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Combined measurements of $^{17}\text{O}_{\text{excess}}$ and d-excess in African monsoon precipitation: Implications for evaluating convective parameterizations

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Water stable isotopes ($\delta^{18}\text{O}$, δD) are useful tools to depict and to understand the atmospheric water cycle. In tropical regions, they record the variations of convective activity and their implementation in convection schemes brings constraints on our understanding and parameterization of this phenomena. Here, we present for the first time measurements of a new isotopic marker of the hydrological cycle ($^{17}\text{O}_{\text{excess}}$ resulting from the combination of $\delta^{17}\text{O}$ and $\delta^{18}\text{O}$ of water) in convective regions on two different time scales: (i) during the African monsoon onset and intra-seasonal variability (Banizoumbou, 2006) and (ii) during the squall line of the 11th of August 2006 (Niamey). $^{17}\text{O}_{\text{excess}}$ responds to the monsoon onset by a ~ 30 per meg increase as well as to different convective processes in squall lines by ~ 20 per meg variations. These variations parallel those of d-excess at first order and display significant correlation with relative humidity in the lower troposphere. Still, higher correlation coefficients are observed between d-excess and relative humidity than between $^{17}\text{O}_{\text{excess}}$ and relative humidity, suggesting a higher influence of relative humidity on d-excess than on $^{17}\text{O}_{\text{excess}}$. Using a simple reevaporation model and a more sophisticated 2D model of a squall line, we show that reevaporation is the process explaining the increase of d-excess and $^{17}\text{O}_{\text{excess}}$ with relative humidity for these two studies. We also show that the combination of $^{17}\text{O}_{\text{excess}}$ and d-excess is a powerful tool to constrain the representation of isotopic processes during rain reevaporation. In turn, a good representation of such processes enables to use water isotopes to evaluate convective parameterization in atmospheric models.

1. Introduction

Water stable isotopes ($\delta^{18}\text{O}$, δD) have long been used as markers for temperature in high latitudes (e.g. polar ice core studies). In the Tropics, the amount effect (i.e. the anticorrelation between the rain amount and the $\delta^{18}\text{O}$ or δD of the corresponding collected precipitation) dominates the isotope variability (Dansgaard, 1964; Rozanski et al., 1993). However, recent studies at a monthly or a longer timescale, combining both observations and modeling, have shown that a variety of factors can affect $\delta^{18}\text{O}$ or δD , such as the history of air masses, regional convection or water vapor transport (Yoshimura et al., 2003; Vuille and Werner, 2005; Vimeux et al., 2005; Villacis et al., 2008; Risi et al., 2008; Lee et al., 2009). Moreover, a recent 1D vertical radiative convective modeling study has highlighted that the amount effect is mainly due to two major convective processes: rain reevaporation and water vapor transport by convective or meso-scale subsidence (Risi et al., 2008). When strong convection occurs, those two processes deplete the

isotopic composition of precipitation: the high humidity level tends to decrease the droplet reevaporation and the important subsidence brings into the subcloud layer water depleted vapor from higher levels.

To better explore these different influences on tropical precipitation δD and $\delta^{18}\text{O}$, two studies have taken advantage of the 2006 intensive observation phase of the AMMA (African Monsoon Multidisciplinary Analysis) campaign (Redelsperger et al., 2006; Janicot et al., 2008) that took place over the Sahel (West Africa), a region of the world associated with both very intense convective systems and a strong reevaporation of the rain due to the dryness of the air in the vicinity of the Sahara. First, an analysis of the event-based isotopic composition (δD and $\delta^{18}\text{O}$) of precipitation collected in the Niamey area (Niger) has revealed that $\delta^{18}\text{O}$ and δD record the monsoon onset (Risi et al., 2008). Second, high-frequency (intra-event) water isotopic analyses along squall lines have shown that the $\delta^{18}\text{O}$ and δD of precipitation were influenced by the squall line dynamics and rain reevaporation (Risi et al., 2010a).

Combining $\delta^{18}\text{O}$ and δD in the so-called d-excess ($\text{d-excess} = \delta\text{D} - 8 \times \delta^{18}\text{O}$; Dansgaard, 1964) is expected to bring additional information on fractionating processes in convective systems. In particular, modeling d-excess in precipitation along a squall line has suggested that d-excess in precipitation was mainly controlled by reevaporation of the

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falling rain, and was thus closely related to relative humidity (Risi et al., 2010a). Indeed, relative humidity variations strongly modify the relative influence of diffusive and equilibrium fractionation at the liquid–vapor transitions. Then, because the fractionation coefficients associated with δD and $\delta^{18}O$ are significantly different for the diffusive and equilibrium fractionation, a change in the proportion of diffusive and equilibrium fractionations results in a change in d-excess.

Recently, it has been made possible to measure the triple isotopic composition of oxygen in water ($\delta^{17}O$ and $\delta^{18}O$). This has led to a new tracer, $^{17}O_{\text{excess}}$ (Barkan and Luz, 2005; Landais et al., 2006). Whereas $\delta D - \delta^{18}O$ in meteoric waters are linearly related (Craig, 1961), this is not the case for $\delta^{17}O - \delta^{18}O$. Rather, the water isotopic composition follows a linear relationship with a constant slope of 0.528 in a $\ln(\delta^{17}O + 1) - \ln(\delta^{18}O + 1)$ plot. Therefore, although the $^{17}O_{\text{excess}}$ definition has been chosen by analogy with the d-excess definition, it uses the logarithm system (Landais et al., 2008): $^{17}O_{\text{excess}} = \ln(\delta^{17}O + 1) - 0.528 \times \ln(\delta^{18}O + 1)$, where δ is defined as $(R_{\text{sample}}/R_{\text{standard}} - 1)$, and R_{sample} and R_{standard} are the considered isotopic ratios ($H_2^{17}O/H_2^{16}O$ or $H_2^{18}O/H_2^{16}O$) of the sample and standard respectively.

Laboratory experiments, simple isotopic models and recent measurements of water vapor at the ocean surface have suggested that $^{17}O_{\text{excess}}$ should be a tracer of relative humidity in the regions of evaporation (Angert et al., 2004; Barkan and Luz, 2007; Uemura et al., 2010). Furthermore, the influences of temperature or condensation altitude on $^{17}O_{\text{excess}}$ are expected to be insignificant in contrast to d-excess. Indeed, while the ratio of the equilibrium fractionation coefficients depends on the temperature for the system $\delta D - \delta^{18}O$ (associated with d-excess), it is temperature independent for the system $\ln(\delta^{17}O + 1) - \ln(\delta^{18}O + 1)$ (associated with $^{17}O_{\text{excess}}$) (Barkan and Luz, 2005). As a consequence, measurements of $^{17}O_{\text{excess}}$ are thought to have an important added value with respect to d-excess, in particular for better disentangling, the parameters affecting the water isotopic composition (temperature, relative humidity). For instance, it has been shown that the contrasted behaviors of d-excess and $^{17}O_{\text{excess}}$ in polar regions could be explained by d-excess being affected by fractionation processes along the distillation pathway from the evaporation sources to polar regions, while the $^{17}O_{\text{excess}}$ of polar snow was recording much more directly the signal acquired at low latitudes during the surface evaporation of water vapor, especially the relative humidity conditions in which tropical evaporation took place (Risi et al., 2010b). However, studies of $^{17}O_{\text{excess}}$ in precipitation have been limited so far to East Antarctica (Landais et al., 2008) and to a few points in temperate regions (Landais et al., 2006). Measurements of $^{17}O_{\text{excess}}$ in tropical regions are needed to confirm this interpretation and thus better interpret $^{17}O_{\text{excess}}$ variations in paleoclimate studies.

