Anonymous Referee #1

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In this manuscript, satellite observations of the isotopic composition of free tropospheric water vapor are used to investigate the processes shaping the moisture budget over the subtropical North Atlantic in summer. The study highlights the importance of the Saharan Heat Low in facilitating the uplift and westward transport of moisture from the continental boundary layer to the oceanic free troposphere. The isotope data are used to shed light on seasonal, interannual and spatial variations in the associated moisture transport and mixing processes. In my view, this is a convincing study that provides important mechanistic insights into the subtropical water cycle and demonstrates the usefulness of isotope observations for such process investigations. I still have quite a few comments that mainly relate to the presentation of the methods and results, which in my opinion could be improved at several places. Nevertheless, most of these comments should be easy to resolve for the authors. Note that the manuscript contains several minor language errors, and I do not attempt to list all of them (I think these could be eliminated in the copyediting stage).

We are grateful to Referee #1 for his/her positive and attentive review. His/her numerous comments have been very useful to improve the manuscript. Our answers to referee's comments are shown in blue and changes in text are shown in grey.

Specific Comments:

Title: I think the wording of the current title is a bit awkward. What about 'Importance of the Saharan Heat Low in controlling the North Atlantic free tropospheric humidity budget deduced from IASI dD observations'?

We now follow your suggestion, thank you.

Abstract/Introduction: The last part of the abstract (from line 7) and last part of the introduction are a bit unconnected to the rest (read more like a report, first we did .., then . . .). I'd try to improve the connection between the different parts (SHL, interannual and spatial variability).

Changes have been made to improve the connection.

Abstract: One a[2]dditional sentence on the more general implications of the work would be good.

Added:

> "More generally, our results demonstrate the utility of δD observations obtained from the IASI sounder to gain insight into the hydrological cycle processes in the West African region."

Page 2, lines 3-4: 'the dryness of...': I don't understand this. Also in a moist atmosphere the humidity can be variable (even more in absolute terms).

We meant to refer to the logarithmic dependence of the OLR to changes in specific humidity where in dry area a small change of specific humidity has a great influence on the OLR (greater than a small change in humid areas). But this is now removed for sake of simplicity.

> ~~"and as the dryness of the subtropical atmosphere allows for important variations of the humidity "~~

P 2, 21: 'representation' is probably not the correct word; 'understanding'?

We changed to 'understanding'

P 2, 29-30: ‘the seasonal cycle . . . in summertime’ is a bit contradictory

This has been changed to:

> “(..) the Saharan Heat Low (SHL) - which is a key component of the West African Monsoon system - has a large influence on the budget of water isotopologues above the North Atlantic in summertime, when the SHL is most active, leading to a strong seasonality of δD.”

P 3, 19: ‘filtered based on the residual fit’: What does this mean?

This is related to the retrieval procedure that requires the fitting of computed spectra on the measured spectra. The residual is the difference between the final computed spectra and the measured one. The sentence needs more details to be properly understood and is not of prime importance, we thus removed it

> ~~“The retrievals are also filtered based on the residual of the fit.”~~

Section 2.1: Please add some information on the averaging procedure. For instance, above Izana, do you first calculate daily means by averaging over the individual observations and then monthly means? May there be a bias due to the diurnal cycle? Do you weight the isotope observations by moisture content?

We now add information on the averaging. IASI overpasses are around 9.30 AM and 9.30 PM local time and are likely to induce a bias if there is a strong diurnal cycle.

> “These data are used at different time scales from the individual observation to monthly averages. Daily means are obtained by averaging individual observations from morning and evening IASI measurements which is likely to introduce a bias if there is a diurnal cycle. Monthly averages are obtained from the daily averages.”

Section 2.3: More details on the trajectory setup would be helpful. At which altitude and time of the day are the trajectories initialized? May there be a bias due to temporal mismatches between observations and trajectories? Wouldn’t it be good to quantify also the uncertainty due to different starting altitudes (by using more than one trajectory per day), since also the satellite observations do represent a vertically extended layer?

More details are now given. Referee is right mentioning potential temporal and spatial mismatches between observations and trajectories. We thus tested if trajectories arriving at different altitudes representative of the IASI sensitivity layer have similar patterns and we also tested the temporal differences. The outcome of this test is that the situation presented is generally valid. We provide the different trajectory analyzes in appendix.

P 4, 11: Which reanalysis data set do you use (add reference)?

This is now specified:

> (..) we use backward trajectory calculations from the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Stein et al., 2015) where NCEP GDAS (Global Data Assimilation System) re-analyses (Kleist et al., 2009) have been used as the meteorological fields

P 4, 14 and P5, 8: Does q denote specific humidity or mixing ratio? Please use a consistent nomenclature.

q is the mixing ratio. This has been corrected.

P 5, 14-15: 'intense convective activity': I'd be more specific at this point. As I understand the Worden-paper, it is the recycling/evaporation of precipitation that leads to this increased depletion.

This has been changed to:

> Noteworthy, intense convective activity act to over deplete water vapor through rain-drop re-evaporation and δD-q pairs can be found below the Rayleigh distillation model (Worden et al., 2007).

P 5, 23: There is also a relatively abrupt increase in q. In my view, the differences in autumn are more pronounced.

The now corrected Figure 2 (see next answer) better highlights a difference between the enrichment (in June) and the progressive moistening (from April).

Figure 2: From inspecting this figure, the individual values (e.g, for July) shown in panel b do not seem to average to the value shown in panel a. Do you weight by q? Is this really what one should do when calculating such a multi-annual mean value?

Thank you for the in-depth inspection of the Figure, there was indeed something wrong. We realized that the monthly averages were not properly done from daily averages. This is now corrected and has been double checked: monthly averages are computed from daily means and composites are derived from monthly averages. The seasonality is thus less pronounced that previously stated, so that this is now also corrected in the revised MS.

P 6, 15: 'before...': I don't understand this insertion. Is it really required? At least you don't need the acronym.

Agreed, we have simplified the discussion here.

Figure 4: Axis labels should be added to the first row. 'daily variations' is unclear; do you show daily averages or individual observations?

