Rain evaporation, snow melt and entrainment at the heart of water vapor isotopic variations in the tropical troposphere, according to large-eddy simulations and a two-column model

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Abstract

The goal of this study is twofold. First, we aim at developing a simple model as an interpretative framework for the water vapor isotopic variations in the tropical troposphere over the ocean. We use large-eddy simulations to justify the underlying assumptions of this simple model, to constrain its input parameters and to evaluate its results. Second, we aim at interpreting the depletion of the water vapor isotopic composition in the lower and mid-troposphere as precipitation increases, which is a salient feature in tropical oceanic observations. This feature constitutes a stringent test on the relevance of our interpretative framework. Previous studies, based on observations or on models with parameterized convection, have highlighted the roles of deep convective and meso-scale downdrafts, rain evaporation, rain-vapor diffusive exchanges and mixing processes. The interpretative framework that we develop is a two-column model representing the net ascent in clouds and the net descent in the higher tropospheric relative humidity. First, when the relative humidity is larger, less snow sublimates before melting and a smaller fraction of rain evaporates. Both effects lead to more depleted rain evaporation and eventually more depleted water vapor. This mechanism dominates in regimes of large-scale ascent. Second, the entrainment of dry air into clouds reduces the vertical isotopic gradient and limits the depletion of tropospheric water vapor. This mechanism dominates in regimes of large-scale descent.

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Key Points:

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10	•	Isotopic enrichment of tropospheric water vapor by rain evaporation is stronger
11		when drier air enhances sublimation and evaporation
12	•	Entrainment of dry air weakens the vertical isotopic gradient and limits the de-
13		pletion of tropospheric water vapor.
14	•	These mechanisms explain the increased depletion of tropospheric water vapor as
15		tropospheric relative humidity increases.

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16 Abstract

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The interpretative framework that we develop is a two-column model represent-27 ing the net ascent in clouds and the net descent in the environment. We show that the 28 mechanisms for depleting the troposphere when precipitation rate increases all stem from 29 the higher tropospheric relative humidity. First, when the relative humidity is larger, less 30 snow sublimates before melting and a smaller fraction of rain evaporates. Both effects 31 lead to more depleted rain evaporation and eventually more depleted water vapor. This 32 mechanism dominates in regimes of large-scale ascent. Second, the entrainment of dry 33 air into clouds reduces the vertical isotopic gradient and limits the depletion of tropo-34 spheric water vapor. This mechanism dominates in regimes of large-scale descent. 35

³⁶ Plain Language Summary

Water molecules can be light (one oxygen atom and two hydrogen atoms) or heavy 37 (one hydrogen atom is replaced by a deuterium atom). These different molecules are called 38 water isotopes, and their relative concentration in water is called the isotopic composi-39 tion. The isotopic composition of the precipitation recorded in ice cores or in speleothems 40 can be used to reconstruct past climates. However, the factors controlling the isotopic 41 composition are complex. Here we aim at developing a simple model as an interpreta-42 tive framework for the water vapor isotopic variations in the tropical troposphere over 43 the ocean. As a guide for developing this framework, we use high-resolution atmospheric 44 simulations that explicitly simulates vertical motions in the storms. As a test for this 45 framework, we try and interpret why in observations, the precipitation and water vapor 46 are more depleted when storm activity is stronger. We find that stronger storm activ-47 ity, when associated with stronger large-scale ascent, is associated with a moister tro-48 posphere. This reduces the sublimation of snow, the fraction of rain that evaporates and 49 the dilution of cloudy air by entrainment, ultimately leading to more depleted water va-50 por and precipitation. 51

52 1 Introduction

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1.1 Looking for an interpretative framework for water vapor isotopic profiles

The isotopic composition of water vapor (e.g. its Deuterium content, commonly 55 expressed as $\delta D = (R/R_{SMOW} - 1) \times 1000$ in %, where R is the ratio of Deuterium 56 over Hydrogen atoms in the water, and SMOW is the Standard Mean Ocean Water ref-57 erence) evolves along the water cycle as phase changes are associated with isotopic frac-58 tionation. Consequently the isotopic composition of precipitation recorded in paleoclimate archives has significantly contributed to the reconstruction of past hydrological changes 60 (Wang et al., 2001). It has also been suggested that observed isotopic composition of wa-61 ter vapor could help better understand atmospheric processes and evaluate their repre-62 sentation in climate models, in particular convective processes (Schmidt et al., 2005; Bony 63

et al., 2008; Lee et al., 2009; Field et al., 2014). Yet, water isotopes remain rarely used beyond the isotopic community to answer today's pressing climate questions. A prerequisite to better assess the strengths and weaknesses of the isotopic tool is to better understand what controls spatio-temporal variations in water vapor isotopic composition (δD_v) through the tropical troposphere, and in particular how convective processes drive these variations.

While there are interpretative frameworks for the controls of free tropospheric hu-70 midity (Sherwood, 1996; Romps, 2014), no such interpretative framework exist for wa-71 ter isotopes beyond the simple Rayleigh distillation or mixing lines (Worden et al., 2007; 72 Bailey et al., 2017). We aim at filling this gap here. The first goal of this paper is thus 73 to design an interpretative framework that could be useful in the future to interpret wa-74 ter vapor isotopic variations in the tropical troposphere in a wide range of contexts. Anal-75 ogous to that for relative humidity, this framework will also allow us to compare the pro-76 cesses controlling relative humidity and isotopic composition. 77

Frameworks do exist to interpret the δD_v in the sub-cloud layer (SCL), such as the Merlivat and Jouzel (1979) closure assumption, later extended to account for mixing with free tropospheric air (Benetti et al., 2015) and for updrafts and downdrafts (Risi et al., 2020). This latter framework highlighted the need to know the steepness of the relationship between δD_v and specific humidity q as they evolve with altitude. This motivates us to develop a framework that allows us to predict the δD_v evolution with altitude in the troposphere.

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1.2 Large-eddy simulation analysis as a guide to design the interpretative framework

Many previous studies investigating the processes controlling tropospheric δD_v have 87 relied on general circulation models that include convective parameterization (Lee et al., 88 2007; Bony et al., 2008; Risi et al., 2008; Field et al., 2010). However, parameterizations 89 include numerous simplifications or assumptions that are responsible for a significant part 90 of biases in the present climate simulated by GCMs and of inter-model spread in climate 91 change projections (Randall et al., 2003; Stevens & Bony, 2013; Webb et al., 2015). Here, 92 we thus use large-eddy simulations (LES) as a guide to design the interpretative frame-93 work. These high-resolution simulations allows us to explicitly resolve convective mo-94 tions. These simulations will also provide the input parameters for our interpretative frame-95 work, and a benchmark to evaluate its results. 96

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1.3 Interpreting the amount effect

In the tropics, it has long been observed that in average over a month or longer, 98 the isotopic composition of the rain is more depleted when the precipitation rate is stronger 99 (Dansgaard, 1964; Rozanski et al., 1993). This phenomenon is called the "amount ef-100 fect". Since most of the precipitation in the tropics is associated with deep convection, 101 understanding the amount effect is a stringent test on our understanding of how convec-102 tive processes affect the water isotopic composition in the tropical troposphere. The ca-103 pacity of our interpretative framework to predict the amount effect will thus be a strin-104 gent test on its relevance. The second goal of this study is thus to better understand the 105 processes underlying the amount effect, using the interpretative framework. 106

Dansgaard (1964) hypothesized that the amount effect could be due to the progressive depletion by convective storms of the vapor from which the rain forms, and to rain evaporation and diffusive exchanges between the rain and the vapor. If the case, the amount effect crucially depends on the isotopic composition of the vapor. From a column-integrated water budget perspective, the isotopic composition of precipitation depends on the relative proportion of the precipitation that originates from horizontal advection and from surface evaporation (Lee et al., 2007; Moore et al., 2014). More precipitation is generally associated with more large-scale ascent and thus more large-scale convergence. Since
vapor from horizontal advection is more depleted than water from surface evaporation
because it has already been processed in clouds, the precipitation is more depleted. In
this view as well, the amount effect crucially depends on the isotopic composition of the
vapor.

Water isotopic measurements in the vapor phase, by satellite or in-situ, have confirmed that increased precipitation was associated with more depleted water vapor (Worden et al., 2007; Kurita, 2013; Lacour et al., 2017). Hereafter we will call this the "vapor amount effect". Actually, the precipitation and water vapor isotopic composition often vary in concert (Kurita, 2013; Tremoy et al., 2014). In this paper, we will thus focus on understanding the processes underlying the "vapor amount effect".