Beyond paleoclimates, there is a strong interest in developing a new tracer for studying the water cycle and atmospheric convection. The parameterization of convective processes in climate models remains a major source of uncertainty in the simulation of the hydrological cycle and its response to climate change (Slingo et al., 1994; Held and Soden, 2006; Zhang and McFarlane, 1995; Jackson et al., 2008). In particular, the representation of reevaporation processes in convective regions, which has been shown to play a key role in the prediction by climate models of the large-scale organization and variability of the tropical atmosphere (Grabowski and Moncrieff, 2004; Zurovac-Jevtic et al., 2006), is difficult to evaluate and to constrain from observations. In this context, water stable isotopes have been suggested as an alternative tool to help evaluate convective parameterizations (Schmidt et al., 2005; Bony et al., 2008; Lee et al., 2009). This is not direct since the representation of isotopic processes is also associated with some uncertainties and tunable parameters. Still we expect from the similarity between d-excess and $^{17}O_{\text{excess}}$ definitions a strong influence of reevaporation on $^{17}O_{\text{excess}}$ if reevaporation is indeed the primary influence on d-excess in precipitation in convective systems. Then, because the relationships between equilibrium and diffusive fractionation coefficients are different in the $\delta D - \delta^{18}O$ (d-excess) system and in the $\ln(\delta^{17}O + 1) -$

$\ln(\delta^{18}O + 1)$ ($^{17}O_{\text{excess}}$) system, the relative variations of $^{17}O_{\text{excess}}$ and d-excess in convective systems should enable us to better describe/parameterize the influence of reevaporation process on water isotopes. In turn, it would enable us to use more confidently the combination of water isotopes measurements to better constrain convective parameterizations themselves.

In this paper, we present the first $^{17}O_{\text{excess}}$ measurements in tropical regions. In order to exploit this new tracer together with $\delta^{18}O$ and d-excess, we have analyzed the water samples collected during the 2006 AMMA field campaign (Risi et al., 2008, 2010a). We first present the $^{17}O_{\text{excess}}$ of the water samples collected after each rain event at Banizoumbou (a village close to Niamey; Fig. 1) to investigate the intra-seasonal to seasonal variability as well as the monsoon offset. Then, we display the $^{17}O_{\text{excess}}$ evolution for water samples collected in Niamey along the 11th of August squall line to explore the effects of convective dynamics. The relationships between d-excess, $^{17}O_{\text{excess}}$ and relative humidity for these two studies are discussed. Finally, we use two different models: a simple reevaporation model and a 2D model representing the transport, microphysical and isotopic processes in the squall line, both fitted with $^{17}O_{\text{excess}}$, to further examine and quantify the influence of reevaporation and thus relative humidity on $^{17}O_{\text{excess}}$ and d-excess of the tropical precipitation associated with convection.

2. Data

2.1. Sampling and previous data

2.1.1. Seasonal variability and monsoon onset

The Sahelian climate is closely tied to the Western African monsoon. It is strongly affected by the position of the ITCZ (Inter-Tropical Convergence Zone), whose northward ($5^{\circ}N$ to $10^{\circ}N$) shift at the beginning of July marks the monsoon onset. During the AMMA 2006 campaign, observations were collected during the dry season (January–February), the monsoon onset (1 June–15 July), the peak of monsoon (15 July–15 September) and the late monsoon (15–30 September) (Janicot et al., 2008). Precipitation was sampled immediately after each rain event at 3 different locations around Niamey (within an area of about 60 km) and measurements of δD and $\delta^{18}O$ performed in 2007 at LSCE with an accuracy of 0.5 and 0.05‰ respectively (Risi et al., 2008). We focus our study on the longest record (derived from Banizoumbou) that extends from the 9th of June to the 14th of September 2006.

2.1.2. Squall line

A squall line is a well-defined form of organized meso-scale convective system that tends to occur in lines or bands. An observationally based conceptual model of squall line cloud-clusters has been proposed by Houze and Betts (1981) and (Gamache and Houze, 1983). Basically, the squall line system can be divided in three adjacent regions: the first (ahead) is the convective region, the second is called the transition zone and the third (behind) is the stratiform region. In the convective region, water and ice are condensed in vigorous convective updrafts which carry moist air from the boundary layer to the upper troposphere and generate strong precipitation. Most of the condensates either precipitate, reevaporate or are detrained to the rear of the squall line. In the stratiform region, the condensate (which is either detrained from the convective zone or formed in the meso-scale updraft of the stratiform zone) precipitates and then potentially reevaporates in unsaturated meso-scale downdrafts. The transition zone is a narrow region of strong subsidence associated with a very low relative humidity (Fig. 2).

Beyond the 4 different squall lines analyzed in Risi et al., 2010a for δD and $\delta^{18}O$, we have chosen to analyze the precipitation samples corresponding to the 11th of August squall line for three reasons. First, this squall line is the one that was subjected to the highest time resolution measurements of δD and $\delta^{18}O$ of rain water. Second, this squall line best depicts the different structures of the convective and stratiform zones associated with strong variations in $\delta^{18}O$ and d-

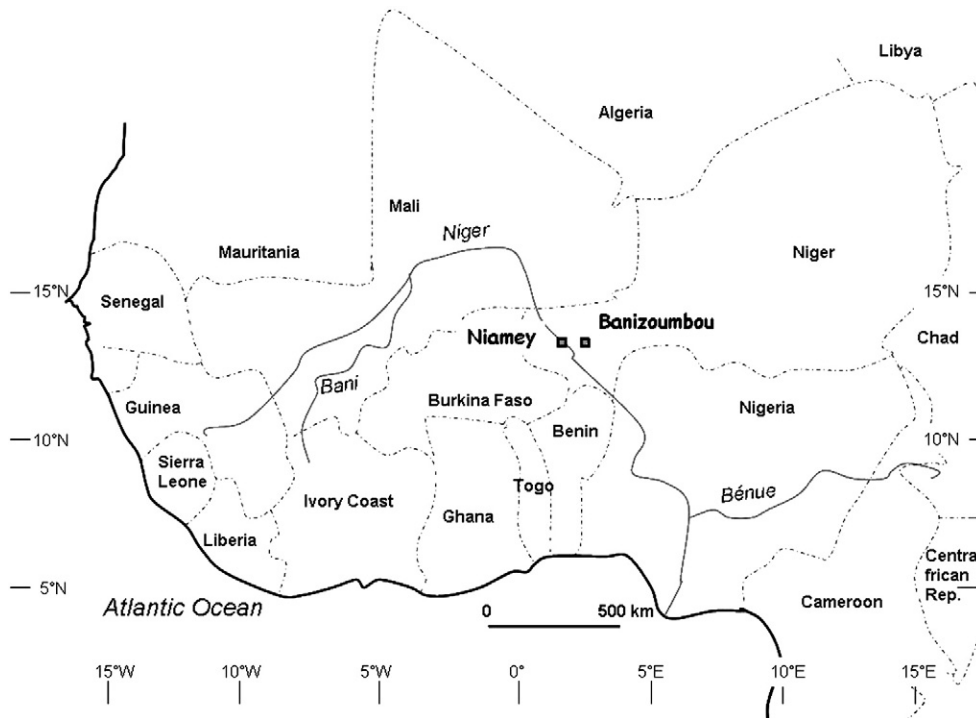


Fig. 1. Map of the West Africa region indicating the location of Banizoumbou and Niamey in Niger.

excess. Third, Risi et al. (2010a) have already analyzed the evolution of δD and $\delta^{18}O$ during this system using a squall line model forced by observed 3D winds (Chong, 2009).

