convective rainfall were provided at both inter- and intra-event 281 scales. The key regional and local controls on the isotopic composition of convective rainfall can be divided in two groups: (i) rainfall produced by different moisture source region(s) represented by inter-event isotopic values, and (ii) local effects 282 283 associated with vertical rainfall structure and surface meteorological conditions. In the summer, thermal conditions dominate 284 convective processes. During autumn and spring, convective rainfall was associated with frontal systems (Fig. 2). In this regard.  $\delta_{wed}$  values better constrained the large-scale processes (such as vapor origin, convective activity and weather systems) 285 286 with stronger rainfall amount dependencies. The individual isotopic patterns influenced by local effects revealed microphysical processes such as coalescence (i.e., higher concentration of raindrops with high Z values) hat are often masked by weighted 287 288 averages and long-term averages during the evolution of individual precipitation systems.

#### 289 4.1 Regional atmospheric controls

290 Regional aspects of atmospheric moisture transport to Rio Claro were illustrated in HYSPLIT backward trajectories 291 (Fig. 3) and maps of vertically integrated moisture flux across the region (Fig. 4). Most of moist air masses arriving at Rio 292 Claro during summer exhibited a common origin in the equatorial Atlantic Ocean and were subjected to a long rainfall rainout, 293 extending over several thousand kilometers. Along this pathway, air masses interacted with the Amazon forest. Intensive 294 moisture recycling resulted in a small continental isotope gradient across the Amazon forest (Salati et al., 1979; Rozanski et 295 al., 1993) and elevated d-excess (Gat, J. R., & Matsui, 1991). At Rio Claro, the arriving air masses are depleted in heavy 296 isotopes ( $\delta_{wgd} \leq -10.0$  %) due to rainout along the trajectories ( $\geq 78$  mm), with consistent  $d_{wgd} > +14.0$  %, inherited through 297 the interaction of maritime moisture with the Amazon forest. In contrast, the summer event on 2021/02/24 was influenced by 298 oceanic moisture and had a short trajectory compared to other typical summer events. The convective events during spring and 299 autumn season exhibited substantially shorter trajectories suggesting that the atmospheric "pump" transporting moisture from

300 the equatorial Atlantic Ocean to the Amazon forest was much weaker or non-existent during this time of the year. Those short 301 trajectories suggest enhanced evapotranspiration of source moisture for rainfall (Salati et al., 1979; Risi et al., 2013; Gat, J. R., 302 & Matsui, 1991; Worden et al., 2007; Brown et al., 2008; Levin et al., 2009; Worden et al., 2021). As a result, those trajectories 303 were characterized by a reduction in the amount of rainfall along the trajectories and enriched  $\delta^{18}O_{wed} = \ge -4.2$  ‰ and higher 304  $d_{\text{wgd}} = \geq +16.5$  %. In addition, the highest  $d_{\text{wgd}}$  (+23.3 %) observed on 2019/11/05 was characterized by a continental moisture 305 trajectory circling around Rio Claro (Fig. 3c) over a greater RH gradient (e.g., sugar cane crop regions) (da Silva et al., 2021). 306 Evaporation from soil increases kinetic fractionation, favoring the evaporation of HDO due to high diffusivity, resulting in a 307 strong *d*-excess changes (Risi et al., 2013).

## 308 4.2 Local atmospheric controls

309 In deep convection, precipitation particles primarily grow through the collection of water (known as coalescence) by larger droplets and/or ice particles (known as riming). These larger particles sweep water from the cloud on their falling paths 310 311 in the presence of strong rising air currents. As a parcel of rising air ascends, the growing particles within it move until they 312 become large enough to fall relative to the air. As the air parcel ascends, particles fall out at each successive height. The 313 remaining lighter particles disperse laterally over a larger area due to the diverging airflow. Convective air movements create 314 concentrated reflectivity peaks in the radar pattern because most of the precipitation mass falls within a few kilometres of the 315 updraft centres (Houghton, 1968; Houze, 1997). The irregular blank bands visible in the vertical MRR profiles (Fig. 5) could 316 be attributed to the lateral dispersion of remaining particles. The concentrated high reflectivity values (Fig. 5) illustrate this 317 pattern, which typically occurs close to the surface and indicates the occurrence of Zmax.