From previous studies, four hypotheses have emerged to explain the "vapor amount effect":

- 1. Hypothesis 1: As precipitation rate increases, convective or meso-scale downdrafts 127 bring more depleted vapor from above into the sub-cloud layer (SCL) (Risi et al., 128 2008; Kurita et al., 2011; Kurita, 2013). This is because δD_v generally decreases 129 with altitude, because as water vapor is lost through condensation and q decreases, 130 heavy isotopes are preferentially lost in the condensed phase. This phenomenon 131 is called Rayleigh distillation and is plotted in a $q - \delta D_v$ diagram in Figure 1 (blue). 132 However, downdrafts would both decrease δD_v and q. This hypothesis is thus in-133 consistent with the observation that q generally increases while δD_v decreases as 134 precipitation rate increases. By itself, this hypothesis cannot be sufficient. 135
- 2. Hypothesis 2: As precipitation rate increases, the moistening effect by rain evaporation increases. If rain evaporation is more depleted than the vapor, then it depletes the vapor (Worden et al., 2007). The effect of rain evaporation is represented in purple in Figure 1. If the evaporated fraction of the rain is small, rain evaporation acts to deplete the vapor because light isotopes preferentially evaporate.
- 3. Hypothesis 3: As precipitation rate increases, the rain evaporation is more depleted. 141 For example, if precipitation rate increases, the fraction of rain that evaporates 142 is smaller. Because heavy isotopes diffuse through air more slowly than $H_2^{16}O$, the 143 initial vapor produced by rain evaporation is more depleted than the average iso-144 topic composition of the rain. As a larger fraction of the raindrop evaporates, the 145 vapor produced by evaporation becomes less depleted and can sometimes be more 146 enriched than the surrounding vapor (Risi et al., 2008, 2010; Tremoy et al., 2014; 147 Risi et al., 2020) (Figure 1, purple). Alternatively, larger precipitation rates typ-148 ically occur in moister environments, which favors rain-vapor diffusive exchanges 149 rather than pure evaporation (Lawrence et al., 2004; Lee & Fung, 2008). Since rain 150 comes from higher altitudes, it is more depleted than if in equilibrium with the 151 local vapor, and thus rain-vapor diffusive exchanges favor more depleted evapo-152 ration. 153
- 4. Hypothesis 4: As precipitation rate decreases, dehydration by mixing dominates 154 relatively to dehydration by condensation. Due to the hyperbolic shape of the mix-155 ing lines in a $q - \delta D$ diagram, dehydration by mixing with a dry source is asso-156 ciated with a smaller depletion than predicted by Rayleigh distillation (Dessler 157 & Sherwood, 2003; Galewsky & Hurley, 2010; Galewsky & Rabanus, 2016) (Fig-158 ure 1 orange). Bailey et al. (2017) argues that in more subsiding regions, mid-tropospheric 159 vapor is more enriched for a given q because air masses result from the mixing be-160 tween air subsiding from a higher altitude and shallow convective detrainment. 161

We notice that hypothesis 2-4 are all associated with an increased steepness as precipitation rate increases (Figure 1), consistent with the key role of the steepness of the $q-\delta D_v$ relationship in depleting the SCL water vapor highlighted by Risi et al. (2020). The



Figure 1. Schematic showing the influence of different processes on q and δD_v . Condensation and immediate loss of condensate in convective updrafts leads to drying and depleting the water vapor following Rayleigh distillation (blue). During evaporation of cloud droplets, each droplet evaporates totally. Since cloud droplets are enriched in heavy isotopes, this moistens the air and enriches the vapor (cyan). In contrast, during evaporation of rain drops, each drop evaporates progressively. Whereas it moistens the air, it depletes the vapor for small evaporation fractions and enriches the vapor for large evaporation fraction (purple). Finally, mixing of subsiding air with air detrained from convective updrafts dehydrates the air and depletes the vapor following a hyperbolic curve, leading to higher δD_v for a given q compared to Rayleigh (orange). The curves are plotted following simple Rayleigh and mixing lines with approximate values taken from the control LES described later in the article.

mechanisms underlying these hypotheses will thus have to be key ingredients of our in terpretative framework.

The LES will be described and analyzed in section 2. The interpretative framework will be designed and used to interpret the "vapor amount effect" in section 3. Finally, section 4 will offer a summary, some discussion and perspectives.

¹⁷⁰ 2 Large-eddy simulations

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2.1 Model and simulations

We use the same LES model as in Risi et al. (2020), namely the System for Atmo-172 spheric Modeling (SAM) non-hydrostatic model (M. F. Khairoutdinov & Randall, 2003), 173 version 6.10.9, which is enabled with water isotopes (Blossey et al., 2010). This model 174 solves anelastic conservation equations for momentum, mass, energy and water, which 175 is present in the model under six phases: water vapor, cloud liquid, cloud ice, precipi-176 tating liquid, precipitating snow, and precipitating graupel. We use the bulk, mixed-phase 177 microphysical parameterization from Thompson et al. (2008) in which water isotopes were 178 implemented (Moore et al., 2016). 179

The control simulation ("ctrl") is three-dimensional, with a doubly-periodic domain of 96 km×96 km. The horizontal resolution is 750 m. There are 96 vertical levels. The simulation is run in radiative-convective equilibrium over an ocean surface. The sea surface temperature (SST) is 30°C. There is no rotation and no diurnal cycle. In this simulation, there is no large-scale circulation.

The amount effect can be seen only if the precipitation increase is associated with 185 a change in the large-scale circulation (Bony et al., 2008; Dee et al., 2018; Risi et al., 2020). 186 To compare ctrl to simulations with larger and smaller precipitation rate, we thus run 187 simulations with a prescribed large-scale vertical velocity profile, ω_{LS} . This profile is used 188 to compute large-scale tendencies in temperature, humidity and water vapor isotopic com-189 position. We compute large-scale vertical advection by a simple upstream scheme (Godunov, 190 1959). In the computation, large-scale horizontal gradients in temperature, humidity and 191 isotopic composition are neglected, i.e. there are no large-scale horizontal advective forc-192 ing terms. The large-scale vertical velocity ω_{LS} has a cubic shape so as to reach its max-193 imum ω_{LSmax} at a pressure p_{max} =500 hPa and to smoothly reach 0 at the surface and 194 at 100 hPa (Bony et al., 2008). We analyze here simulations with ω_{LSmax} =-60 hPa/d 195 ("HighPrec"), corresponding to typical deep convective conditions in the inter-tropical 196 convergence zone, and ω_{LSmax} = +20 hPa/d ("LowPrec"), corresponding to subsiding trade-197 wind conditions. The mean precipitation rates are 1.5, 2.5 and 8.5 mm/d respectively in LowPrec, ctrl and HighPrec. 199

The simulations are run for 50 days and the last 10 days are analyzed. We use instantaneous outputs that are generated at the end of each simulation day.

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2.2 Simulated amount effect and basic features

Figure 2a shows that the ctrl, HighPrec and LowPrec simulations allow us to cap-203 ture the amount effect both in the near-surface vapor and in the precipitation, which vary 204 in concert. In HighPrec, the domain-mean relative humidity h is larger than in ctrl by 205 more than 10% (Figure 2b), while δD_{η} is more depleted by more than 50%, in most of 206 the troposphere (Figure 2c). We can see that the δD_v difference at all altitudes is sim-207 ilar to that in the SCL. This confirms that understanding what controls the SCL δD_v is key to understand what controls δD_v at all altitudes (Risi et al., 2020). This also ex-209 plains why models that assume constant SCL δD_v show very little sensitivity to all kinds 210 of convective and microphysical processes (Duan et al., 2018). We can also see that Rayleigh 211 distillation alone (dashed line) is a poor predictor of δD_v profiles and of their sensitiv-212 ity to large-scale circulation. 213

2.3 Steepness of the $q - \delta D_v$ relationship

With the goal of understanding the amount effect, as a first step Risi et al. (2020)215 focused on understanding what controls the δD_v in the SCL, because the SCL ultimately 216 feeds the water vapor at all altitudes in the troposphere. They identified the key role of 217 the steepness of the $q - \delta D_v$ relationship of vertical profiles in the lower troposphere. 218 This steepness determines the efficiency with which updrafts and downdrafts near the 219 SCL top deplete the SCL. To understand what controls δD_v in the SCL and thus ev-220 erywhere in the troposphere, we thus need to understand what controls the steepness 221 of the $q - \delta D_v$ relationship. 222

The vertical profiles of $ln(R_v)$ as a function of ln(q) for each simulation show a nearly linear relationship (Figure 2d), consistent with a Rayleigh-like distillation process (Figure 1). If the vertical profiles were dominated by mixing processes, as in hypothesis 4, the relationship would look concave down (Bailey et al., 2017) (Figure 1 orange). Rather, in HighPrec, the curve looks concave up near the melting level, consistent with an effect of rain evaporation (Figure 1 purple).