We show individual observations, this is now stipulated in the legend and axis labels have been added.

Are the Rayleigh and mixing models the same as in Fig. 1 (with the same end members)? Over which levels has the temperature lapse rate been computed? This lapse rate is currently not discussed in the main text (but should be, I think).

They weren't exactly the same, now they are.

We now add the information on the temperature gradient:

> The colour indicates the gradient of temperature computed between 5.5 and 1.5 km.

And is briefly discussed in the text. But this is more discussed in the following section.

P 9, 10: What is the data source for the precipitation amount? Also the reanalysis, which would mean that it's actually a model forecast? What is the accumulation period? This should be mentioned, as it may introduce some uncertainty.

Yes, the precipitation also come from the reanalysis.

The average precipitation amount (computed from the re-analysis at a time step of 3 hours)

Figure 5: 'Daily variations': see above. Please specify how the Richardson number is calculated. The vertical velocity is not discussed and could thus be removed. Potential temperature could be shown as an alternative (which would probably also illustrate the deep mixed layer for the green box).

It is now stated that these are daily variations and not individual observations. Richardson number is from MERRA re-analysis. We removed the vertical velocity panel and choose to not add the potential temperature for sake of simplicity.

Section 3.4: In my opinion, this section disturbs the flow of the paper. I would shift it after section 4, as it provides a transition to the detailed spatial analysis in section 5.

We agree that it could fit before the spatial analysis (it was there in a previous section) but we prefer to have it in Section 3 "Seasonal variations: Influence of the SHL on _D in the subtropical North Atlantic" since we use the seasonality to derive the spatial influence.

P 11, 20 – P12, 2: I don't understand this sentence (the connection between the seasonality in q and the mixing processes).

The bottom panel of Figure 6, which shows the seasonality for the specific humidity (in percent), reveals a different behaviour. The observed maximum in δD wich does not correspond to the maximum of humidity can also be interpreted as the signature of the SHL, as mixing processes produce a stronger isotopic signal for a given specific humidity than any other hydrological processes (Galewsky2010,Noone2012).

Figure 7: The two upper panels could be combined by adding the red line to the uppermost panel.

We now combine the two panels.

Figure 9: The caption says that the ratio in b and c was normalized by the number of air masses from the African continent, but the main text and the axis label suggest that it is normalized by the total number of air masses.

Thank you, it is the ratio of #of western air-massess/# total number

P 14, 7: The wording of the first sentence is unclear.

Changed to:

> In this section we translate the control of the airmass origins in terms of mixing fraction of the mixing.

P 14, 17: 'very similar values' instead of 'always the same value' (it does vary a bit due to changes in SST)

Yes referee is right, this has been corrected.

Figure 11: Note that each point represents one location over the North Atlantic. Why do the arrows indicate linear pathways (your simple models describe curved paths in the q-dD space)?

The idea of the figure was to show that the δD-q pairs distribution can be decomposed into different pathways from three different points (sources). The arrows have no physical meaning here.

P 16, 12: 'can easily be distinguished': This is a bit subjective. How do you do this? Are the circles just positioned subjectively?

We try to be less subjective:

> "The latter are identified as the moist members of the different branches visually identified of the δD-q pairs scatter plot."

Are all data points within the circles shown in panel a of Fig. 12 (this should be explicitly mentioned in the caption)?

The circles were "drawn by hand" and some contours on q and δD were arbitrarily defined. The contours are now better defined. See also our answer to your last comment.

P 17, 25: 'processes . . . are horizontal': I don't think that this can be concluded from the present analysis. I'm pretty sure that the descent or ascent of air masses is important for shaping these patterns (as you have demonstrated, e.g., for the SHL).

Agreed. This is conclusion was a bit simplistic as the stronger subsidence is likely to contribute to the depletion and drying Northward. This has been removed and we now just mentioned:

> "Note that the sources show quasi constant δD and q values while the dehydration pathways, on the other hand, show an important variability . The dehydration and depletion is mainly latitudinal."

Figure 12: S2 and S3 are interchanged in panel c. Why are there gaps in the geographical locations of the pathways in the tropics in panel d? More general: Are the pathways defined in geographical or in the q-dD space? Why and how? For instance, in panel c there are some green points (P3) that I would visually attribute to P4.

The S2-S3 swap is now corrected, thank you.

The best would have been to dissociate the different pathways from their position in the q-δd diagrams. However to simplify the procedure, we sometimes used the geographical locations of the q-δd pairs. This is why the geographical limits between the different pathways are sometimes sharp. This is now explicitly stated. We could indeed think that some green point are related to P4 however their geographical position prevent us to link them to P4.

P 19, 3: Figure 12e instead of 11

ok

Section 5.2: I think some discussion should be added to this section. How unambiguous is the definition of the different pathways? Basically, one could reach every position in the q-dD space (in between your simple models of Fig. 1) by combining different Rayleigh and mixing lines.

The analysis proposed here suggest that the combined observation of water vapor and its isotopic composition can be very useful to identify the different sources of humidity, which are key actors of the hydrological and dynamical cycle of the region, and their interactions. It is however impossible to unambiguously assess the processes responsible of the position of δD-q pairs as combination of different processes can lead to a same δD-q position. Nevertheless, the coherence of our interpretation with the actual understanding of the SHL dynamic suggest that it is a reasonable interpretation.

Anonymous Referee #2

In this manuscript, the authors present the isotopic composition of water vapour in the subtropical North Atlantic free troposphere investigated with IASI measurements. This work can be seen as a further step of previous water vapour isotopologues studies carried out in the same region, involving in-situ, ground-based and space-based techniques. In these studies, the observed H2O-δD distribution was characterized as a function of the origin of the airmasses. Here, the authors focus on summer time, where H2O-δD distributions show the mixing between dry and depleted upper tropospheric air with humid and enriched boundary layer air transported within the Saharan air layer. The novelty of the work relies on the identification of the Saharan Heat Low (SHL) as the mechanism controlling the moisture budget in the subtropical Atlantic during summer. This work also shows a simple technique for interpreting the interannual variations of δD as a function of the fraction of western to eastern airmasses arriving at Izaña. Overall, this is a well-written and very interesting manuscript. I recommend publication subject to minor revisions.