2.2. $^{17}O_{\text{excess}}$ measurements at LSCE

2.2.1. Method

The $^{17}O_{\text{excess}}$ measurements presented here have been obtained following the experimental setup described by Barkan and Luz (2005).

Basically, water is converted to oxygen by fluorination with CoF_3 reagent and the produced pure oxygen is trapped in a tube manifold immersed in liquid helium. Each oxygen tube is then analyzed by dual inlet on a Thermo Delta V for $\delta^{17}O$ and $\delta^{18}O$ (2 runs of 16 dual inlet measurements). Each water sample has been converted and measured 3 times.

Two flasks of working water standard are analyzed daily to check the performance of both the fluorination line and the mass spectrometer. The $\delta^{17}O$ and the $\delta^{18}O$ are expressed with respect to V-SMOW after calibration of our oxygen standard vs. V-SMOW. The resulting uncertainty on $^{17}O_{\text{excess}}$ (see discussion in Landais et al., 2006) is 5 per meg or 0.005‰ (pooled standard deviation).

Note that the $\delta^{18}O$ obtained by the fluorination method and presented in this paper agree well with $\delta^{18}O$ previously obtained by the classical equilibration method (Risi et al., 2008; Risi et al., 2010a), the difference between both being less than 0.05‰.

2.2.2. Seasonal variability and monsoon onset

The evolution of isotopic composition at Banizoumbou between July and September 2006 depicts an abrupt change at the monsoon onset on the 15th of July (Risi et al., 2008; Fig. 3, Supplementary material). The $^{17}O_{\text{excess}}$ also records the offset very clearly, with a shift from -10 per meg to 20 per meg (Fig. 3). In addition, it appears that (1) this shift is slightly delayed in the $^{17}O_{\text{excess}}$ signal compared to the d-excess signal (the $^{17}O_{\text{excess}}$ of the 17th of July is still at -1 per meg while d-excess already reaches a value of $+15.9\text{‰}$) and that (2) $^{17}O_{\text{excess}}$ shows less intra-seasonal variability before the monsoon onset: while the d-excess record depicts a “U-shape” probably related to the degree of organization of the convective systems (Risi et al., 2008), $^{17}O_{\text{excess}}$ remains rather stable. After the monsoon offset, $^{17}O_{\text{excess}}$ exhibits high values, except on the 7th of August where it exhibits an anomalously low precipitation amount, δD and d-excess.

2.2.3. Squall line

$^{17}O_{\text{excess}}$ displays also clear variations along the squall line of the 11th of August 2006 at Niamey (Fig. 4, Supplementary material). The general pattern is a high $^{17}O_{\text{excess}}$ in the convective region (high precipitation rate)

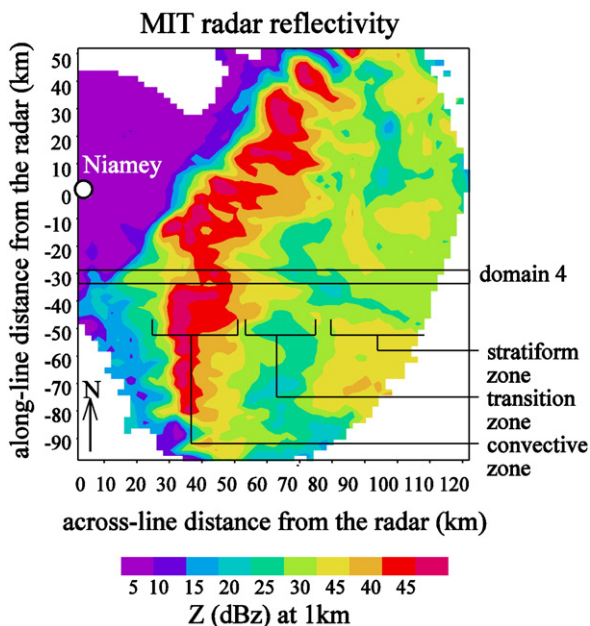


Fig. 2. Horizontal reflectivity map from the MIT radar at 1 km on 11 August 2006 between 02:41 and 03:11 UTC, in the domain of the 3D wind retrieval extending over 120 and 150 km in the west-east and south-north directions, respectively. The domain selected for our analysis as well as the position of the radar in Niamey ($x=0$ km, $y=0$ km) are indicated.

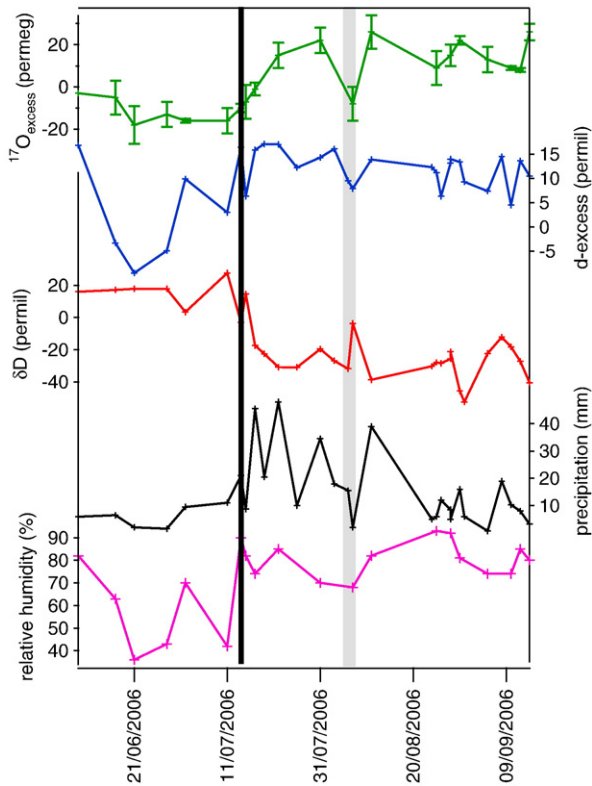


Fig. 3. Evolution of isotopic composition on the event-based sampling during summer 2006 in Banizoumbou (Niger). From top to bottom:

- $^{17}\text{O}_{\text{excess}}$ (green)
- d-excess (blue) (Risi et al., 2008)
- δD (red) (Risi et al., 2008)
- precipitation amount during the individual event (mm) (black)
- relative humidity (%) from radio-sounding in the lowest level (pink)

The thick black line indicates the date of the monsoon onset and the light grey rectangle the location of an isolated precipitation event.

and a $^{17}\text{O}_{\text{excess}}$ lower by 20 per meg over the stratiform region when precipitation rate is low. This general pattern is similar to the d-excess evolution.