To better quantify the steepness of the $q - \delta D_v$ relationship, we define the $q - \delta D_v$ steepness α_z , as the effective fractionation coefficient that would be needed in a distillation to fit the simulated joint $q - \delta D_v$ evolution (Risi et al., 2020):

$$\alpha_z = 1 + \frac{\ln \left(\frac{R_v(z)}{R_v(z - dz)} \right)}{\ln \left(\frac{q(z)}{q(z - dz)} \right)} \tag{1}$$

The steepness α_z in the ctrl simulation is smaller than that predicted by Rayleigh 232 distillation, i.e. $\alpha_z < \alpha_{eq}$, especially at higher altitudes (Figure 2e) (section 3.2.2 will 233 demonstrate that it is due to entrainment). Just above the SCL top, $\alpha_z - 1$ is more than 234 three times larger in HighPrec than in ctrl. The increased steepness leads the updrafts 235 and downdrafts to deplete more efficiently the SCL water vapor (Risi et al., 2020), and 236 eventually the full tropospheric profile through mixing by deep convection. Conversely, 237 in LowPrec, the steepness is smaller and responsible for more enriched SCL. Our inter-238 pretative framework will allow us to interpret these features (section 3). 239

2.4 Effect of de-activating rain-vapor exchanges

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According to hypotheses 2 and 3, the isotopic composition of the rain plays a key role in the "vapor amount effect". At a given instant and for a small increment of rain evaporation fraction, the isotopic composition of the evaporation flux R_{ev} is simulated following Craig and Gordon (1965):

$$R_{ev} = \frac{R_r / \alpha_{eq} - h_{ev} \cdot R_v}{\alpha_K \cdot (1 - h_{ev})}$$

where R_r and R_v are the isotopic ratios in the liquid water and water vapor, α_{eq} and α_K are the equilibrium and kinetic fractionation coefficient and h_{ev} is the relative humidity. In order to test hypotheses 2 and 3, we run additional simulations similar to ctrl and HighPrec but without any fractionation during rain evaporation, named "nofrac", where $R_{ev} = R_r$. We also run additional simulations with fractionation during evaporation, but with rain-vapor diffusive exchanges de-activated, named "nodiff", where $R_{ev} =$ $R_r/\alpha_{eq}/\alpha_K$.

When fractionation during rain evaporation is de-activated, δD_v is more enriched, 252 consistent with a more enriched composition of rain evaporation (Figure 3a). In addi-253 tion, the δD_v difference between HighPrecand ctrl is reduced by about 70% compared 254 to when all isotopic exchanges are considered (Figure 3c, red). This confirms that frac-255 tionation during rain evaporation plays a key role in the "vapor amount effect". When 256 rain-vapor diffusive exchanges are de-activated, the δD_v difference between HighPrec and 257 ctrl is reduced by about 30% compared to when all isotopic exchanges are considered (Fig-258 ure 3c, green). Rain-vapor vapor diffusive exchanges thus play an important role as well. 259

We note that the δD_v difference between the simulations is remarkably constant 260 with altitude (Figure 3a,c), although we expect strong vertical variations in rain evap-261 oration. This is consistent with the important role of the SCL δD_v as an initial condi-262 tion for the full δD_v profile. We also note that more enriched δD_v profiles are associated 263 with a reduced lower-tropospheric steepness α_z just above the SCL, and larger δD_v dif-264 ferences between simulations are associated with larger differences in lower-tropospheric 265 α_z . This is consistent with the SCL δD_v being mainly driven by the steepness α_z just 266 above the SCL (Risi et al., 2020). Finally, the reduced "vapor amount effect" in "nofrac" 267 leads to a reduced amount effect in the precipitation δD as well (Figure 3c, circles). This 268 shows that the column-integrated water budget (Lee et al., 2007; Moore et al., 2014) can-269 not by itself predict the amount effect, since it depends on the isotopic composition of 270 the advected vapor, which can greatly vary depending on the detailed representation of 271 rain evaporation processes. 272

To summarize, in the total δD_v difference between HighPrec and ctrl, there is about one third due to fractionation during evaporation, one third due to rain-vapor diffusive



Figure 2. (a) Domain-mean water vapor (circles) and precipitation (squares) δD_v as a function of precipitation rate. Vertical distribution of relative humidity (b), δD_v (c) and α_z (e) in ctrl (black), HighPrec (blue) and LowPrec (orange). (d) $ln(R_v(z)) \cdot 1000$ as a function of ln(q(z)) for different altitudes. In c and e, dashed lines indicate the prediction by Rayleigh distillation. The horizontal lines show the altitude of the melting level.



Figure 3. (a) Vertical distribution of δD_v for ctrl, when fractionation during liquid evaporation is turned on (black) or off (red) and when liquid-vapor equilibration is turned off (green). (b) Same as (a) for the vertical profiles of α_z .

(c) δD_v difference between the HighPrec and ctrl, with (black) and without (red) fractionation during evaporation and when liquid-vapor equilibration is turned off (green). The circles illustrate the difference in the precipitation δD .

(d) Same as (c) but for α_z .

exchanges, and one third that would remain even in absence of any fractionation during evaporation. These tests suggest that hypotheses 2 and/or 3 play a key role in the "vapor amount effect". In the next sections, we aim at better understanding how rain evaporation impacts δD_v profiles.

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2.5 Vertical profiles binned by moist static energy

Previous studies have shown that analyzing variables in isentropic coordinates was a powerful tool to categorize the different convective structures: undiluted updrafts, diluted updrafts, saturated and unsaturated downdrafts, and the environment (Kuang & Bretherton, 2006; Pauluis & Mrowiec, 2013). This method also has the advantage of filtering out gravity waves. It has been applied to the analysis of a wide range of convective systems (Mrowiec et al., 2015, 2016; Dauhut et al., 2017; Chen et al., 2018).

Here we use the frozen moist static energy m as a conserved variable because it is conserved during condensation and evaporation of both liquid and ice water (C. J. Muller & Romps, 2018; Hohenegger & Bretherton, 2011).

$$m = c_{pd} \cdot T + g \cdot z + L_v \cdot q_v - L_f \cdot q_i$$

where c_{pd} is the specific heat of dry air, T is temperature, g is gravity, z is altitude, L_v and L_f are the latent heat of vaporization and fusion, and q_i is the total ice water content (cloud ice, graupel and snow). At each level, we categorize all grid points into bins of m with a width of 0.4 kJ/kg.

The domain-mean m decreases from the upper troposphere down to about 5 km, due to the loss of energy by radiative cooling, and then increases down to the surface due to the input of energy by surface fluxes (Figure 4, solid black line). Based on this diagram, we can identify four kinds of air parcels:

- Environment. They correspond to air parcels whose m is close to the domain-mean (solid black). They are the most numerous (Figure 4a). Their vertical velocity is slightly descending (Figure 4b), but because they are very numerous, they account for most of the downward mass flux (Figure 4c). Their relative humidity is close to the domain-mean (Figure 4d), they contain only a small cloud water and rain content and phase changes are very slow (Figure 4e-g). However, because they cover most of the domain, they contribute significantly to the evaporation in the domainmean (Figure 4h).
- 2. Cloudy updrafts. They correspond to air parcels on the right of the domain-mean m and whose bin-mean vertical velocity is ascending (Figure 4b). If air rose adi-306 abatically from the SCL, they would conserve their m and they would be located 307 completely on the right of the diagram. In practice, m decrease because the en-308 vironment air is progressively entrained into ascending parcels. In the diagrams, 309 parcels are more diluted when they are closer to the domain-mean, and less di-310 luted when they are more to the right. In spite of their dilution with the environ-311 ment, their humidity is at saturation (Figure 4d). They contain a lot of cloud and 312 precipitating water, and vapor undergoes condensation (Figure 4e-g). 313
- 3. Cloudy downdrafts. They correspond to air parcels on the right of the domainmean m and whose bin-mean vertical velocity is descending (Figure 4b). They are more diluted than cloudy updrafts. Their humidity is below saturation (Figure 4d). They contain cloud and precipitating water that undergo evaporation (Figure 4e-g). Located around the cloudy updrafts in the real space, they mainly correspond to subsiding shells (e.g. Glenn and Krueger (2014)).
- 4. Precipitating downdrafts. They correspond to air parcels on the bottom-left of the diagrams, with lower *m* relative to the domain-mean. They are among the most strongly descending air parcels (Figure 4b) but since they are scarce (Figure 4b),



Figure 4. Variables binned as a function of frozen moist static energy m and of altitude, for the ctrl simulation: (a) number of samples, (b) vertical velocity anomaly, (c) vertical mass flux (vertical velocity multiplied by the proportion of samples and density), (d) relative humidity, (e) cloud water content mixing ratio (liquid and ice), (f) precipitating water mixing ratio (rain, graupel and snow), (g) evaporation and condensation tendency dq/dt (positive in case of evaporation, negative in case of condensation), (h) dq/dt multiplied by the number of samples. The solid black line show the domain-mean frozen moist static energy, while the dashed black line shows the frozen moist static energy at saturation.



Figure 5. (b,e) As for Figure 4 but for (a) δD_v anomaly, (d) $(\phi - 1) \cdot 1000$, where $\phi = R_{ev}/R_v$; it is expressed in %. (a,d) As for (b,e) but for HighPrec. (c,f) As for (b,e) but for LowPrec.

contribute little to the total descending mass flux (Figure 4c). They are very dry, with no cloud water, but with precipitating water (Figure 4d-f). We interpret these parcels as unsaturated, precipitating downdrafts. Strong evaporation of rain occur in these downdrafts (Figure 4g), but because they cover only a small fraction of the domain, they contribute little to the evaporation in the domain-mean (Figure 4h).