The Authors would like to thank the reviewer #2 for reviewing this manuscript and for providing comments. Our answers to referee's comments are shown in blue and changes in text are shown in grey.

The specific comments described below are in relation of a general concern of lack of highlighting previous works developed in this region, which would help to justify the used tools and support the findings.

SC#1. Section 2.1 IASI δD retrievals: The authors use δD IASI retrievals to demonstrate the role of the SHL on the seasonal cycle of the water isotopologue budget above the North Atlantic in summertime. The 5-year ERC Project MUSICA focused on the long-term, global and high-resolution observations of tropospheric H2O-δD. This project used Izaña as a multiplatform site for improving the retrievals of ground-based FTIR and IASI sensors, by comparing with in-situ measurements and airborne profiles. Besides the relevance of the named project on the results of this work, there is no specific mention of it. I recommend including in line 20 of section 2.1, the more recent results of IASI observational errors that can be found in Schneider et al. (2015*, 2016, 2017*), and discuss the use of different approximations for the IASI retrievals.

* Schneider et al., Atmos. Meas. Tech., 8, 483–503, doi:10.5194/amt-8-483-2015, 2015

**Schneider et al., Atmos. Meas. Tech., 10, 507-525, doi:10.5194/amt-10-507-2017, 2017.

We followed your recommendation concerning the lack of references to the work done within the MUSICA project in that region. More references on previous work related to the sensitivity of δD to airmasses history are added in the introduction. See also our more detailed reply to Matthias Schneider who also pointed out a lack of discussion on the MUSICA work.

We also now make sure that it is clear for the reader that we use the retrieval of δD from IASI developed at ULB/LATMOS and not the MUSICA one. Concerning the description of IASI error, we thus only refer to the corresponding error characterization works (Lacour et al.,2012 and Lacour et al., 2015).

SC#2.

Section 2.2 TES δD retrievals: Please include a line describing the observational error for TES retrievals (section 2.2).

Added:

> The observational error on δD retrieved values from TES has been evaluated to 30‰ (Worden et al., 2012; Herman et al., 2014).

SC#3. Section 3.1 Seasonal cycle of water vapour and its isotopic composition over Izaña: Is the composite seasonal cycle representative of different years? Are this data in agreement with the inter-annual variability observed with the in-situ records at Izaña? Please, check and discuss.

Yes the composite is realized from 5 years of IASI data (2009-2013). However, it is difficult to compare with in situ data at Izana since the ground-based data described in Gonzalez et al.(2015) is for the years 2012 to 2013 at one site and 2013 to 2015 for another site, hence with little temporal overlap with our dataset.. Moreover Gonzalez et al. (2015) showed there was an important diurnal cycle of δd due to the development/displacement of the boundary layer with night measurements being the most representative of the free troposphere. A comparison of in-situ measurements with IASI ones is thus not straightforward and would require the development of a cautious frame to do so properly. Nevertheless, this would be a work of interest.

SC#4. Section 3.3 Relationship between the SHL and the summer enrichment over the Atlantic: The four clusters described in the δD-q distribution plot (Figure 5a) were already observed in the situ-measurements at Izaña. Please, link and discuss the observations. I would also like to see some references on the discussion on the dynamics of the Saharan Air Layer.

Referee is right, this is a link we should have made.

> Noteworthy, the four boxes delimiting the different δD-q signatures (here defined arbitrarily) are very similar to the dissociation of in situ δD measurements by Gonzales et al., 2016 based on the temperature of the last condensation of air parcels.

We also add a discussion on the SAL:

> The spatial pattern drawn by the high seasonality of δD can be linked to the dynamics of the SAL. Tsamalis et al. (2013) have shown that the SAL displays clear seasonal cycle (both in latitudinal extent and in vertical structure), using 5 years of data from the space-borne Cloud-Aerosol Lidar with Orthogonal Polarization (Winker et al., 2010). The SAL occurs at higher altitudes and farther north during the summer than during winter. Near the African coastline, the SAL is found between 5-30°N in summer, its northern edge being observed just north of the Canary Islands. The northern edge of the SAL migrates to 15_N during the winter, and is generally observed to be south of the Canary Islands from September to May (see Figure 2 of Tsamalis et al. 2013). During the summer, the SAL is found to be thicker and higher off the coast of Africa, between 1 and 5 km above mean sea level, while it is observed between 1 and 3 km during the winter (see Figure 4 of Tsamalis et al. 2013), i.e. below the altitude of maximum sensitivity of IASI-derived δD products.

SC#6. Please correct typo in page 15, line 17. It is read “2103”, instead of “20

Corrected, thank you.

Matthias Schneider short comment

The paper presents and interprets δD and {H2O,δD} pair distributions obtained from IASI spectra for the North Atlantic region. It gives interesting insight into the possibilities of such measurements for investigating tropospheric moisture pathways. Similar studies have already been made during the project MUSICA and the respective results are published in several papers: Schneider et al. (2015), Dyroff et al. (2015), González et al. (2016), Schneider et al. (2016). The MUSICA works have been focused on demonstrating the quality and the potential of the remote sensing data whereas this study focuses more on the scientific interpretation of the data. So there are similarities but also clear differences with respect to the MUSICA studies and this new work is very interesting for the scientific community. However, I think it would be important to relate this new work better to the previous MUSICA studies, mention the similarities, and highlight the new aspects.

We thank Dr. Schneider for having taken the time to comment on our manuscript. We agree with him that similar studies have been conducted in the framework of the MUSICA project. The different seasonal δd-q signatures above Izana have been observed from our IASI retrieval for a few years (i.e. Lacour J.-L. ULB PhD thesis in 2015) and much work has been done trying to understand the link with the dynamics of the region. It is the purpose of the present study to improve our knowledge of the influence of the regional atmospheric dynamics on the water vapor budget over the Northeastern Atlantic, and of the processes leading to the seasonal q-δd distribution. We agree with him that some references on MUSICA work related to the sensitivity of q-δd pairs to moisture pathways in that region were missing and we believe this has been now corrected.