2.4. Relative humidity data

To interpret isotopic variations at the seasonal and intra-seasonal timescales, we examine relative humidity from radio-sounding measurements at Niamey. We use here the average of the relative humidity between 200 and 1000 m, but results are not significantly sensitive to this choice of the atmospheric layer.

During the August 11th squall line, no proper measurements of relative humidity have been performed at a high frequency: the ARM (atmospheric radiation measurements) mobile facility microwave radiometer profiler does not provide relative humidity data when it is raining and the surface captor gives the relative humidity only at the surface, which is not representative of the low atmospheric layers where reevaporation takes place. However, the ARM data give information on the relative humidity in the atmospheric levels immediately after the rain. It indicates that between 500 and 3000 m, the relative humidity was around 40% at the end of the stratiform zone. In the absence of additional data, we used a reconstruction of the low level relative humidity (925–1000 hPa) derived from Risi et al. (2010a) who forced a 2D squall line model by observed wind fields to reconstruct relative humidity and isotopic composition of the rain (cf Section 3.2). This reconstruction produces an almost saturated air (relative humidity

higher than 90%) during the convective zone as expected and a relative humidity of 40% at the end of the stratiform zone as measured by ARM. Finally, in order to display the relative humidity reconstruction on the same axis than the records of isotopic composition (Fig. 4), we have synchronized the temporal evolutions of the modeled and measured precipitation rates so that the convective maximum, the stratiform maximum, the transition zone, the beginning and the end of the rain occur at the same moment in both series. Then a linear interpolation has been performed for the intermediate periods.

3. Models

3.1. Description of the simple reevaporation model

In order to study the pure effect of reevaporation on $^{17}\text{O}_{\text{excess}}$ and d-excess evolution, we use a simple model describing reevaporation of falling droplets. This model was first developed by Stewart (1975) and revisited by Bony et al. (2008) who took into account the evolution of the water vapor isotopic composition during evaporation. The Bony et al. (2008) model gives the evolution of the isotopic ratio of the droplet and of the surrounding water vapor (unsaturated downdraft) as a function of relative humidity, initial composition of the liquid and vapor phases, initial fraction of liquid water and residual fraction of rain droplets. Relative humidity has a major influence on fractionation during reevaporation. Indeed, for a saturated air (relative humidity of 100%), reevaporation is very low and there is an equilibrium between the droplets and the surrounding water vapor. On the other

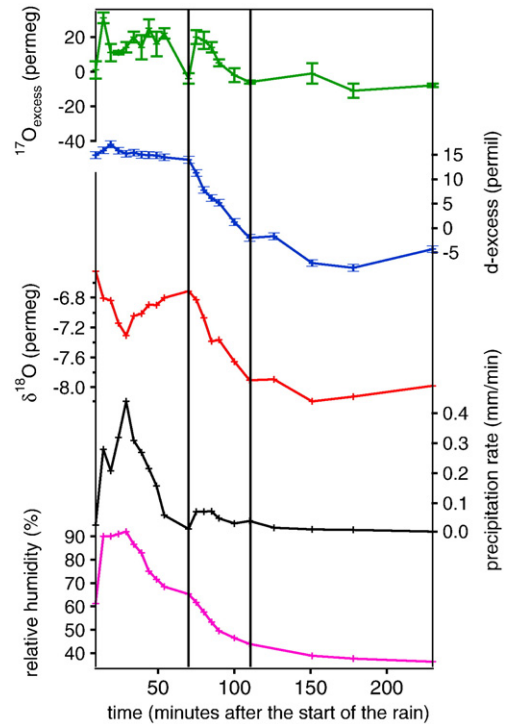


Fig. 4. Evolution of water isotopic composition during the squall line of the 11th of August 2006 (Niamey, Niger). From top to bottom:

- $^{17}\text{O}_{\text{excess}}$ (green)
- d-excess (blue) (Risi et al., 2010a)
- $\delta^{18}\text{O}$ (red) (Risi et al., 2010a)
- precipitation rate (mm min^{-1}) (black) (Risi et al., 2010a)
- relative humidity between 925 and 1000 hPa diagnosed from the 2D squall line model of Risi et al. (2010a) (pink)

The black lines represent the limit between the convective and transition zones (left) and between the transition and stratiform zones (right).

hand, when relative humidity is low, no water vapor can equilibrate with the droplets so that their associated water isotopic ratios increase. The equations used in this paper are exactly those given in the appendix of Bony et al. (2008).

To adapt this model to the $^{17}\text{O}_{\text{excess}}$ description, we used the fractionation coefficients determined by Barkan and Luz (2005) for equilibrium fractionation ($\ln ^{17}\alpha_{\text{eq}} = 0.528 \times \ln ^{18}\alpha_{\text{eq}}$ with $^{17}\alpha_{\text{eq}}$ and $^{18}\alpha_{\text{eq}}$ the equilibrium fractionation coefficients respectively associated with $\text{H}_2^{17}\text{O}/\text{H}_2^{16}\text{O}$ and $\text{H}_2^{18}\text{O}/\text{H}_2^{16}\text{O}$) and Barkan and Luz (2007) for diffusive fractionation ($\ln ^{17}\alpha_{\text{k}} = 0.518 \times \ln ^{18}\alpha_{\text{k}}$ with $^{17}\alpha_{\text{k}}$ and $^{18}\alpha_{\text{k}}$ the kinetic fractionation coefficients for diffusion of water vapor in air respectively associated with $\text{H}_2^{17}\text{O}/\text{H}_2^{16}\text{O}$ and $\text{H}_2^{18}\text{O}/\text{H}_2^{16}\text{O}$).

The Bony et al. (2008) model includes two parameters: (1) an exponent n that modulates the effect of diffusivity of the different water isotopes in air that depends on drop size and that has been tuned to 0.58 (Stewart, 1975; Bony et al., 2008), and (2) ϕ , a parameter used to compute the relative humidity (h_{eff}) of the vapor around the raindrops, which is intermediate between the relative humidity h_b of the surrounding (downdraft) air and h_s , the relative humidity at saturation ($h_s = 1$): $h_{\text{eff}} = \phi \times h_s + (1 - \phi) \times h_b$; ϕ is classically set to 0.9 (Bony et al., 2008; Risi et al., 2010a) but it is a very poorly constrained parameter and the largest source of uncertainty in the simulation of tropical precipitation isotopic composition (Bony et al., 2008). In the next sections, we examine the extent to which an additional tracer of the water cycle ($^{17}\text{O}_{\text{excess}}$) can help to constrain ϕ .