The isotopic composition of water vapor is most enriched in the least diluted updrafts, 329 and most depleted in the precipitating downdrafts (Figure 5b). To assess the effect of 330 phase changes, we plot $\phi = R_{ev}/R_v$, where R_{ev} is the ratio of the water vapor tendency 331 associated with phase changes (evaporation in downdrafts and in the environment, or 332 condensation in cloudy updrafts) and R_v is the isotopic ratio of the water vapor in the 333 same *m*-altitude bin. In cloudy updrafts, $\phi - 1$ is about 100% in the lower troposphere 334 and increases with height (Figure 5e). This roughly corresponds to equilibrium fraction-335 ation during condensation. In cloudy downdrafts, $\phi - 1$ is also about 100%. This means 336 that cloud droplets evaporate totally without fractionation. In contrast, in precipitat-337 ing downdrafts, $\phi - 1$ is much lower. It is around 30% below 1 km. The fact that $\phi - 1$ 338 1 is positive is consistent with the fact that rain evaporation in the SCL acts to slightly 339 enrich the water vapor (Risi et al., 2020). In contrast, between 2 and 3 km, ϕ -1 is around 340 -100%: at these levels, rain evaporation acts to deplete the water vapor, consistent with 341 Worden et al. (2007). 342

These diagrams look qualitatively similar for the other simulations. One notice-343 able difference is that in HighPrec, the δD_v contrast between the environment and the 344 cloudy regions is larger (Figure 5a). This may be associated with the more depleted evap-345 oration of the rain in precipitating downdrafts and of cloud droplets in cloudy downdrafts 346 (Figure 5d). Conversely in LowPrec, the δD_v contrast between the environment and the 347 cloudy regions is larger (Figure 5c). To quantitatively compare the different simulations, 348 now we plot vertical profiles of variables in average over cloudy regions and over the en-349 vironment. 350

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2.6 Vertical profiles for cloudy regions and for the environment

Here we chose to define cloudy regions as all parcels with a cloud (liquid or ice) wa-352 ter content greater than 10^{-6} g/kg (e.g. Thayer-Calder and Randall (2015)). In this loose 353 definition, "cloudy regions" correspond to both cloudy updrafts and downdrafts, while 354 the "environment" includes both the environment and precipitating downdrafts. Includ-355 ing the cloudy downdrafts into the cloudy regions is justified by the fact that a signif-356 icant portion of the water condensed in cloudy updrafts subsequently evaporate in these 357 cloudy downdrafts, without directly affecting the environment. Our results below are not crucially sensitive to the definition of the cloudy regions and of the environment, pro-359 vided that the definition of cloudy regions is not too restrictive (Text S1). 360

Cloudy regions cover only a few percent of the domain (Figure 6a). The fraction of water condensed in cloudy regions that evaporates into the environment, estimated as $f_{ev} = -(dq/dt)_{env}/(dq/dt)_{cloud}$, where $(dq/dt)_{env}$ and $(dq/dt)_{cloud}$ are the humidity tendencies associated with phase changes in average in the environment and in the cloudy region respectively, varies between 30% and 90%, depending on altitude (Figure 6b). It is smaller in HighPrec and than in ctrl, because the environment is moister.

Figure 6c plots $\phi = R_{ev}/R_e$, where $R_{ev} = (dq_{HDO}/dt)_{env}/(dq/dt)_{env}, (dq_{HDO}/dt)_{env}$ is the HDO tendency associated with phase changes in the environment and R_e is the isotopic ratio in the environment. In all simulations except in HighPrec near 4.5 km, $\phi >$ 1: the evaporation has an enriching effect on the environment. The overall enriching effect of evaporation contradicts hypothesis 2. Yet in all cases, $\phi < \alpha_{eq}$: the evaporation is not as enriching as if there was total evaporation of condensate. The ϕ is smaller in



Figure 6. (a) fraction of the domain covered by cloudy regions. (b) Fraction of the water condensed in cloudy regions that evaporates into the environment, f_{ev} . (c) $(\phi - 1) \cdot 1000$ (solid) and $(\alpha_{eq} - 1) \cdot 1000$ (dashed), where $\phi = R_{ev}/R_e$ and α_{eq} is the equilibrium fractionation coefficient. Both are expressed in ∞ . The black, red and green lines are for ctr, HighPrec and LowPrec respectively.

HighPrec and larger in LowPrec than in ctrl: rain evaporation has a weaker enriching effect in HighPrec and and a stronger enriching effect in LowPrec. This supports hypothesis 3. In HighPrec near 4.5 km, near the melting level, there is even a small layer where $\phi < 1$: at this level, the rain evaporation has a depleting effect on the water vapor.

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2.7 What controls the isotopic composition of rain evaporation?

³⁷⁸ Why is ϕ smaller in HighPrec and higher in LowPrec than in ctrl? It could be be-³⁷⁹ cause rain-vapor exchanges in a moister environment leads the evaporation to have a more ³⁸⁰ depleting effect (Lawrence et al., 2004; Risi et al., 2008), or because rain evaporation is ³⁸¹ more depleted when the evaporated fraction is small (Risi et al., 2008; Tremoy et al., 2014), ³⁸² or because the rain itself is more depleted. We aim here at quantifying these different ³⁸³ effects.

Figure 7a plots the vertical profiles of rain δD (solid). Below the melting level, the 384 rain is very close to isotopic equilibrium with the vapor (dashed). Above the melting level, 385 the rain is more enriched than if in equilibrium due to rain lofting. Near the melting level 386 for simulation HighPrec, the rain is anomalously depleted. This is due to snow melt. Since 387 the snow forms higher in altitude, it is more depleted than the rain. It thus imprints its 388 depleted signature on the rain when melting. In HighPrec, the moist middle troposphere 389 prevents most of the snow from sublimating: 24% of the precipitation is made of snow 390 at the melting level. The rain is thus strongly depleted by snow melt. In contrast, in ctrl 391 and LowPrec, the drier middle troposphere favors snow sublimation: only 8% and 3%392 of the precipitation is made of snow at the melting level respectively. 393

The quick equilibration between the rain and vapor motivates us to use a simple equation in which some mass q_{l0} of rain, with isotopic ratio R_{l0} , partially evaporates and isotopically equilibrates with some mass q_{e0} of environment vapor, with isotopic ratio R_{e0} . As explained in text S2, if $q_{l0} \gg q_{e0}$, we get:

$$\phi = \frac{\lambda}{1 + (1 - f_{ev}) \cdot (\alpha_{eq} - 1)} \tag{2}$$

where $\phi = R_{ev}/R_e$, $\lambda = R_{l0}/R_{e0}$, R_{ev} is the isotopic ratio of the rain evaporation flux, α_{eq} is the equilibrium fractionation coefficient and f_{ev} is the fraction of the rain that evaporates. Equation 2 tells us that the rain evaporation is more depleted as the rain is more depleted relative to the vapor (quantified by λ) and as the evaporated fraction f_{ev} is smaller. This simple equation (Figure 7b, red) is able to approximate the simulated values of ϕ (black) for the ctrl simulation and is able to capture the smaller and larger values of ϕ for HighPrec and LowPrec respectively (Figure 7c-d).

We find that below the melting level, ϕ is smaller in HighPrec than in ctrl mainly because f_{ev} is smaller (Figure 7c, green). Near the melting level, ϕ is smaller in High-Prec than in ctrl both because f_{ev} is smaller and because λ is smaller, i.e. the rain is more depleted due to snow melt (Figure 7c, purple). In LowPrec, the effect of f_{ev} dominates at most levels (Figure 7d).

2.8 Summary

To summarize, the previous sections suggest that rain evaporation in the lower tro-411 posphere is a key ingredient of the vapor amount effect. The isotopic composition of the 412 rain evaporation flux mainly depends on the evaporated fraction of the rain, consistent 413 with Risi et al. (2008); Tremoy et al. (2014). Near the melting level in regimes of large-414 scale ascent, it is also impacted by snow melt. We hypothesize that the isotopic effect 415 of rain evaporation propagates downward down to the SCL. To test this hypothesis and 416 to understand the underlying mechanisms, in the next section we develop a simple two-417 column model. 418

A simple two-column model to quantify the relative contributions of different processes

The previous section and previous studies provide a guide for developing our sim-421 ple interpretative framework. First, the model needs to represent the effect of rain evap-422 oration, highlighted as a key process in the previous section. Second, alternative hypothe-423 ses for the "vapor amount effect" involve mixing between the subsident environment and 424 detrained water (Bailey et al., 2017) (hypothesis 4). This process also needs to be rep-425 resented in our model. Third, the steepness of the $q - \delta D_v$ relationship must be a key 426 ingredient, since it drives δD_v in the SCL and thus δD_v everywhere. Finally, the pre-427 vious section has relied on the distinction between the environment and cloudy regions. 428 Keeping this distinction, we develop a two-column model. 429

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3.1 Model equations and numerical application to LES outputs

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3.1.1 Balance equations

This model is inspired by the two-column model used to predict tropospheric relative humidity in Romps (2014) and δD_v profiles in Duan et al. (2018). The first column represents the cloudy regions, including cloudy updrafts and downdrafts, as a bulk entraining plume. The second column represents the subsiding environment and precipitating downdrafts (Figure 8).