First, I would very much like to see a statement in the Abstract and/or in the Introduction Section telling the reader that for this new study IASI data generated by the ULB IASI retrieval processor (Lacour et al., 2012; Pommier et al., 2014) are used. In this new study the subtropical North Atlantic moisture pathways are studied for the first time with the ULB IASI retrieval processor data. The here presented data are not generated by the MUSICA MetOp/IASI retrieval processor (Schneider and Hase, 2011; Schneider et al., 2016). The MUSICA MetOp/IASI data have already been used previously for documenting the different moisture pathways in the subtropical North Atlantic region. The technical details of these retrieval processor differences should maybe not be discussed in an ACP manuscript however, I think it is important to mention that there are different processors. The reason is that the retrieval processor differences can importantly affect the products (Worden et al., 2012, http://www.atmos-meas-tech.net/5/397/2012/): For instance, while the MUSICA processor works with a broad spectral window (Frank and Hase, 2011; Wiegele et al., 2014) similar to the new TES retrieval processor (Worden et al., 2012), the ULB IASI processor fits smaller spectral windows (Lacour et al., 2012). A brief summary of the differences of the processors is given in the Appendix of Schneider et al. (2016).

We agree that the information on the retrieval processor might not be clear for the reader. This is now clarified in the introduction:

> In this study, we use δD and humidity profiles retrieved from IASI at ULB/LATMOS (Lacour et al., 2012; Lacour et al., 2015).

This is also clearly stated in the data section.

We added the missing references about the identification of moisture pathways from IASI and FTIR MUSICA products.

> From in situ measurements at Izana, González et al. (2016) have shown that different airmass pathways could be detected in H2O-δD pairs distribution. The sensitivity of δD observations to different moisture pathways have also been reported from ground based FTIR and IASI measurements (Schneider et al., 2015) within the MUSICA project (Schneider et al., 2016). Here, we use IASI H2O and δD ULB/LATMOS retrieval products (..)

Second, I would like to recommend setting the here presented data interpretation approaches and the achieved results better in relation to the respective MUSICA activities. In my opinion it would be good to clarify what aspects have already been addressed in the MUSICA papers and what aspects go beyond previous MUSICA works. For example the interpretation of the MUSICA NDACC/FTIR and MUSICA MetOp/IASI {H2O,δD} remote sensing data as shown in Schneider et al. (2015 and 2016) is very similar to what is shown in this new paper in the Sections 3.1, 3.2, and 3.3. Some differences exist in the use of the backward trajectories (in the MUSICA studies the trajectories end at the last condensation point and here they can go beyond the condensation point) and this new work provides an analysis of individual months (taking the summer 2012 as example), whereas the MUSICA studies are mainly limited to an analyses of the overall situation.

> Aspects that have not been addressed by the respective MUSICA studies or that have been addressed by using a different approach could then be better highlighted: For instance, Section 3.4 presents a geographically expanded picture if compared to the MUSICA studies and Section 4 shows that quantifying the strength of the Saharan boundary layer mixing signal is possible by a simple backward trajectory analyses whereas in MUSICA the link to the Saharan boundary layer has been documented by coinciding observations of dust concentrations. The discussion of pathways (Section 5) has similarities to the MUSICA works, but also offers interesting new aspects, like the consideration of a wider geographic region. The clear message from the example study of July 2012 is that such satellite data can be really helpful for investigating geographically varying moisture pathways.

There are indeed similarities with previous work done within MUSICA but we believe the general approach proposed here is somehow different than the previous studies you report and can thus not be compared easily. We went through our manuscript again and made sure the previous works are properly addressed. Similarities with the work by Gonzalez is also now better addressed. (See referee #2 comments). Schneider et al. (2015 and 2016) clearly showed the sensitivity of δD to different air parcels trajectories, which is now clearly acknowledged in our manuscript but we do not think it is a good idea to compare every similarity in that study. In Section 3.1 3.2 and 3.3 of our paper we use the sensitivity of δD to different moisture sources to show that we have a clear signal associated to the arrival of the SHL. The message of the section 3 is the concomitant signal in δD and the SHL. The use of backward trajectory analyses is frequent in isotope analysis and we believe it is not worth to mention that some differences occur from one backward trajectory analyze to another. We agree with referee that we could link the section 4 with Schneider et al. (2015) concerning the coincidence of dust and high δd values. So we added:

> Schneider et al., (2015) already documented the link between high δD values and the continental origin of the airmass by coinciding observations of dust concentrations.

I have furthermore an important remark on the discussion provided in Section 2.4 and on the use of {H2O,δD} pair remote sensing data. As shown in Schneider et al. (2016, and references therein) it is important to ensure that the H2O and δD remote sensing products represent the same vertical altitudes, otherwise defective interpretations of the {H2O,δD} pair distributions are very likely. For this purpose the MUSICA {H2O,δD} pairs are generated by an a posteriori processing (the Type 2 product, which ensures that the {H2O,δD} pair distributions can be correctly interpreted). I think it would be important to clarify how this problem has been addressed for the here presented {H2O,δD} pair data.

We are aware of the post processing methodology the referee proposed to ensure that δD and humidity vertical profiles are representative of the exact same vertical profile. We agree that such post processing simplifies the interpretation of δd-H2O pairs for quantitative purpose. However in this study, we did not apply the post processing method on our IASI retrievals. The reason for that is the data used in that study have been processed a long time ago (the a posteriori processing on IASI data is now operational) and after verification on some samples of our data, we found that applying the post processing does not affect significantly the interpretation of our results. Moreover it is fine from an optimal estimation point of view to do so (in that case the smoothing contribution to the error is greater for δD than for H2O and δD are less precise than q). We show an example of the post processing in Figure S1 for one day of data above the North Atlantic (0-40°N/40°W-5°E) for data at 3.5 km and data at 5.5 km. As one can see there are indeed changes. The changes are especially important for humid data at 3.5 km (remind that we do not use data at 3.5 km) within convective area which is because when degrading the humidity profile at δD resolution, the vertical resolution (of H2O) becomes larger and at this altitude, more sensitive to the boundary layer which is very humid within this area. At 5.5 km there are also differences but the latter are unlikely to affect our analysis in δD-q space. We should also be careful when translating the results of Schneider et al., 2016 to our retrievals. As you mentioned the retrieval schemes are different and are likely to be differently sensitive. For example, we indeed use a relatively small spectral range centered on HDO maximum Jacobians and avoid fitting a large region where there is a lot of information on H2O. Our retrieval is thus not optimized to provide high resolution vertical profile of H2O and their AVK are more similar to δD ones than in the MUSICA retrieval scheme. Nevertheless, while we believe the post-processing should not affect the interpretation of the present study, it is worth noting that the a posteriori processing has recently been adopted in the processing of IASI data for simplifying the analysis made by end-users.