3.2. Description of the 2D model of water isotopes in squall line

In addition to reevaporation, $^{17}\text{O}_{\text{excess}}$ and d-excess variability are likely to be affected by other microphysical and dynamical processes. To examine the role of these different processes, we use the 2D squall line model fitted with water isotopes (Risi et al., 2010a). Water vapor, cloud water and rain are advected by winds using an upstream advection scheme. The 3D wind field derived from the MIT radar by Chong (2009) for the 11th of August squall line is used for the advection. Water vapor condenses as soon as it reaches saturation. To sample the complex dynamical structure of the convective system and reduce the observed 3D wind fields to 2D wind fields needed as inputs by the model, Risi et al. (2010a) have considered several 2D cross-sections along the squall line (Fig. 2 for an example). For each cross-section, 2D winds are averaged over 5 km in the north-south direction (details given in Risi et al., 2010a). The different cross-sections (defined as domains in Risi et al., 2010a; e.g. domain 4 on Fig. 2) induce different wind fields as inputs of the 2D squall line model (Fig. 5).

At each phase change, fractionation occurs and modeled changes in isotopic composition (δD , $\delta^{18}\text{O}$) associated with each process (including rain reevaporation) are represented as in Bony et al. (2008) and Risi et al. (2010a). The adaptation to $^{17}\text{O}_{\text{excess}}$ description has been done in the same way as depicted in the previous section using the differentiation between equilibrium and diffusive fractionation coefficients.

As for the parameters used in the model, ϕ was varied between 0.5 and 0.9 (tests performed outside of this range led to very unrealistic evolution of $\delta^{18}\text{O}$, d-excess and $^{17}\text{O}_{\text{excess}}$). An idealized isotopic profile based on a Rayleigh distillation, to which the evolution of the precipitation composition is not significantly sensitive, is applied at lateral boundaries (Risi et al., 2010a). No air or water can be exchanged at the top and bottom boundaries defined by the model. The surface evaporation is neglected, which is justified both by the fact that the squall line occurred during night-time, and that it lasted only a few hours (Risi et al., 2010a).

Ten different cross-line transects of the squall line were used to model the evolution of precipitation rate, relative humidity as well as $\delta^{18}\text{O}$, d-excess and $^{17}\text{O}_{\text{excess}}$ of the precipitation. For each simulation (referred to as domain 1 to 10), Risi et al. (2010a) have shown that the model was able to reproduce the observed precipitation rate and

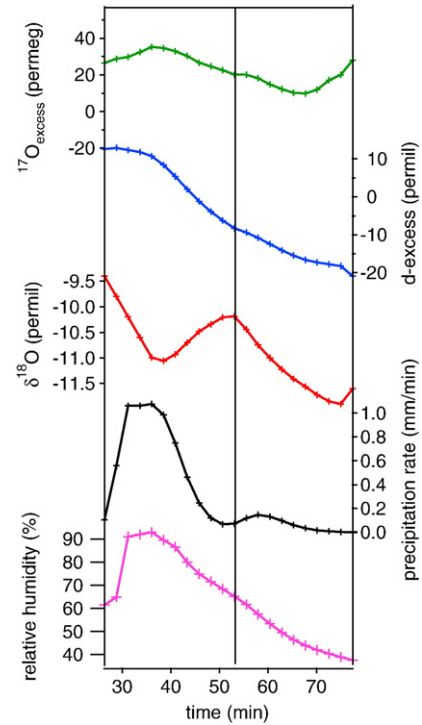


Fig. 5. Outputs from the 2D model of water isotopic composition for the 11 of August 2006 squall line for domain 4. From top to bottom:

- modeled $^{17}\text{O}_{\text{excess}}$ (green)
- modeled d-excess (blue)
- modeled $\delta^{18}\text{O}$ (red)
- modeled precipitation rate (mm min^{-1}) (black)
- modeled relative humidity for the lowest level (1000–925 hPa)

The black lines represent the limit between the convective and transition zones (left) and between the transition and stratiform zones (right).

isotopic evolution. In parallel, the simulated low level relative humidity decreased from almost 100% in the convective zone down to 40–70% in the stratiform zone, with some variability across the different domains. Domain 4 is the domain for which the model results best reproduce the observed evolutions of both $\delta^{18}\text{O}$ and d-excess over the convective and transition zones, and that simulates a drying (40%) at the end of the system in agreement with the ARM data (section 2.4). We thus hereafter focus on this domain.

4. Discussion

From the seasonal variability and squall line analyses, the observed evolutions of $^{17}\text{O}_{\text{excess}}$ and d-excess were shown to display similar variations at first order (monsoon onset, convective to stratiform zones transition) although the details are different (i.e. d-excess exhibits some variability before the monsoon onset while $^{17}\text{O}_{\text{excess}}$ remains stable). In order to learn from these common and different features, we discuss here the relationships between $^{17}\text{O}_{\text{excess}}$, d-excess, precipitation rate and relative humidity and compare with the predictions of the simple reevaporation model.

4.1. Relationship between $^{17}\text{O}_{\text{excess}}$ (d-excess) and precipitation rate

No clear relationship is observed at the seasonal scale between $^{17}\text{O}_{\text{excess}}$ or d-excess and precipitation rate during the summer of 2006 at Banizoumbou (Fig. 6). Along the squall line, some coherency between $^{17}\text{O}_{\text{excess}}$ and the precipitation rate is observed for the low precipitation rates, but no correlation appears for higher precipitation rates (Fig. 6). A

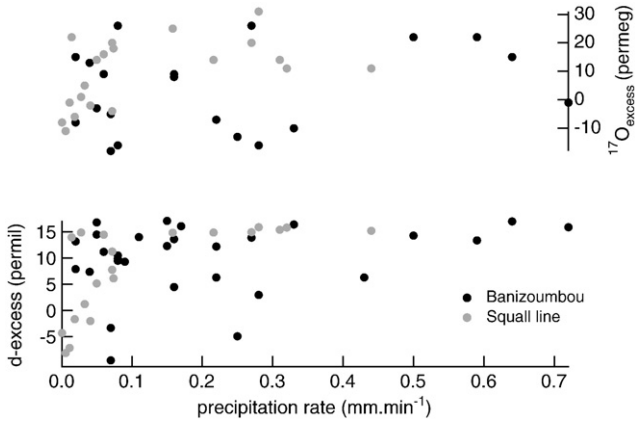


Fig. 6. Relationship between $^{17}\text{O}_{\text{excess}}$ (d-excess) and precipitation rate for (1) the event-based sampling during the whole summer season at Banizoumbou (black points) and for (2) the squall line study (grey points).

coherent feature that still arises from the two studies is that negative values of d-excess and $^{17}\text{O}_{\text{excess}}$ are only encountered for a very low precipitation rate. In contrast to the amount effect that depicts some relationship between $\delta^{18}\text{O}$ or δD and the amount of precipitation, Fig. 6 thus suggests that the precipitation rate does not have a strong direct influence on the second order parameters $^{17}\text{O}_{\text{excess}}$ and d-excess.

4.2. Relationship between $^{17}\text{O}_{\text{excess}}$ (d-excess) and relative humidity

If reevaporation is indeed the process that explains the variations of the isotopic composition of precipitation in the African monsoon, we expect a link between $^{17}\text{O}_{\text{excess}}$ (d-excess) and relative humidity because relative humidity is a key parameter in the droplets reevaporation model (Bony et al., 2008).