The mass balance for the air in the cloudy regions writes:

$$\frac{dM}{dz} = M \cdot (\epsilon - \delta) \tag{3}$$

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Figure 7. (a) δD profile for rain water (solid) and snow (dotted) falling in the environment. The liquid that would be in equilibrium with the vapor in the environment is shown in dashed. (b) Profile of $\phi = R_{ev}/R_e$ simulated by the ctrl simulation (black, same as in Figure 6c black) and predicted by equation 2 (red). (c) Difference of ϕ between HighPrec and ctrl simulated by the LES (black), predicted by the equation 2 (red), predicted by equation 2 if only f_{ev} varies (green) and if only λ varies (purple). (d) Same as (c) but for the difference between LowPrec and ctrl.

where M is the bulk mass flux in the cloudy regions (positive upward), ϵ and δ are the fractional entrainment and detrainment rates.

We assume that the q in the cloudy regions is at saturation, and call it q_s . The water balance in the cloudy regions writes:

$$\frac{d(Mq_s)}{dz} = \epsilon \cdot M \cdot q_e - \delta \cdot M \cdot q_s - c \tag{4}$$

where c is the condensation rate and q_e is the specific humidity in the environment. The terms on the right hand side represent the water input by entrainment of environment air, the water loss by detrainment of cloudy air, and the water loss by condensation respectively. We assume that all the condensed water is immediately lost by the cloudy regions to the environment, and evaporation of this lost water can occur in the sub-saturated environment only, as in Romps (2014).

We assume that mass is conserved within the domain, so that the flux in the environment is -M. The large-scale ascent, when present, is taken into account through a humidity tendency, consistent with the LES set-up. We assume that the large-scale humidity tendency applies to the environment only, which is a first-order approximation justified by the small fraction of the domain that is covered by cloudy updrafts (less than 10%). The water balance in the environment writes:

$$\frac{d\left(-Mq_e\right)}{dz} = -\epsilon \cdot M \cdot q_e + \delta \cdot M \cdot q_s + f_{ev} \cdot c - \eta \cdot M \cdot \frac{\partial q_e}{\partial z}$$
(5)

where f_{ev} is the fraction of the cloud or precipitating water that evaporates in the environment, $\eta = M_{LS}/M$ and M_{LS} is the domain-mean large-scale mass flux. The terms on the right hand side represents the water loss by entrainment into cloudy regions, water input by the detrainment of cloudy air, partial evaporation of condensed water and water input by large-scale vertical advection.

Regarding water isotopes, we assume that the cloud water removed by condensation is in isotopic equilibrium with the cloudy region water vapor. The isotopic balance
in the cloudy regions thus writes:

$$\frac{d\left(Mq_s\cdot R_s\right)}{dz} = \epsilon \cdot M \cdot q_e \cdot R_e - \delta \cdot M \cdot q_s \cdot R_s - c \cdot \alpha_{eq} \cdot R_s \tag{6}$$

where α_{eq} is the equilibrium fractionation coefficient, R_s is the isotopic ratio in the cloudy regions and R_e is the isotopic ratio in the environment.

The isotopic balance in the environment writes:

$$\frac{d\left(-Mq_e\cdot R_e\right)}{dz} = -\epsilon\cdot M\cdot q_e\cdot R_e + \delta\cdot M\cdot q_s\cdot R_s + f_{ev}\cdot c\cdot\phi\cdot R_e - \eta\cdot M\cdot\frac{\partial\left(q_eR_e\right)}{\partial z} \quad (7)$$

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where $\phi = R_{ev}/R_e$ and R_{ev} is the ratio of the precipitation evaporation flux.

466 3.1.2 Other simplifying assumptions and differential equations

To simplify the equations, as in Romps (2014) we assume that q_s is an exponential function of altitude:

$$q_s = q_s(z_0) \cdot e^{-\gamma \cdot (z - z_0)} \tag{8}$$



Figure 8. Schematic view of the simple two-column model, and definition of the main variables.

where γ is a lapse rate in m^{-1} calculated as $d \ln(q_s)/dz$.

For isotopes, we assume that the R_s is a power function of q_s , consistent with a Rayleigh distillation:

$$R_s = R_s(z_0) \left(q_s / q_{s0} \right)^{a_s - 1}$$

472 Coefficient α_s represents the steepness of the $q - \delta D_v$ gradient in cloudy regions 473 and remains to be estimated. As in Duan et al. (2018), R_s is thus an exponential func-474 tion of altitude:

$$R_s = R_s(z_0) \cdot e^{-(\alpha_s - 1) \cdot \gamma \cdot (z - z_0)} \tag{9}$$

475 We set:

 $q_e = h \cdot q_s$ $R_e = H \cdot R_s$

476 Combining equation 5 with equations 3 and 8, we get the following differential equa-477 tion for h:

$$\frac{\partial h}{\partial z} = h \cdot \gamma - \frac{\delta}{1 - \eta} (1 - h) - \frac{f_{ev} \cdot \mu \cdot \gamma}{1 - \eta} \tag{10}$$

where $\mu = c/(M \cdot q_s \cdot \gamma)$ represents the ratio of actual condensation (c) relative to the condensation if the ascent was adiabatic $(M \cdot q_s \cdot \gamma)$. Similarly, combining equations 7 with equations 5 and 9, we get the following differential equation for H:

$$\frac{\partial H}{\partial z} = H \cdot \gamma \cdot (\alpha_s - 1) - \frac{\delta}{h \cdot (1 - \eta)} \cdot (1 - H) - \frac{f_{ev} \cdot \mu \cdot \gamma}{h \cdot (1 - \eta)} \cdot H \cdot (\phi - 1)$$
(11)

⁴⁸¹ Note that these equations are only valid as long as $\eta < 1$, which will be the case ⁴⁸² in all our simulations (section 3.1.4). We now have two equations with four unknowns: ⁴⁸³ h, H, μ and α_s . The condensation efficiency μ can be deduced from equations 4:

$$\mu = 1 - \frac{\epsilon}{\gamma} \cdot (1 - h) \tag{12}$$

⁴⁸⁴ This equation, similar to one in Romps (2014), reflects the fact that condensation ⁴⁸⁵ efficiency decreases when entrainment ϵ increases and when the entrained air is drier. If ⁴⁸⁶ $\epsilon = 0$ or h = 1, then $\mu = 1$.

Similarly, the $q - \delta D_v$ steepness α_s in cloudy air can be deduced from equation 6:

$$\alpha_s - 1 = \mu \cdot (\alpha_{eq} - 1) + \frac{\epsilon}{\gamma} \cdot h \cdot (1 - H)$$
(13)

This equation tells us that two effects control the steepness of the $q - \delta D_v$ gradient. First, there is a "dilution effect": if dry air is entrained, then the condensation efficiency μ decreases. This reduces α_s compared to α_{eq} , i.e. compared to what we would expect from Rayleigh distillation. Second, there is an "isotopic contrast effect": if depleted water vapor is entrained (H < 1), then α_s becomes steeper. This is how a depleting effect of rain evaporation in the environment can translate into a larger steepness in both regions, and eventually more depleted SCL.

496 3.1.3 Numerical solutions

To get analytical solutions for h and H, Romps (2014) and Duan et al. (2018) as-sume that $h \cdot \frac{\partial q_s}{\partial z} \gg q_s \cdot \frac{\partial h}{\partial z}$ and that $H \cdot \frac{\partial R_s}{\partial z} \gg R_s \cdot \frac{\partial H}{\partial z}$. This allows them to calculate h497 498 and H as the solutions of a simple linear equation and of a second order polynomial re-499 spectively. However, there are two issues with these solutions. First, although these so-500 lutions behave reasonably for h (Romps, 2014), they become very noisy, unstable or un-501 realistic for H when values for ϵ , δ and f_{ev} that are diagnosed from LES outputs. This 502 is because a powerful positive feedback exists between α_s and H: as H decreases, more 503 depleted vapor is entrained in updrafts which increases the steepness α_s ; in turn, the stronger 504 steepness α_s makes the subsidence more efficient at depleting the environment, further 505 decreasing H. Duan et al. (2018) circumvented this problem by assuming ϵ and δ that 506 are constant with altitude and equal to each other, but it is at the cost of artificially re-507 ducing freedom for the solutions. Second, our hypothesis is that rain evaporation near 508 the melting level affects the isotopic profiles down to the SCL. We thus want each al-509 titude to feel the memory of processes at higher altitudes. The term with $\frac{\partial H}{\partial z}$ is thus a 510 key ingredient in our framework. 511

Therefore, we choose to numerically solve the differential equations 10 and 11. We start from an altitude of 5 km with h = 0.8 and H - 1 = -10%. We do not start above 5 km because entrainment is more difficult to diagnose above the melting level (section 3.1.4). We integrate equations 10 and 11 down to the SCL top around 500 m. The resulting h profile is a function of the profiles of 5 input parameters: γ , ϵ , δ , f_{ev} and η 517 . The *H* profile is a function of 7 input parameters: γ , ϵ , δ , f_{ev} , η , α_{eq} and ϕ . These in-518 put parameters are all diagnosed from the LES simulations as detailed below. In each 519 LES level, the input parameters are assumed constant and equations 10 and 11 are in-520 tegrated within each layer over 50 sub-layers.