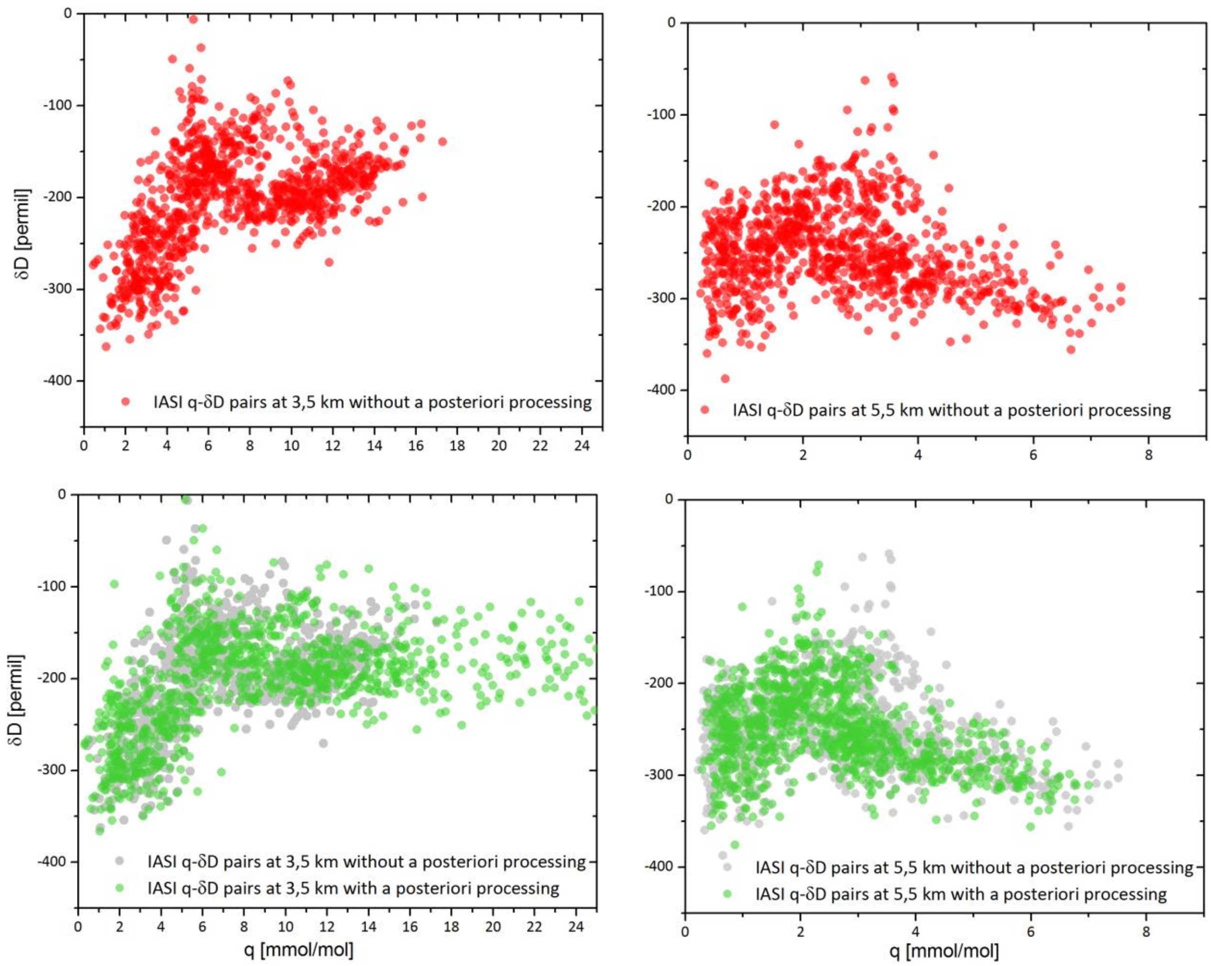

Figure 1 Differences between retrieved δD -q pairs at 3.5 and 5.5 km with and without a posteriori processing

In the data section, we now specify that the sensitivity of Dd and q are not the same:

> It is also important to mention that the δD and humidity retrieved profiles are not exactly representative of the same atmosphere, the humidity profile having more vertical information than δD. It is thus important to keep in mind that when δD-q pairs are considered, the δD estimate is representative of a thicker layer of the atmosphere than the q estimate.

I think it would be also important to mention that the transport out of the Saharan boundary layer to the atmosphere above the Canary Archipelago has been studied since many years mainly by the aerosol community (there are leading experts at the Izaña Observatory) and there are a lot of publications available, which I would like to recommend considering (e.g. Rodriguez et al., 2011, http://www.atmoschem-phys.net/11/6663/2011/ or Rodríguez et al., 2015, http://www.atmos-chemphys.net/15/7471/2015/, and references therein). Also interesting in this context could be to have a look on the works published for a current ACP/AMT special issue (http://www.atmos-chemphys.net/special_issue382.html).

We feel that discussing the transport of aerosols towards the Izana Observatory is out of the scope of the study and for the sake of clarity and conciseness we have decided not to dedicate a specific paragraph on the aerosol transport.

As a minor remark I would like to recommend not talking about “data above Izaña” (Izaña is a hill on Tenerife Island and the name of an observatory on this hill). When talking about a region that covers actually the whole Canary Archipelago I would like to recommend using something like “data representative for the Canary Archipelago region”. Finally, I would like to recommend considering for the Introduction Section a reference to the review of Galewski et al. (2016, Rev. Geophys., 54, doi:10.1002/2015RG000512).