Fig. 7a and b show a clear increase of d-excess and $^{17}\text{O}_{\text{excess}}$ with increasing relative humidity at seasonal scale. It appears however that the relationship between d-excess and relative humidity is stronger ($R^2=0.68$) than the relationship between $^{17}\text{O}_{\text{excess}}$ and relative humidity ($R^2=0.34$). This suggests that some other factors may participate in the $^{17}\text{O}_{\text{excess}}$ evolution, such as large-scale processes controlling the isotopic composition of the vapor from which the precipitation forms (origin of air masses, convection and continental recycling upstream air mass trajectories, etc). The occurrence of additional large-scale processes controlling the isotopic composition of the rain is indirectly confirmed by the temporal variations of the positive correlation between precipitation rate and relative humidity (stronger correlation before the monsoon onset than after).

Fig. 8a and b show the relationships between d-excess and relative humidity and between $^{17}\text{O}_{\text{excess}}$ and relative humidity along the Aug 11th 2006 squall line in Niamey. Again, we note a clear increase of d-excess and $^{17}\text{O}_{\text{excess}}$ with increasing relative humidity. Similarly to the event-based sampling experiment, the correlation coefficient is on average higher for the relationship between d-excess and relative humidity ($R^2=0.81$) than for the relationship between $^{17}\text{O}_{\text{excess}}$ and relative humidity ($R^2=0.52$).

Interestingly, the slopes between (1) $^{17}\text{O}_{\text{excess}}$ and relative humidity on the one hand and (2) d-excess and relative humidity on the other hand are very similar for the inter-event variability study at Banizoumbou (respectively 0.53 per meg. $\%^{-1}$ and $0.38\%.\%^{-1}$) and for the squall line (respectively 0.46 per meg. $\%^{-1}$ and $0.41\%.\%^{-1}$). This suggests a similar mechanism at play in the two cases.

4.3. Relationship between $^{17}\text{O}_{\text{excess}}$ and d-excess

For both studies, the slope of $^{17}\text{O}_{\text{excess}}$ vs. d-excess is ~ 1 per meg. $\%^{-1}$ (Fig. 9). Note also that the correlation coefficient is higher for the squall

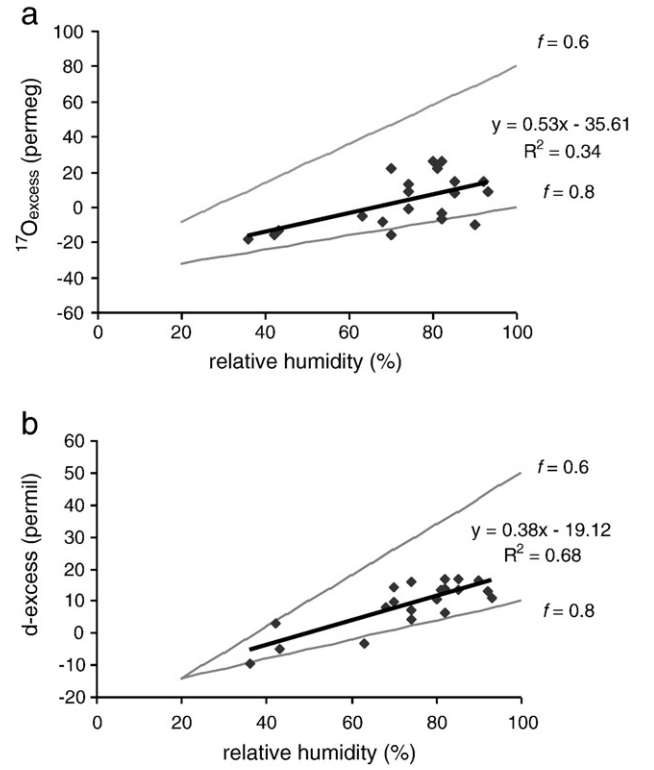


Fig. 7. Variations of $^{17}\text{O}_{\text{excess}}$ with low level relative humidity at the seasonal time scale derived from Banizoumbou data. The grey lines correspond to the lowest (highest) slope of the relationship between $^{17}\text{O}_{\text{excess}}$ and relative humidity that can be obtained with the simple model of reevaporation for $\phi=0.6$ (0.8). Note that the exact fit of the data can be obtained with many different combinations of ϕ between 0.6 and 0.8 , f , p and n in their realistic range of variations. b: Same as panel a but for d-excess.

line ($R^2=0.49$) than for the seasonal and intra-seasonal variability ($R^2=0.29$). This could correspond to the higher R^2 between $^{17}\text{O}_{\text{excess}}$ /d-excess and relative humidity in the squall line than for the inter-event variability at Banizoumbou. This supports the hypothesis that at the seasonal and intra-seasonal time scales, other factors than reevaporation processes may contribute to the evolution of $^{17}\text{O}_{\text{excess}}$. These may be large-scale processes affecting the isotopic composition of the vapor playing at time scales longer than the precipitating system scale, such as the origin of air masses or processes upstream air mass trajectories (upstream convection, continental recycling). Such a hypothesis would also explain why $^{17}\text{O}_{\text{excess}}$ remains almost stable before the monsoon onset while d-excess exhibits large variation with the degree of organization of convective system (Risi et al., 2008).

4.4. Simple reevaporation model – tuning of the parameter ϕ

We have evidenced strong links between $^{17}\text{O}_{\text{excess}}$, d-excess and relative humidity from the two studies detailed above. In order to assess how much rain reevaporation controls the isotopic composition of precipitation, and especially the second order parameters d-excess and $^{17}\text{O}_{\text{excess}}$ (as suggested by Risi et al., 2010a, for d-excess), we use a droplet reevaporation model (Bony et al., 2008) to explore associated variations of d-excess and $^{17}\text{O}_{\text{excess}}$.

With this aim, we performed hundreds of droplet reevaporation simulations under various realistic conditions (varying independently relative humidity, remaining water fraction in the droplet – f , initial isotopic composition of the water vapor, ratio between liquid water and total water mass – p , temperature) with n varying within 20% and with the less constrained parameter ϕ varying between 0.5 and 0.9 . Varying independently relative humidity, f and p is not necessarily very realistic since during reevaporation, f and p should decrease with

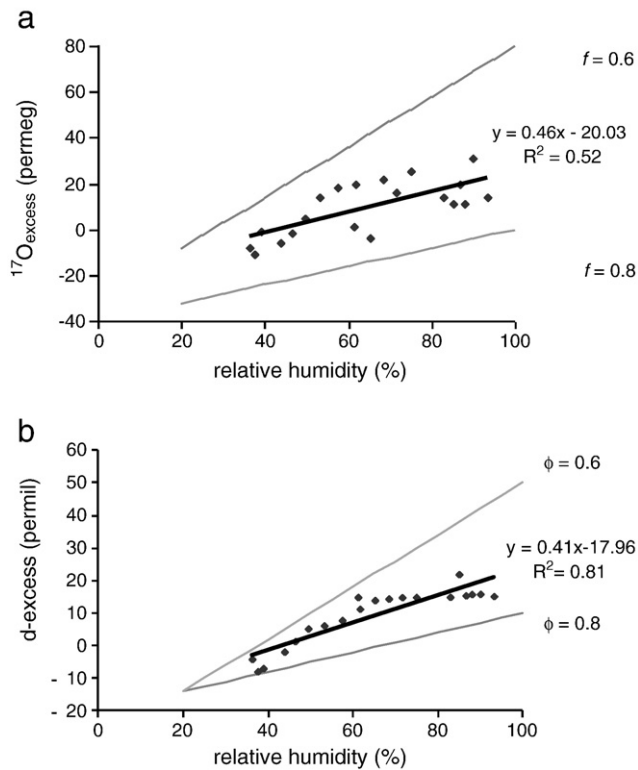


Fig. 8. Variations of $^{17}\text{O}_{\text{excess}}$ with low level relative humidity (reconstructed from domain 4) along the passage of the Aug 11th 2006 squall line (black). The lowest (highest) grey line displays the highest (lowest) slope of $^{17}\text{O}_{\text{excess}}$ vs. relative humidity that can be obtained from the simple reevaporation model with $\phi = 0.8$ (0.6). Note that we cannot discuss the intercepts for relative humidity = 0 with this model so that they have been arbitrary fixed for the clarity of the figure. Note that the exact fit of the data can be obtained with many different combinations of ϕ between 0.6 and 0.8, f , p and n in their realistic range of variations. b: Same as Fig. 8a but for d-excess.