3.1.4 Diagnosed input parameters

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Parameters f_{ev} , α_{eq} and ϕ were already plotted in Figure 6 and discussed in sec-522 tion 2.6. Parameter γ is calculated from domain-mean profiles. It is steeper in ctrl than 523 in ω -60 because of the steeper temperature gradient resulting from the drier air (Fig-524 ure 9a). Parameter $\eta = M_{LS}/M$ is calculated from the net upward mass flux in cloudy 525 regions M (Figure 9b), which is calculated as the average vertical velocity in cloudy re-526 gions multiplied by the area fraction of the cloudy region. Entrainment ϵ is diagnosed 527 by using the conservation of the frozen moist static energy m (e.g. Hohenegger and Brether-528 ton (2011); Del Genio and Wu (2010)): 529

$$\frac{\partial m_s}{dz} = \epsilon \cdot (m_e - m_s)$$

where m_s and m_e are the frozen moist static energy in the cloudy region and the environment respectively. The application of this equation is limited to the lower troposphere. Above the melting level, we would need to account for the precipitation of ice (Pauluis & Mrowiec, 2013) and for the lofting of rain. Therefore, we arbitrarily set a minimum of $\epsilon = 0.5 \, km^{-1}$ above the melting level. Entrainment is maximal in the sub-cloud layer, and decreases exponentially with height (Figure 9c), consistent with previous studies (Del Genio & Wu, 2010; De Rooy et al., 2013).

Finally, detrainment δ is deduced from ϵ and M using equation 3. Detrainment shows the typical trimodal distribution (Johnson et al., 1999) (Figure 9d), with a first maximum just above the SCL top corresponding to the detrainment of shallow convection, a second maximum near the melting level corresponding to the detrainment of congestus convection, and a third maximum in the upper troposphere corresponding to the deep convection (not shown in Figure 9d).

3.1.5 Closure in the sub-cloud layer

To calculate the full δD profiles, we need as initial condition the isotopic ratio in 544 the SCL. With this aim, we use a simple version of the SCL model of Risi et al. (2020). 545 We assume that water enters the SCL through surface evaporation and through down-546 drafts at the SCL top, and exits the SCL through updrafts at the SCL top. We neglect 547 large-scale forcing and rain evaporation, since they have a small impact in the SCL (Risi 548 et al., 2020). The air flux of updrafts equals that of downdrafts. We define $r_u = q_u/q_1$ 549 and $r_d = q_d/q_1$, where q_1 is the mixing ratio in the SCL and q_u and q_d are the mixing 550 ratios in updrafts and downdrafts at the SCL top. We assume that the water vapor is 551 more enriched as the air is moister, following a logarithmic function: $R_u = R_1 \cdot r_u^{\alpha_u - 1}$ 552 and $R_d = R_1 \cdot r_d^{\alpha_d - 1}$ where R_u and R_d are isotopic ratios in updrafts and downdrafts, 553 and α_u and α_d are the $q - \delta D_v$ steepness coefficients for updrafts and downdrafts. Wa-554 ter and isotopic budgets yield: 555

$$R_{1} = \frac{R_{oce}/\alpha_{eq}(SST)}{h_{1} + \alpha_{K} \cdot (1 - h_{1}) \cdot \frac{r_{u}^{\alpha_{u}} - r_{d}^{\alpha_{d}}}{r_{u} - r_{d}}}$$
(14)

where R_{oce} is the isotopic ratio at the ocean surface, $\alpha_{eq}(SST)$ is the equilibrium fractionation coefficient at the sea surface temperature, α_K is kinetic fractionation coefficient (Merlivat & Jouzel, 1979) and h_1 is the relative humidity normalized at the SST



Figure 9. Input parameters for the simple model, for ctrl (black), HighPrec (blue) and Low-Prec (orange). (a) saturation specific humidity lapse rate γ ; (b) ratio of large-scale vertical mass flux over the cloudy mass flux; (c) entrainment rate; (d) detrainment rate.

and accounting for ocean salinity: $h_1 = q_1/q_{sat}^{surf}(SST)$, $q_{sat}^{surf}(SST) = 0.98 \cdot q_{sat}(SST)$ and q_{sat} is the humidity saturation as a function of temperature at the sea level pressure. We assume $\delta D_{oce} = 0\%$ and h_1 is diagnosed from the LES.

For r_u and r_d , we use values for the ctrl simulation, because small changes in r_u and r_d across simulations have only a marginal impact on R_1 (Risi et al., 2020). Following Risi et al. (2020), we set $r_u - 1 = 1.44\%$ and $r_d - 1 = -0.38\%$. For α_u and α_d , Risi et al. (2020) had shown that they scale with α_z values just above the SCL top, but with larger values especially for simulations with large-scale ascent. We use an empiricallyfitting function: $\alpha_u = \alpha_d = 1 + 100 \cdot (\widetilde{\alpha_z} - 1)^3$, where $\widetilde{\alpha_z} = 1 + \frac{ln(R(z_{SCT})/R(z_{SCT} + 1\,km))}{ln(q(z_{SCT})/d(z_{SCT} + 1\,km))}$ and z_{SCT} is the altitude of SCL top.

Finally, since the updraft region covers only a very small fraction of the domain, we assume that $R_e(z_{SCT}) \simeq R_1$.

The procedure to calculate the full δD_v profiles is as follows:

- ⁵⁷² 1. vertical profiles for h, H and α_s are calculated through a downward integration ⁵⁷³ of equations 10-13 following section 3.1.3.
 - 2. The vertical profile for a normalized version of R_s , $R_{s,norm}$ that satisfies $R_{s,norm}(z_{SCT}) = 1$, is calculated based on the α_s profile through an upward integration.
 - 3. The vertical profile for a normalized version of R_e , $R_{e,norm}$, is calculated as $R_{e,norm} = R_{s,norm} \cdot H$.
 - 4. From the $R_{e,norm}$ profile, $\widetilde{\alpha_z}$ is estimated.
 - 5. From h_1 and $\widetilde{\alpha_z}$, R_1 is estimated.
- 6. The full R_e profile can finally be calculated so that $R_e(z_{SCT}) \simeq R_1$: $R_e = R_{e,norm} \cdot R_1/H(z_{SCT})$.
 - 3.1.6 Evaluation of the two-column model

The two-column model successfully captures the order of magnitude and the shape of the vertical profile of h for the ctrl simulation (Figure 10a), as well as the moister troposphere in HighPrec and the drier troposphere in LowPrec (Figure 10b-c).

It successfully captures the vertical profile of δD_v (Figure 10b) and the more depleted troposphere in HighPrec but underestimate the δD_v difference by about half (Figure 10e). It also captures the more enriched troposphere in LowPrec but again underestimate the δD_v difference especially in the middle troposphere (Figure 10f). Similarly, it approximately captures the steepness α_z and the sign of the α_z differences across simulations, but underestimates the α_z differences (Figure 10g-i).

These mismatches are caused by mismatches in the estimate of the relative enrich-592 ment of the environment relative to the cloudy region H. Although it is reasonably well 593 predicted for the ctrl simulation (Figure 10j), the model fails to simulate the smaller H594 for HighPrec in the middle troposphere and the larger H for LowPrec almost everywhere. 595 The two-column model overestimates the impact of η and predicts a behavior for H that 596 is too similar to that of h. We could not find the exact reason for this shortcoming, but 597 we have to acknowledge that the two-column model hides many horizontal heterogeneities. 598 We will have to keep this shortcoming in mind when interpreting the results. 599

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3.2 Decomposition of relative humidity and δD_v variations

To estimate the impact of the different input parameters on the h and δD_v profiles, we modify them one by one from the ctrl simulation to the HighPrec and from the ctrl simulation to LowPrec simulations.



Figure 10. (a) Relative humidity h simulated by the LES (black) and predicted by the twocolumn model (red) for the ctrl simulation. (b) Same as (a) but for the difference between High-Prec and ctrl. (c) Same as (b) but for the difference between LowPrec and ctrl. (d-f) Same as (a-c) but for the water vapor δD . (g-i) Same as (a-c) but for the steepness α_z . (j-l) Same as (a-c) but for the relative enrichment of the environment relative to the updrafts H.



Figure 11. (a) Relative humidity difference between HighPrec and ctrl predicted by the twocolumn model (black) and its contributions from variations of input parameters one by one: η (pink), γ (green), ϵ and δ (red) and f_{ev} (blue). (b) Same as (a) but for the difference between LowPrec and ctrl.

3.2.1 Decomposition of relative humidity

⁶⁰⁵ The moister troposphere in HighPrec is mainly due to the larger η , i.e. the direct ⁶⁰⁶ moistening effect of large-scale ascent (Figure 11a). The thermodynamic structure, en-⁶⁰⁷ trainment, detrainment and rain evaporation have a much smaller effect. Similarly, The ⁶⁰⁸ drier troposphere in LowPrec is mainly due to the more negative η , i.e. the direct dry-⁶⁰⁹ ing effect of large-scale descent (Figure 11b).

Note that the direct effect of η on h in the environment may be overestimated in our simulations by prescribing a large-scale vertical velocity profile that is horizontally constant (Bao et al., 2017).