We now speak of the Canary Archipelago region.

We now cite the review paper by Galewsky.

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Importance of the Saharan Heat Low ~~on~~ in controlling the ~~control of the~~ North Atlantic free tropospheric humidity budget deduced from IASI δD observations

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Abstract. The isotopic composition of water vapour in the North Atlantic free troposphere is investigated with IASI measurements of the D/H ratio (δD) above the ocean. We show that in the vicinity of West Africa, the seasonality of δD is particularly strong (~~160~~130‰), which is related with the installation of the Saharan Heat Low (SHL) during summertime. The SHL indeed largely influences the dynamic in that region by producing deep turbulent mixing layers, yielding a specific water vapor isotopic footprint. The influence of the SHL on the isotopic budget is analysed at various time and space scales and is shown to be large, highlighting the importance of the SHL dynamics on the moistening and the HDO-enrichment of the free troposphere over the North Atlantic. ~~We also report important~~ The potential influence of the SHL is also investigated at the inter-annual scale as we also report important variations of δD above ~~Izana (Canary Islands) that we interpret~~ the Canary Archipelago region. We interpret the variability of the enrichment, using backward trajectory analyses, in terms of the ratio of air-masses coming from the North Atlantic and air-masses coming from the African continent. Finally, ~~we present~~ the interest of IASI high sampling capabilities is further illustrated by presenting spatial distributions of δD and humidity above the North Atlantic ~~and~~ from which we show that the different sources and dehydration pathways controlling the humidity can be disentangled thanks to the added value of δD observations. More generally, our results demonstrate the utility of δD observations obtained from the IASI sounder to gain insight into the hydrological cycle processes in the West African region.

1 Introduction

In the North Atlantic, the free tropospheric humidity is the result of a complex interplay between moistening and dehydrating processes of air parcels originating from different sources. While the large scale subsidence largely controls the dryness of the subtropics, numerous other processes have been shown to moisten the subsiding air, such as large scale transport from the tropics (Sherwood, 1996) or vertical mixing associated with convection, or evaporation of condensate in the convective towers (Sun and Lindzen, 1993). Due to the difficulty to disentangle the relative contributions of these processes and of the different sources, controls on the tropospheric humidity remain imprecise. Additionally, these different sources and processes may be affected by the modulation of regional environmental influences such as the migration of the Inter Tropical Convergence Zone (ITCZ) (Wilcox et al., 2010), the activity of the Saharan Heat Low (SHL) (Lavaysse et al., 2010) or the West African monsoon and associated mesoscale convective systems. Furthermore, the subtropical North Atlantic is a particularly climate sensitive area (Spencer and Braswell, 1997) as the radiative forcing associated with changes in water vapour is the strongest in the free troposphere (Held and Soden, 2000)~~and as the dryness of the subtropical atmosphere allows for important variations of the humidity~~. Efforts in understanding the controls on the North Atlantic humidity are thus crucial.

The measurement of water vapour isotopologues has proved to be a helpful observational diagnostic to study the atmospheric moistening/dehydrating processes in a novel way (e.g. ~~Worden et al. 2007; Frankenberg et al. 2009; Risi~~ This is because the water isotopologues (HDO, $H_2{}^{16}O$, $H_2{}^{18}O$) preferentially condense/evaporate during the phase changes of water, and therefore their isotopic ratio is sensitive to key processes of the hydrological cycle such as airmass mixing (Galewsky et al., 2007), convection (Risi et al., 2008), transport (Galewsky and Samuels-Crow, 2015). The observation of water isotopologues in the vapour provide thereby useful information on the processes that affected the air-masses downwind. While the number of δD ($\delta\mathrm{D} = 1000 * [(\mathrm{HDO}/\mathrm{H_2O})/\mathrm{Rsmow} - 1]$, Rsmow being the HDO/H_2O ratio in the standard mean ocean waters) measurements has increased this last decade (e.g. Lacour et al. 2012; Worden et al. 2012; Schneider et al. 2016), it is necessary to understand the factors controlling their variations in order to apprehend their utility for studying hydrological processes. With its demonstrated capabilities to provide δD measurements in the free troposphere (Schneider and Hase, 2011; Lacour et al., 2012, 2015), the Infrared Atmospheric Sounding Interferometer (IASI) flying onboard MetOp has since a decade a key role in supplying δD observations. IASI has the advantage to make about 1.3 millions measurements a day, which almost ensures one measurement everywhere twice a day. Up to now, IASI δD retrievals have been sparsely used for geophysical analyses (Bonne et al., 2015; Tuinenburg et al., 2015).

In this study, we ~~explore~~ use δD ~~distributions derived~~ and humidity profiles retrieved from IASI at ULB/LATMOS (Lacour et al., 2012, 2015) to explore the isotopic signal at various time and space scales above the North Atlantic near the West African coast and we interpret their seasonal to interannual variability as well as their spatial variations. This enables us to investigate the potential of such observations to improve our ~~representation~~ understanding of the moistening processes in this region. Because the retrieval of δD above deserts is difficult due to uncertainties in the surface emissivity and the presence of dust, we have chosen to not analyse the measurements above the Sahara. Nevertheless, because of the integrated nature of δD, we show that some information can be derived from δD signature of air parcels coming from the desert. Former studies have already been dedicated to the interpretation of isotopic variations observed in precipitation and in water vapour in West Africa (Frankenberg et al., 2009; Risi et al., 2010; Okazaki et al., 2015) by combining models with observations, but solely focusing on the role of convection. From in situ measurements at Izana, González et al. (2016) have ~~also~~ shown that different airmass pathways could be detected in H_2O-δD pairs distribution. ~~Here, we use IASI measurements of~~ The sensitivity of δD observations to different moisture pathways have also been reported from ground based FTIR and IASI measurements (Schneider et al., 2015) within the MUSICA project (Schneider et al., 2016). Here, from IASI H_2O and δD ULB/LATMOS retrieval products, we use this property of isotope to first show that the Saharan Heat Low (SHL) - which is a key component of the West African Monsoon system - has a large influence on the ~~seasonal cycle of the~~ budget of water isotopologues above the North Atlantic in summertime, when the SHL is most active, leading to a strong seasonality of δD. Then, we present inter-annual variability of the isotopic composition ~~at Izana (Canary Islands, 100 km off the coast of Africa~~above the Canary Archipelago Region (CAR) and analyse the causes of the variability. Finally, we detail the spatial variations of water vapour and its isotopic composition above the North Atlantic for July 2012 and discuss the different sources and dehydration pathways controlling the free tropospheric humidity.