318 Variations in the isotopic composition of the rainfall reflect changes in this mechanism of raindrop formation (Sun et 319 al., 2022; Aggarwal et al., 2016). This is shown by the vertical variation in the Z values of the events on 2020/02/10, 320 2021/02/24, 2020/06/09 and 2019/11/05 (grey band in Fig. 6, 7, 8). A possible reason for this change is that the process of 321 coalescence during the falling raindrops towards the surface may have been altered. This can be seen in the higher Zampl 322 values (40  $\sim$  50 dBZ), which suggest that water particles were being incorporated into the raindrop during the fall at the surface, 323 resulting in a larger water particle and consequently a higher concentration in the raindrops and the occurrence of Zmax close 324 to the surface. The  $\delta^{18}$ O values generally increased while the *d*-excess decreased, resulting in a change in the isotopic variation 325 pattern, reflecting the diffusive exchange process between the surrounding vapor and the raindrops (Gedzelman and Lawrence, 326 1990; Celle-Jeanton et al., 2004). In contrast, the isotopic pattern of the events on 2020/02/01 (Fig. 5), 2020/01/30 (Fig. 5), 327 and 2020/05/23 (Fig. 7) exhibited small variation due to the low variability in Z values. Therefore, the main local control on 328 the isotopic variability of intra-events corresponded to the vertical structure of the rainfall event.

The *d*-excess values decreased and the  $\delta^{18}$ O values increased at the end of the events on 2020/02/01, 2021/02/24 and 2020/02/10 (black dotted cycle in Figs. 5a, 6a and 6b, respectively). This was due to the formation of residual rainfall at low altitudes and a decrease in rainfall intensity during the dissipation phase of the convective cell. On 2020/11/18 (Fig. 8b), the  $\delta^{18}$ O values constantly decreased, illustrating a typical depletion of heavy isotopes based on Rayleigh distillation processes due to the progressive condensation of convective systems. Previous studies have widely observed these mechanisms during intra-events, and both interpretations are supported (Adar et al., 1991; Coplen et al., 2008, 2015; Barras and Simmonds, 2009; Celle-Jeanton et al., 2004; Muller et al., 2015).

### 336 5 Concluding remarks

337 This study used high-frequency isotopic composition of rainfall as well as meteorological data to investigate the 338 regional and local mechanisms controlling the isotopic characteristics of convective precipitation. Based on the inter-event 339 analysis, the regional isotopic characteristics are different between summer and autumn-spring seasons. The  $\delta_{wed}$  is determined 340 by moisture transport mechanisms and convection features. The main factors are the gradual reduction of heavy isotopes along 341 moisture trajectories. The rainfall produced along these tracks was pronounced during summer events, associated with the 342 longer moisture transport pathway from the Amazon Forest, producing depleted isotope values. In contrast, reduced autumn 343 and spring rainfall along trajectories is associated with a shorter moisture transport pathway from the Atlantic Ocean and 344 southern Brazil. This produces enriched isotope characteristics and high  $d_{wgd}$  values associated with transpiration and soil 345 evaporation along the moisture transport pathway.

346 Within convective events, the regional  $\delta_{wed}$ -signature was altered by local effects, as shown by the intra-event isotopic 347 evolution. The vertical structure of rainfall, described by the Z parameters in the vertical MRR profile, is the main local control. 348 During falling raindrops, a microphysical change can cause a vertical change in Z values, resulting in abrupt variations in isotopic patterns. These findings were supported by significant and strong correlations between the MRR and isotopic 349 350 parameters in each event. Therefore, the isotopic composition of convective rainfall is controlled by an interplay of regional 351 and local factors. The complex and dynamic conditions of convective rainfall formation across the tropics can be understood 352 using high-frequency analysis. Although high-frequency rainfall sampling is logistically difficult, we encourage future studies 353 of this type in different geographical regions across the tropics, to better understand the factors controlling the isotopic 354 composition of convective rainfall during rainy period. Extensive monitoring of local meteorological parameters and modeling 355 of regional moisture transport to the rainfall collection site, along with the application of more robust below-cloud models, 356 should accompany such studies.

#### 357 Data availability

A complete database (isotope characteristics of rainfall as well as selected meteorological parameters characterizing these
events) are available at: <u>https://doi.org/10.17632/kk3gs8zn4s.1</u> (dos Santos et al., 2023). Monthly GNIP data:
<u>https://www.iaea.org/services/networks/gnip</u>. GOES-16 imageries are available at:
<u>https://home.chpc.utah.edu/~u0553130/Brian\_Blaylock/cgi-bin/goes16\_download.cgi</u>. The weather systems are available at:

- https://www.marinha.mil.br/chm/dados-do-smm-cartas-sinoticas/cartas-sinoticas 362 and 363 http://tempo.cptec.inpe.br/boletimtecnico/pt. Reanalysis data available are at: 364 (https://cds.climate.copernicus.eu/cdsapp#!/search?type=dataset. The Global Modeling and Assimilation Office (GMAO) data are available at: https://goldsmr4.gesdisc.eosdis.nasa.gov/data/MERRA2/M2T1NXFLX.5.12.4/). 365 366 Goddard Earth Sciences Data and Information Services Center (GES DISC) data are available at: https://disc.gsfc.nasa.gov/datasets/AIRS3STD 7.0/summary. 367 368
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Season		Spring		Autumn		Summer			
Data		05/11/2019	18/11/2020	09/06/2020	23/05/2020	30/01/2020	10/02/2020	01/02/2020	24/02/2021
Number of samples		21	8	12	4	6	18	5	16
Duration		82	141	96	131	23	86	18	55
δ <sup>18</sup> Ο	Median	-3.1	-4.2	-3.4	-2.9	-10.0	-13.9	-10.4	-6.8
	Weighted average	-3.0	-4.2	-2.7	-2.9	-10.0	-13.4	-11.1	-7.2
$\delta^2 H$	Median	0.8	-13.7	-5.6	-6.9	-64.4	-92.0	-73.5	-44.8
	Weighted average	-1.2	-14.9	-4.9	-6.8	-63.9	-90.4	-75.0	-47.2
d-excess	Median	22.9	19.7	17.3	16.3	15.7	17.5	13.4	7.2
	Weighted average	23.3	19.1	17.3	16.5	16.5	16.7	14.2	11.1
Automatic Weather Station	Rain rate	0.4	0.2	0.3	0.0	0.4	0.5	0.6	0.5
	RH	96	86	95	87	93	97	93	86
	Т	21	20	19	19	23	22	23	21
	Tdw	20	17	18	17	21	21	21	18
	LCL	146	489	168	449	247	93	253	468
Micro Rain Radar	Z	46	38	42	33	38	41	39	35
	W	8	7.1	7.7	6.6	6.6	6.7	7.1	7.1
GOES-16	BT	-63	-63	-50	-56	-53	-39	-60	-51

Table 1. Summarizing overall convective rainfall events, isotope and meteorological parameters

Duration (minutes); Isotopes parameters (‰); Median values of meteorological variables: Rain rate (mm.min<sup>-1</sup>), Relative Humidity – (RH %), Temperature (T °C), Dew Temperature (Tdw °C), Lifting Condensation Level (LCL meters), Reflectivity (Z dBZ), Vertical Velocity (m.s<sup>-1</sup>) and Brightness temperature (BT °C).





**Figure 1**. Regional and local context of study area. (a) Localization of sampling site in Rio Claro (black star) (b) regional synoptic context across Brazil and main weather systems (CF – cold front and SACZ – Southern Atlantic Convergence Zone). (c) GOES-16 satellite imagery of convective rainfall (d) Micro Rain Radar (MRR) image of convective rainfall.



**Figure 2.** Seasonal variation of isotope and convective parameters. (a) Temporal distribution of monthly  $\delta^{18}$ O and *d*-excess values during study period, with aggregated median of  $\delta^{18}$ O values for high-frequency convective rainfall events (b) AQUA/AIRS latent heat flux. (c) MERRA-2 outgoing longwave radiation (monthly averaged daytime and night-time data) (d) monthly rainfall amounts at Rio Claro separated into day and night fraction (no rainfall types distinguished). The black symbol indicates weather systems described in section 3.1. The monthly isotopic composition used in this figure was collected by the first authors of the article and determined by the UNESP laboratory, following the same procedures mentioned in section 2.2.



Figure 3. Ten-day backward trajectories arriving at Rio Claro station of eight convective events. (a) Summer, (b) Autumn and (c) Spring.
 Twenty-seven ensembles are grey lines, and the mean trajectory is the colors lines. The colours of the mean trajectories indicate the origin of air masses. The authors used trivial information, the borders of the countries and the ocean provided by the ESRI base map.



**Figure 4.** ERA-5 vertical integral of eastward water vapor flux. (a, b, c, d) summer convective events (e, f) autumn and (g, h) spring aggregated. The maps corresponded to the days when convective rainfall events occurred. Positive values indicate the direction of moisture vapor flux from left to right, and negative values from right to left.



Figure 5. Summer intra-events. (a, b)  $\delta^{18}$ O (red lines) and *d*-excess (purple lines) (c, d) Temperature (orange lines) and Relative Humidity (blue lines) (e, f) brightness temperature (BT – green bars) and rainfall amount (blue lines) (g, h) Reflectivity parameters, Z maximum (red lines), Z amplitude (green lines) and Z median (black lines) (i, j) radar reflectivity of Micro Rain Radar. The black dotted cycle refers to the low *d*-excess value.



Figure 6. Summer intra-events. Refer to Fig. 5 for legend description.



Figure 7. Autumn intra-events. Refer to Fig. 5 for legend description.



Figure 8. Spring intra-events. Refer to Fig. 5 for legend description.