3.2.2 Dilution effect on δD_v

⁶¹⁴ A first effect impacting δD_v profiles is the dilution by entrainment (section 3.1.2). ⁶¹⁵ In the absence of entrainment ($\epsilon = 0$), the steepness in the updraft column would be ⁶¹⁶ $\alpha_s = \alpha_{eq}$ (Figure 12a, black). Because dry air is entrained, the condensation rate is re-



Figure 12. (a) Fractionation coefficient α_{eq} (black), corresponding to the steepness in the cloudy column α_s if h = 1 and H = 1; steepness α_s predicted if h < 1 and H = 1 $(\alpha_s = 1 + \mu \cdot (\alpha_{eq} - 1))$ (green); steepness α_s from the full equation 13 (purple). (b) Difference in δD_v from ctrl to HighPrec predicted by the two-column model (black) and predicted if accounting only for the dilution effect (green). (c) Same as (b) but for LowPrec.

duced by the factor μ following equation 12. According to equation 13, this reduces the steepness (Figure 12a, green). This effect of entrainment can be understood as a mixing process: as the air rises and condensation proceeds, the remaining air is mixed with dry air from entrainment and with droplets that evaporate. Consistent with the concavedown shape of the mixing lines, this leads to a reduction of the $q - \delta D_v$ steepness (Figure 1, orange and cyan).

As a consequence of this "dilution effect", tropospheric δD_v is less depleted than 623 predicted by Rayleigh distillation. Since the troposphere is moister in HighPrec, entrained 624 air leads to less evaporation of cloud droplets than in ctrl. This weaker "dilution effect" 625 contributes to more depleted δD_v in HighPrec (Figure 12b, green). Reciprocally, since 626 the troposphere is drier in LowPrec, the sronger "dilution effect" contributes to the more 627 enriched δD_v in LowPrec (Figure 12c, green). Quantitatively, the contribution of this 628 dilution effect on the SCL δD_v difference is 29% for HighPrec and 47% for LowPrec (ta-629 ble 2). The contribution increases with altitude. 630

⁶³¹ Note that the two-column model likely overestimates this contribution, because of ⁶³² the shortcoming mentioned in section 3.1.6. The fact that only one third of the δD_v dif-⁶³³ ference remains when post-condensation effects are turned off (section 2.4) confirms that ⁶³⁴ these contributions are overestimated.

3.2.3 Decomposition of δD_v

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In HighPrec, the more depleted troposphere is driven primarily by the effect of the smaller ϕ , i.e. the more depleted rain evaporation (Figure 13a, cyan). It explains 147% of the δD_v difference in the SCL (Table 2). The smaller rain evaporated fraction (smaller f_{ev}) is the second main contributor (Figure 13a, blue, 43% in the SCL). This positive contribution is explained by the fact that evaporation has an overall enriching effect. The third main contributor is the larger η (i.e. large-scale ascent), contributing to 26% of the

Table 1. Difference of δD_v in the SCL between HighPrec and ctrl and between LowPrec and ctrl simulated by the LES and predicted by the two-column model, and the contribution of the dilution effect.

Difference in SCL δD_v from ctrl	HighPrec	LowPrec
Total simulated by the LES $(\%)$	-40	10
Total predicted by the two-column model $(\%)$	-30	11
Dilution effect (‰, %)	-9 (29%)	5 (47%)

 δD_v difference. This contribution corresponds mainly to the "dilution effect" explained in section 3.2.2. The sum of these contributions exceeds 100%, because there are some dampening effects, especially h_1 : the moister surface relative humidity reduces the kinetic fractionation during surface evaporation.

⁶⁴⁶ In LowPrec, η becomes the main contribution to the δD_v difference in the SCL (126%), ⁶⁴⁷ through the dilution effect (Figure 13b, pink, Table 2). The effect of the larger ϕ , i.e. the ⁶⁴⁸ more enriched rain evaporation, contributes to 36% to the δD_v difference in the SCL.

This decomposition can be reconciled with the result that about one third of the δD_v difference from ctrl to HighPrec remains when the fractionation during condensate evaporation is de-activated. This remaining difference is associated with (1) the dilution effect, and (2) the portion of the ϕ contribution that is due to the more depleted rain due to more snow melt. The fact that the sum of this two contributions exceeds one third suggests that the underestimate of δD_v variations by the simple model is due to underestimating the effect of rain evaporation.

We note that the relative contributions of the different processes are very homogeneous in the vertical. For example, in the SCL, half of the contribution of ϕ comes from ϕ above 3 km. This shows the strong "memory" of water vapor δD , which integrates processes downwards in the environment column, and then upward in the cloudy column.

660 4 Conclusion

661

4.1 Summary

The amount effect, i.e. the observed decrease in precipitation δD as precipitation rate increases, is the most salient feature in monthly-mean isotopic observations over tropical oceans (Dansgaard, 1964). We confirm here that it is intimely related to the "vapor amount effect", i.e. the observed decrease in water vapor δD as precipitation rate increases (Worden et al., 2007). This study gives a comprehensive and quantitative understanding of the processes underlying the vapor amount effect, at least in our LES simulations. This understanding is illustrated in Figure 14:

- When the troposphere is moister (in terms of relative humidity), less snow sublimates and thus more snow is available for melting. Snow melt results in rain that
 is more depleted relative to a liquid in equilibrium with the vapor, which leads to
 more depleted rain evaporation flux. When the troposphere is moister, the rain
 evaporated fraction is also smaller, making the rain evaporation flux even more
 depleted.
- ⁶⁷⁵ 2. The more depleted evaporation depletes the environment more efficiently relative ⁶⁷⁶ to clouds. When this more depleted environment is entrained into the clouds, it



Figure 13. (a) δD_v difference between HighPrec and ctrl predicted by the two-column model (black) and its contributions from variations of input parameters one by one: η (pink), γ and α_{eq} (green), ϵ and δ (red), f_{ev} (blue), ϕ (cyan) and h_1 (orange). (b) Same as (a) but for the difference between LowPrec and ctrl.

Table 2. Difference of δD_v in the SCL between HighPrec and ctrl and between LowPrec and ctrl simulated by the LES and predicted by the two-column model, and the contribution of different effects. The sum of all the different effects, except the line "Including ϕ above 3 km", is 100% of the predicted δD_v difference. The line "Including ϕ above 3 km" is a part of "Effect of ϕ "

SCL δD_v difference from ctrl	HighPrec		LowPrec
Total simulated by the LES $(\%)$	-40		10
Total predicted by the two-column model $(\%)$	-30		11
Effect of γ and α_{eq} (‰, %)	8 (-25%)		-3 (-30%)
Effect of ϵ and δ (‰, %)	6 (-19%)		2(14%)
Effect of η (‰, %)	-8 (26%)		14 (126%)
Effect of f_{ev} (‰, %)	-13 (43%)		-2 (-22%)
Effect of ϕ (‰, %)	-44 (147%)		4(36%)
Including ϕ above 3 km (‰, %)	-23 (76%)		2(23%)
Effect of h_1 (‰, %)	22 (-72%)		-3 (-24%)

677		makes the $q - \delta D_v$ vertical gradient steeper. In turn, the steeper $q - \delta D_v$ gradi-
678		ent makes the subsidence more efficient at depleting the environment, in a pos-
679		itive feedback that makes the $q - \delta D_v$ gradient even steeper. Overall, this mech-
680		anism allows to propagate the isotopic anomalies associated with rain evapora-
681		tion downwards.
682	3.	When the troposphere is moister, the dilution of cloudy air by entrainment is weaker.
683		Water vapor condenses more efficiently, which also contributes to the steeper $q-$
684		δD_v vertical gradient.
685	4.	The steeper $q - \delta D_v$ gradient in the lower troposphere makes updrafts and down-

drafts at the SCL top more efficient in depleting the SCL water vapor (Risi et al., 2020).

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5. Finally, since the more depleted SCL vapor serves as the initial condition for the full δD_v vertical profiles, the water vapor is more depleted at all altitudes in the troposphere.

Coming back to our initial hypotheses to explain the vapor amount effect, the dominant role of rain evaporation and rain-vapor diffusive exchanges confirms hypothesis 3 (Lawrence et al., 2004; Risi et al., 2008; Lee & Fung, 2008). The role of entrainment in diluting cloudy air and reducing their condensation efficiency is reminiscent of hypothesis 4.

We notice that the root of the vapor amount effect in the water vapor is higher rel-696 ative humidity, with a triple effect on reducing (1) the sublimation of snow aloft, (2) the 697 fraction of rain that evaporates, and (3) the dilution of cloudy air by entrainment. This 698 explains why the amount effect can be observed only when the precipitation increase is 699 associated with a change in the large-scale circulation (Bony et al., 2008; Moore et al., 2014; Bailey et al., 2017; Risi et al., 2020). While the tropospheric relative humidity is 701 very sensitive to the large-scale circulation, it is almost invariant with sea surface tem-702 perature (Romps, 2014). For example, if precipitation increases because sea surface tem-703 perature increases without any change in large-scale circulation, then the tropospheric 704



Figure 14. Schematic summarizing how a moister troposphere leads to more depleted vapor in the troposphere. The black and red boxes represent standard water processes and isotopic processes respectively.

humidity would remain almost constant (Romps, 2014), so the above-mentioned mechanism cannot take place and there is no amount effect.