In section 2 we present the IASI datasets and the different numerical weather prediction model re-analysis that we have used and we provide some guidance on the interpretation of δD-humidity variations. We analyse the seasonal and inter-annual δD variations observed ~~at Izana~~ above the CAR in sections 3 and 4, respectively. Then in section 5, we describe the spatial distribution of δD observed in July 2012. Finally, the results are discussed in the conclusion section.

2 Data and methods

2.1 IASI δD retrievals

This study is mainly based on H_2O and δD profiles derived from IASI radiances measurements ~~(Lacour et al., 2012)~~(Lacour et al., 2012, 2015) from the retrieval processor developed at ULB/LATMOS. IASI is a Fourier Transform spectrometer flying onboard the MetOp platform measuring the thermal infrared emission of the Earth and the atmosphere (Clerbaux et al., 2009). The high quality spectra (good spectral resolution -0.5cm^{-1}- and low radiometric noise) allow to retrieve information on H_2O and δD in the troposphere after an inversion procedure following the optimal estimation method (Rodgers, 2000) adapted for the requirements of δD retrieval (Worden et al., 2006; Schneider et al., 2006). With respect to supplying δD observations in the free troposphere, IASI is the unique successor of the Tropospheric Emission Spectrometer (TES) instrument which has been used in many isotopic applications studies (i.e. Worden et al. 2007; Risi et al. 2010, 2013). IASI with its high spatio temporal sampling (a measurement almost everywhere on the globe, twice a day) is of great interest for studies on short terms variations of δD (Bonne et al., 2015; Tuinenburg et al., 2015) and for an optimal sampling of δD natural variability (Lacour et al., 2015).

The δD profiles retrieved from IASI have limited information on the vertical, with degrees of freedom (dofs) varying between 1 and 2 depending on the local conditions (thermal contrast, temperature and humidity profiles, e.g. Pommier et al. 2014). In general, the maximum of sensitivity of the retrieval lies in the free troposphere between 3 and 6 km. For our analysis we use only the retrieved δD profiles that have more than 1.5 degrees of freedom and that yield a maximum of sensitivity between 4 and 6 km. ~~The retrievals are also filtered based on the residual of the fit.~~ It is also important to mention that the δD and humidity retrieved profiles are not exactly representative of the same atmosphere, the humidity profile having more vertical information than δD. It is thus important to keep in mind that when δD-q pairs are considered, the δD estimate is representative of a thicker layer than the q estimate. On an individual basis, the observational error on δD between 4 and 6 km has been estimated and cross-validated to 38‰ (Lacour et al., 2015). When several retrieved values are averaged (from N measurements), this error is reduced by a factor $\sqrt{N}$. Because of the large number of IASI measurements, there is presently no near-real-time processing of IASI radiances for δD. The availability of this quantity is thus limited. In this study we use three different datasets to investigate the isotopic characteristics of water vapour in the North Atlantic:

- a 5 year (2009-2013) dataset above ~~Izana~~ the CAR (26°N-30°N, 18°W-14°W) with an average of 65 measurements available per day;
- a 1 year dataset (2011) along a latitudinal band of narrow longitudinal extent (0°N-60°N, 30°W-25°W), which is used in section 3.4 to evaluate the variations of δD seasonality along the Western African coast;
- a 1 month dataset (July 2012) above the North Atlantic (0°N-40°N, 40°W-5°W) which is also used to illustrate the spatial extent of the influence of the SHL (subsection 3.4) and to analyse the different sources and processes controlling the humidity above the North Atlantic in section 5.

These data are used at different time scales from the individual observation to monthly averages. Daily means are obtained by averaging individual observations and monthly averages are obtained from the daily averages.

There exists another δD retrieval processor from IASI spectra (Schneider and Hase, 2011) developed within the MUSICA project (Schneider et al., 2016) which we do not use in this study, a brief summary of the differences of the processors is given in the Appendix of Schneider et al. (2016).

The MUSICA MetOp/IASI data have already been used previously for documenting the different moisture pathways in the subtropical North Atlantic region (Schneider et al., 2015).

2.2 TES δD retrieval

In order to derive a climatology of δD seasonality at a global scale, we also used δD profiles retrieved from the Tropospheric Emission Spectrometer (TES) measurements (Worden et al., 2012). The TES instrument is, like IASI, a thermal infrared sounder but with a better spectral resolution which makes it more sensitive to the lower troposphere. The observational error on δD retrieved values from TES has been evaluated to ± 30‰ (Worden et al., 2012; Herman et al., 2014). The spatiotemporal sampling is however lower than IASI. Nevertheless, δD retrievals from TES are available since September 2009 and allow for global analysis of δD.

2.3 Backward trajectories analyses and reanalysis data

To help in the interpretation of δD data we use backward trajectory calculations from the Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Stein et al., 2015) where NCEP GDAS (Global Data Assimilation System) re-analyses (Kleist et al., 2009) have been used as the meteorological fields. Backward trajectories have been mainly used in the analysis of the dataset above ~~Izana for which we computed a trajectory reaching the centre of the 4~~the CAR. Three trajectories arriving at 26°~~by 4~~N, 28°~~box for every day .~~N and 30°N at the longitudinal center of the box are computed for each day of IASI observation. In our analysis the trajectories we used are initialized from midday at altitude of 5.5 km. As the retrieved values of IASI are sensitive over a large layer of the true δD variations, we checked that the air trajectories arriving at 4.5 and 3.5 km were similar. This is shown in Appendix for year 2011. It is also shown that we do not expect temporal mismatch to significantly affect the air parcels history. We also used ECMWF (Dee et al., 2011) and MERRA (Rienecker et al., 2011) re-analysis data to characterize atmospheric dynamics.