decreasing relative humidity. Indeed, the air becomes drier and the precipitation weaker towards the end of the stratiform zone (Hauser et al., 1988). Therefore, lowest values of relative humidity are generally associated with strong reevaporation and a low proportion of liquid water.

First, we observe that the reevaporation model reproduces well the general increase of both $^{17}\text{O}_{\text{excess}}$ and d-excess with relative humidity. The slope dependence of $^{17}\text{O}_{\text{excess}}$ and d-excess with relative humidity is primarily influenced by parameter ϕ . For $\phi < 0.6$, we found slopes for the relationship between $^{17}\text{O}_{\text{excess}}$ and relative

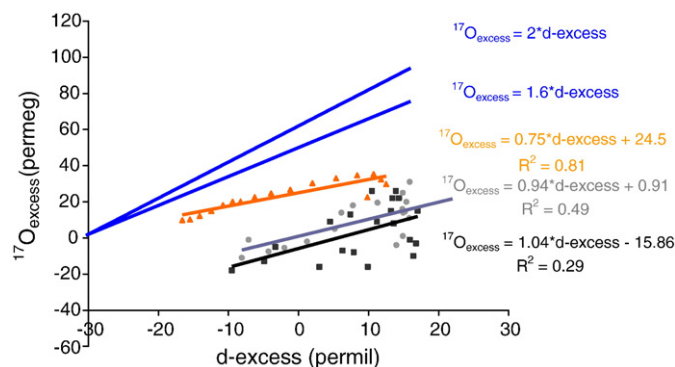


Fig. 9. Variation of measured $^{17}\text{O}_{\text{excess}}$ with d-excess for the squall line study (grey) and the seasonal variability (black). Variations of $^{17}\text{O}_{\text{excess}}$ vs. d-excess calculated with the 2D squall line model for domain 2 (orange). The two blue lines display the slope of $^{17}\text{O}_{\text{excess}}$ vs. d-excess predicted by the simple reevaporation model providing a constant isotopic composition of the water vapor.

humidity higher than 1.1 per meg. $\%^{-1}$ without varying the other parameters concomitantly with relative humidity (Figs. 7 and 8). Variations of f and p associated with those of relative humidity will further increase the slope of $^{17}\text{O}_{\text{excess}}$ with relative humidity. It follows that the modeled slopes $^{17}\text{O}_{\text{excess}}$ (d-excess) vs. relative humidity are significantly higher than the measured ones if $\phi < 0.6$, so that in the following we exclude simulations performed with $\phi < 0.6$. Then, for $\phi > 0.8$, the dependence of $^{17}\text{O}_{\text{excess}}$ and d-excess with relative humidity is very low: even when considering maximum dependences of p and f with relative humidity that can be obtained in extreme conditions in models of convection (Bony et al., 2008; Risi et al., 2010a), the slope of d-excess with relative humidity stays below $0.3\%.\%^{-1}$ and the slope of $^{17}\text{O}_{\text{excess}}$ with relative humidity below 0.4 per meg. $\%^{-1}$ (Figs. 7 and 8). Based on these calculations, we exclude simulations performed with $\phi > 0.8$.

As for the slope $^{17}\text{O}_{\text{excess}}$ vs. d-excess, the sensitivity experiments performed with this simple reevaporation model show that variations in relative humidity, f and p lead to a relationship of $1.6\text{--}2$ per meg. $\%^{-1}$ for the modeled slope $^{17}\text{O}_{\text{excess}}$ vs. d-excess (Fig. 9). This slope remains robust if f co-varies with relative humidity (increasing reevaporation is associated with both lower relative humidity and lower f). The only way to obtain a relationship of 1 per meg. $\%^{-1}$ as given by the data is to include variations induced by changes in the isotopic composition of the water vapor. Indeed, $^{17}\text{O}_{\text{excess}}$ of the water vapor is expected to increase with increasing reevaporation. This is due to input of high $^{17}\text{O}_{\text{excess}}$ vapor by reevaporation upstream of the rear-to-front trajectories in the stratiform zone (the model of Bony et al., 2008, already takes into account the increase in vapor $^{17}\text{O}_{\text{excess}}$ as the local drops reevaporate). We can calculate that an increase of $^{17}\text{O}_{\text{excess}}$ of the water vapor by 15 per meg when decreasing the relative humidity from 100% to 50% (keeping d-excess unchanged) permits to reproduce the observed slope of 1 per meg. $\%^{-1}$ for the relationship between $^{17}\text{O}_{\text{excess}}$ and d-excess. Such a modification of the water vapor isotopic composition is within the range of $^{17}\text{O}_{\text{excess}}$ (25 per meg) predicted in the low level vapor by the 2D squall line model concomitantly with the decrease of relative humidity from the convective to the stratiform zone. In contrast, d-excess of the water vapor remains constant within 8% in this model.

In summary, we have shown that reevaporation explains the evolution of d-excess and $^{17}\text{O}_{\text{excess}}$ with relative humidity well in the convective regions, both at the seasonal and convective time scales. Our data suggest that a ϕ value of 0.7 ± 0.1 should be used in the model of reevaporation formulated by Bony et al. (2008). Then, the relationship between $^{17}\text{O}_{\text{excess}}$ and d-excess can only be explained if we invoke variations in the isotopic composition of the water vapor as the reevaporation proceeds. Going further in the tuning of the reevaporation model as well as inferring the temporal evolutions of f and p from our data is not realistic. First, we are lacking essential data like accurate measurements of relative humidity in the surface layer or of the isotopic composition of the water vapor. Second, with this simple reevaporation model, f , p , the isotopic composition of the water vapor and relative humidity are varied independently one from each other while they are actually related in convective systems. To circumvent this limitation, we now use the 2D squall line model fitted with water isotopes.

4.5. 2D squall line model fitted with water isotopes

Risi et al. (2010a) showed that the temporal evolution of $\delta^{18}\text{O}$ and d-excess along the Aug 11th 2006 squall line could be reproduced with a simple stationary two-dimensional transport model including a representation of cloud microphysics and isotopic fractionation. They also showed that rain reevaporation was a key process in this model to explain the d-excess evolution. Since the present study suggests an important role of reevaporation in the control of both d-excess and $^{17}\text{O}_{\text{excess}}$ along the squall line, we expect a good agreement

between measured $^{17}\text{O}_{\text{excess}}$ and the $^{17}\text{O}_{\text{excess}}$ issued from the 2D model of water isotopes in the squall line.