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4.2 Discussion and perspectives

This study has investigated processes controlling isotopic profiles in idealized conditions. In particular, large-scale horizontal gradients in humidity and δD_v were neglected. In reality, these gradients are expected to dampen the humidity and δD variations as a function of large-scale vertical velocity (Risi et al., 2019).

To assess to what what extent our idealized simulations in radiative-convective equilibrium over the ocean are relevant for interpreting observations, it would be useful to compare our LES simulations with different large-scale velocities to in-situ and remotesensing observations. This raises the question of the spatial scales at which the amount effect can be observed and of the spatial representativeness of both observations and LES simulations. This will also be investigated in a future study.

This paper highlights the important role of snow melt and rain evaporation in de-718 pleting the water vapor in case of large-scale ascent. These processes are expected to be 719 even stronger in stratiform regions of meso-scale systems, where all the rain arises from 720 the widespread melting of snow near the melting level, and where the rain evaporation 721 is boosted by the meso-scale downdraft that dries the lower troposphere (Houze, 1977). 722 This may explain why observations show that stratiform regions are often more depleted 723 than convective regions in squall lines (Risi et al., 2010; Tremov et al., 2014), and why 724 the amount effect is stronger where the fraction of stratiform clouds is larger (Kurita, 725 2013; Aggarwal et al., 2016; Sengupta et al., 2020). To check this hypothesis, we plan 726

to analyze in a future study the dependence of water vapor isotopic profiles to large-scale

- circulation in LES with different convective organizations, such as squall lines (Robe &
- Emanuel, 2001; C. Muller, 2013) or tropical cyclones (M. Khairoutdinov & Emanuel, 2013;
- ⁷³⁰ C. J. Muller & Romps, 2018).

Finally, this study highlights the key role of both microphysical processes (evap-731 oration, snow melt) and macrophysical processes (entrainment) in the amount effect. While 732 entrainment is partly resolved by grid-scale motions, LES models rely strongly on mi-733 crophysical and subgrid-scale turbulence parameterizations in representing these processes. 734 What is the sensitivity of the amount effect to these parameterizations? These processes 735 are even more crudely parameterized in general circulation models (GCMs). How do GCMs 736 represent these processes? More generally, what would be the added value of adding iso-737 topic diagnostics when routinely comparing single-column versions of GCMs to LES sim-738 ulations? This is yet another question that we plan to address in the future. 739

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- All simulation outputs used in this article will be submitted to the PANGEA data repository.

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Supporting Information for "Rain evaporation, snow melt and entrainment at the heart of water vapor isotopic variations in the tropical troposphere, according to large-eddy simulations and a two-column model"

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Contents of this file

1. Text S1: Robustness of the results with respect to the definition for clouds and the environment

2. Text S2: Simple equation for rain evaporation

Introduction

This supporting information assess the robustness of the results with respect to the definition for clouds and the environment (Text S1) and details how the simple equation for rain evaporation is derived (Text S2).

Text S1: Robustness of the results with respect to the definition for clouds and the environment X - 2

In our simple two-column framework, we decide to separate cloudy regions from their environment based on a threshold on cloud water content (e.g. Thayer-Calder and Randall (2015)): we define parcels as "cloudy" when the cloud water content exceeds 10^{-6} g/kg. In the previous studies, alternative definitions have been based on vertical velocity (e.g. Hohenegger and Bretherton (2011)) and/or buoyancy (e.g. Siebesma and Cuijpers (1995)), or position in altitude-equilvalent potential temperature diagrams (Pauluis & Mrowiec, 2013). We thus test here the robustness of our results to different definitions, by defining "very cloudy regions" with cloud water content larger than 10^{-3} g/kg, "cloudy updrafts" with cloud water content larger than 10^{-6} g/kg and ascending vertical velocity, "saturated drafts" with relative humidity larger than 99%, "nearly-saturated drafts" with relative humidity larger than 95%, and (7) "moist static energy updrafts" including all parcels falling into bins of frozen moist static energy in which the vertical velocity is positive (Pauluis & Mrowiec, 2013).

"Cloudy updrafts" and "nearly-saturated regions" are the most and least restrictive definitions respectively (Figure S1a,f). In all definitions, the cloudy region fraction remains below 10% except in the free lower and middle troposphere. In stricter definitions, the cloudy regions are characterized by a larger vertical velocity (Figure S1b) and a larger cloud water content (not shown). The entrainment is not strongly sensitive to the definition in the free troposphere (Figure S1c).

The ratio of the isotopic ratio in the rain evaporation over that in the environment vapor ($\phi = R_{ev}/R_v$) is not very sensitive to the definition for the ctrl (Figure S1e), but its value near the melting level is quite sensitive (Figure S1g). In all definitions, we can see the negative anomaly near the melting level, but it is much more negative in the loosest

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definitions. This is because in stricter definitions, the non-fractionating evaporation of cloud water droplets takes place in the environment. Since droplet evaporation takes place in shells around convective updrafts, and does not directly affect the environment, we chose a loose definition for the "cloudy regions".

The ratio of the large-scale mass flux over the cloudy mass flux, η , for HighPrec is larger in loose definitions (Figure S1h). This is because the cloudy regions incorporates cloudy downdrafts that compensate for the upward mass flux in cloudy updrafts. This large η in the loose definition may contribute to the overestimate of the direct effect of large-scale forcing on δD by the two-column model, and ultimately to the underestimate of the "vapor amount effect".

Text S2: Simple equation for rain evaporation

The quick equilibration between the rain and vapor motivates us to use a simple equation in which some mass q_{l0} of rain, with isotopic ratio R_{l0} , partially evaporates and isotopically equilibrates with some mass q_{e0} of vapor (subscript *e* for environment), with isotopic ratio R_{e0} . After the evaporation and equilibration process, the masses of rain and vapor are noted q_l and q_v :

$$q_l = q_{l0} - q_{ev}$$
$$q_e = q_{e0} + q_{ev}$$

where q_{ev} is the mass of evaporated rain water. The corresponding isotopic budget writes:

$$R_l \cdot q_l = R_{l0} \cdot q_{l0} - R_{ev} \cdot q_{ev}$$

 $R_e \cdot q_e = R_{v0} \cdot q_{v0} + R_{ev} \cdot q_{ev}$ October 20, 2020, 8:35am where R_l , R_e and R_{ev} are isotopic ratios in the final rain, final vapor and evaporation flux. Isotopic equilibrium writes:

$$R_l = \alpha_{eq} \cdot R_e$$

where α_{eq} is the equilibrium fractionation coefficient.

We define:

$$f_{ev} = \frac{q_{ev}}{q_{l0}}$$
$$g = \frac{q_{l0}}{q_{e0}}$$
$$\lambda = \frac{R_{l0}}{R_{e0}}$$
$$\phi = \frac{R_{ev}}{R_{e0}}$$

Re-arranging these equations, we get:

$$\phi = \frac{\lambda \cdot (1 + f \cdot g) - (1 - f) \cdot \alpha}{f \cdot (g \cdot (1 - f) \cdot \alpha + 1 + f \cdot g)}$$

If the mass of rain is much greater than than of vapor, i.e. $g \gg 1$, the equation becomes:

$$\phi = \frac{\lambda}{1 + (1 - f_{ev}) \cdot (\alpha_{eq} - 1)}$$

Therefore, ϕ scales with λ . In addition, ϕ increases with f_{ev} from $\phi = \lambda/\alpha_{eq}$ for $f_{ev} = 0$ (first order approximation) to $\phi = \lambda$ pour $f_{ev} = 1$ (total evaporation).

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(a)

6

2

(d)

6

4

2

0

(f)

6

2

4 8 cloud fraction (%) HighPree

altitude (km)

altitude (km)

altitude (km)



altitude (km)

 $1\dot{2}$ 16 20 6

4 2

-60

 $\begin{array}{ccc} 0 & 60 \\ \phi & (\%) \\ \text{HighPree} \end{array}$

120 180

Figure S1. (a-e): Vertical profiles for the ctrl simulation. (a) fraction of the domainarea covered by cloudy regions. (b) Vertical velocity w in average over the cloudy regions. (c) Entrainement rate ϵ diagnosed from the frozen moist static energy budget as explained in the article. (d) $\phi = R_{ev}/R_e$, expressed in %; (e) rain evaporated fraction f_{ev} . The different colors show the different definitions for the cloudy regions: "cloudy regions" (purple), "very cloudy regions" (blue), "cloudy updrafts" (green), "saturated drafts" (yellow), "nearly saturated drafts" (red), and "moist static energy updrafts" (black). (f) Same as (a) but for HighPrec. (g) Same as (d) but for HighPrec. (h) Ratio of the large-scale mass flux over the cloudy mass flux, η , for HighPrec. October 20, 2020, 8:35am

altitude (km)

6

0

 $\begin{array}{ccc} & 60 \\ \eta & (\%) \\ \text{HighPrec} \end{array}$

90

30