In a first attempt, ϕ was set to 0.9 in the squall line model using domain 4 as in Risi et al. (2010a). However, with this parameterization, $^{17}\text{O}_{\text{excess}}$ did not display any variations with relative humidity. Actually, the observed evolution of $^{17}\text{O}_{\text{excess}}$ with time (Fig. 4) could only be reproduced with $\phi = 0.7$. Fig. 5 thus displays the modeled precipitation rate, $\delta^{18}\text{O}$, d-excess and $^{17}\text{O}_{\text{excess}}$ with $\phi = 0.7$ using the domain 4 forcing. There is a generally good agreement between data and model for the evolution of precipitation rate, $\delta^{18}\text{O}$ and d-excess with time. The decrease of $\delta^{18}\text{O}$ and d-excess over the transition and stratiform zones is well reproduced with a modeled decrease of 1‰ and 24‰, respectively, for $\delta^{18}\text{O}$ and d-excess while the measurements indicate decrease of 1.5‰ and 23‰. Regarding $^{17}\text{O}_{\text{excess}}$, the agreement is very good: in parallel with the d-excess decrease, the modeled $^{17}\text{O}_{\text{excess}}$ exhibits a slow decrease of 20 per meg from the convective to the end of the stratiform zone, consistent with observations. The main difference appears in the transition zone: while the measured $^{17}\text{O}_{\text{excess}}$ profile depicts a clear decrease, the modeled $^{17}\text{O}_{\text{excess}}$ evolution only marks a pause in the decreasing trend.

As shown for the link between modeled d-excess and relative humidity (Risi et al., 2010a), there is also a strong link between $^{17}\text{O}_{\text{excess}}$ and relative humidity. The inferred slope of the modeled d-excess vs. relative humidity for domain 4 is $0.62\%.\%^{-1}$ while the slope between modeled $^{17}\text{O}_{\text{excess}}$ and relative humidity is lower (0.47 per meg. $\%^{-1}$) with a lower correlation coefficient ($R^2 = 0.86$ for $^{17}\text{O}_{\text{excess}}$ vs. relative humidity and $R^2 = 0.98$ for d-excess vs. relative humidity). These slopes are comparable to the data inferred slopes ($0.41\%.\%^{-1}$ and 0.46 per meg. $\%^{-1}$) and in the range predicted by the pure reevaporation model. This is expected since in this 2D model, the process explaining the positive correlation between d-excess or $^{17}\text{O}_{\text{excess}}$ and relative humidity is reevaporation. Moreover, the fact that R^2 is higher for the correlation between d-excess and relative humidity than for the correlation between $^{17}\text{O}_{\text{excess}}$ and relative humidity is consistent with the larger influence of water vapor $^{17}\text{O}_{\text{excess}}$ variations than water vapor d-excess variations. Finally, the slope between $^{17}\text{O}_{\text{excess}}$ and d-excess in the 2D model is around 0.75 per meg. $\%^{-1}$ (Fig. 9). This is of the same order of magnitude than the measured slope (1 per meg. $\%^{-1}$) and significantly lower than the slope of ~ 2 per meg. $\%^{-1}$ predicted by the simple reevaporation model when the isotopic composition of the water vapor is kept constant. This low slope in the 2D model is indeed due to the simulated variations of water vapor isotopic composition (especially $^{17}\text{O}_{\text{excess}}$) along the squall line.

6. Conclusion

We have presented the first data of the triple isotopic composition of oxygen in water in a tropical and convective atmosphere. In Banizoumbou, the evolution of $^{17}\text{O}_{\text{excess}}$ during the 3 months of the western African summer season clearly records the monsoon onset. Moreover, the evolution of $^{17}\text{O}_{\text{excess}}$ during the passage of a squall line reflects the different dynamical and microphysical processes at play along the convective and stratiform zones of the squall line. We have thus shown that, as $\delta^{18}\text{O}$ and d-excess, this new marker is highly sensitive to convective processes.

The relationship between $^{17}\text{O}_{\text{excess}}$ or d-excess and low level relative humidity shows that relative humidity is key in controlling the water isotopic composition, both at the intra-seasonal and convective time scales. Interestingly, this relationship is comparable for the two sampling time scales, suggesting that the same dominant process is at play. This is confirmed by a simple reevaporation model and by a more sophisticated 2D squall line model, that both show that reevaporation is a key controlling process for the controlling water isotopic composition (especially d-excess and $^{17}\text{O}_{\text{excess}}$) in this region.

However, we also show that the reevaporation processes do not explain all the $^{17}\text{O}_{\text{excess}}$ variability. Indeed, at the seasonal and intra-

seasonal timescale, the correlation between $^{17}\text{O}_{\text{excess}}$ and relative humidity is not very high and systematically lower than the correlation between d-excess and relative humidity. Moreover, the $^{17}\text{O}_{\text{excess}}$ evolution is different from the d-excess one (i.e. less variations of $^{17}\text{O}_{\text{excess}}$ before the monsoon onset, the slowest change of $^{17}\text{O}_{\text{excess}}$ during the monsoon onset). This suggests that while relative humidity variations explain the common signal of d-excess and $^{17}\text{O}_{\text{excess}}$ through the reevaporation process, the influence of large-scale processes on the isotopic composition of water vapor (conditions at the evaporative source, convection and recycling along trajectories) can explain the second order difference between the two proxies. This hypothesis will be checked in future studies by measuring the isotopic composition of the water vapor and by using general circulation models equipped with water isotopes.

The combination of d-excess and $^{17}\text{O}_{\text{excess}}$ data in the simple evaporation model and in the squall line 2D model has allowed us to tune the calculation of the effective relative humidity around the droplet (h_{eff}) used in our simple reevaporation model (Bony et al., 2008; Risi et al., 2010a). In this study, a value of 0.7 for ϕ leads to a better agreement with observations than a value of 0.9. We emphasize that this tuning was made possible only thanks to the additional constraint provided by $^{17}\text{O}_{\text{excess}}$ measurements in addition to $\delta^{18}\text{O}$ and d-excess measurements.

As a conclusion, we stress that the new parameter $^{17}\text{O}_{\text{excess}}$ is essential to better constrain the parameterization of isotopic processes during rain reevaporation. Once this parameterization is constrained, water isotope measurements can be combined to further constrain convective parameterization themselves (i.e. reevaporation rate, downdrafts intensity,...). Moreover, the observational and modeling analyses of the relationship between $^{17}\text{O}_{\text{excess}}$ and d-excess show that variations of the water vapor isotopic composition are non-negligible in convective systems. Therefore, measurements of the water vapor isotopic composition are strongly needed for (1) a more accurate parameterization of the isotopic fractionation associated with rain reevaporation, (2) a possible quantification of the reevaporation rate through simultaneous measurements of precipitation and water vapor isotopic composition and (3) the study of large-scale controls on $^{17}\text{O}_{\text{excess}}$.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.epsl.2010.07.033.

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