2.4 Background on δD analysis

Variations in δD are to a first order tied to those in ~~specific humidity (~~absolute humidity (here we use water vapour mixing ratio, noted q). For this reason the interpretation of the information contained in δD is generally done in the δD-q space, which allows for a joint analysis of their variations. While the interpretation of δD-q pairs can be complicated as numerous processes can produce a same δD-q combination, simple models are helpful to understand their position in the δD-q space (Noone, 2012). The isotopic depletion of water vapour that undergoes condensation at equilibrium can be described by a Rayleigh distillation model as:

$$\delta D = (\alpha - 1)\ln\frac{q}{q_0} + \delta D_0; \qquad (1)$$

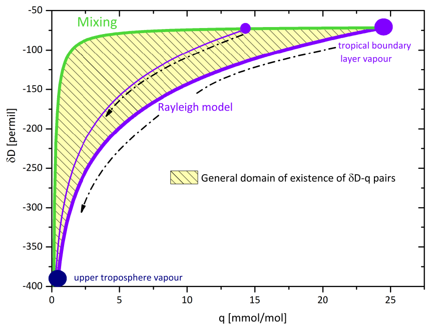

Figure 1. Simple models describing the domain of existence of δD-q pairs. The two purple curves describe the progressive depletion of a tropical boundary layer source and a drier one according to a Rayleigh distillation. The green curve describes the mixing between humidity from the tropical boundary layer source with humidity of the upper troposphere.

with q_0 and δD_0 are the specific humidity and the isotopic composition of the water vapour source, and α is the coefficient of fractionation. This model is shown in Figure 1 for two different sources of water vapour (purple lines). A Rayleigh model with a tropical water vapour source can generally be used to describe the lower limit of the domain of existence of δD-q pairs. The superior limit of this domain can be described by a mixing model between depleted and dry air from the upper troposphere that mixes with enriched and humid air from the tropical boundary layer (green line in Figure 1). The mixing between a source A and B produces a resulting air parcel of mixing ratio q which is the weighted average of the mixing ratio of the two air parcels:

$$q = f[H_2O]_A + (1-f)[H_2O]_B, \tag{2}$$

with f, the mixing fraction. The resulting ratio of isotopologues is given as (Galewsky and Hurley, 2010):

$$R_{\textbf{mix}} = \frac{f[HDO]_A + (1-f)[HDO]_B}{f[H_2O]_A + (1-f)[H_2O]_B}. \tag{3}$$

The mixing model is shown in green in Figure 1. Mixing and distillation of water vapour from different sources can occur over a wide range of combinations and produce δD-q pairs in between these two boundary models. Noteworthy, intense convective activity act to over deplete water vapour through rain-drop re-evaporation and δD-q pairs can be found below the Rayleigh distillation model (Worden et al., 2007).

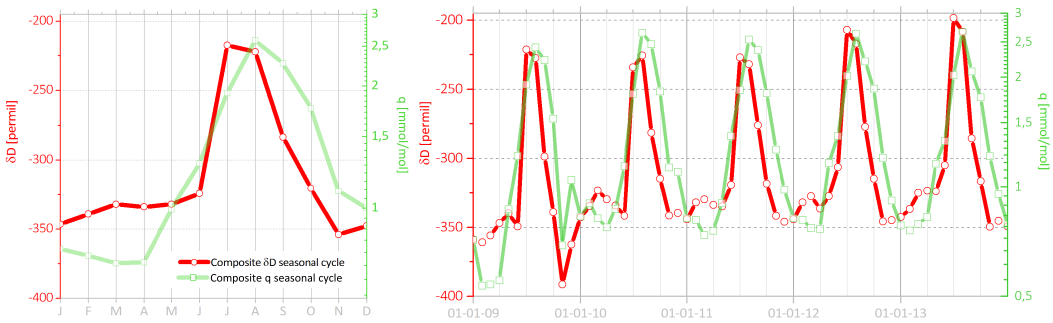

Figure 2. a) Composite seasonal cycles of δD (red) and specific humidity (green) above ~~Izana~~ the CAR for the 2009-2013 time period. b) Monthly variations of δD (red) and specific humidity (green) above ~~Izana~~ the CAR for the 2009-2013 time period

3 Seasonal variations: Influence of the SHL on δD in the subtropical North Atlantic

3.1 Seasonal cycle of water vapour and its isotopic composition over ~~Izana~~the CAR

Figure 2-a shows the composite (2009-2013) seasonal cycles of specific humidity and its isotopic composition at 5.5 km above ~~Izana~~the CAR. The troposphere is moistened from April to August as the large scale subsidence weakens due to the northward migration of the ITCZ. Then, it progressively dries as the ITCZ retreats south. Interestingly, the δD composition of water vapour does not follow the exact same cycle as humidity: the air masses are indeed progressively enriched (~~8~~5‰ per month) from January to June, before exhibiting an abrupt enrichment from June to July (~~140~~110‰ in one month). The enrichment persists throughout in August with values nearly as important as in July. Afterwards, the content in HDO rapidly decreases from August to November, by ∼~~150~~130‰while humidity stays high until October. This strong seasonality in δD values is particularly striking and exceeds the seasonality generally found elsewhere (see section 3.4). The period of enrichment in HDO over ~~Izana~~ the CAR appears to coincide with the period when the SHL is present over the Western Sahara as the climatological onset of the SHL occurs at the end of June as the SHL retreats toward the South at the end of September (Lavaysse et al., 2009). In the following, we conduct our analyses with the aim at understanding the factors controlling this strong seasonality and its link with the SHL. Figure 2 (right panel) also shows important inter-annual variability observed in δD signal not correlated to the humidity variability. We focus on the inter-annual variations in section 4.

3.2 The Saharan Heat Low and the associated atmospheric dynamics

In summer, strong heating of the Saharan surface creates a low pressure system (SHL) which enhances convergence in the low levels (see left panel of Figure 3 illustrating the low level circulation for July 2012). In the lower troposphere, the cyclonic circulation around the SHL strengthens simul-

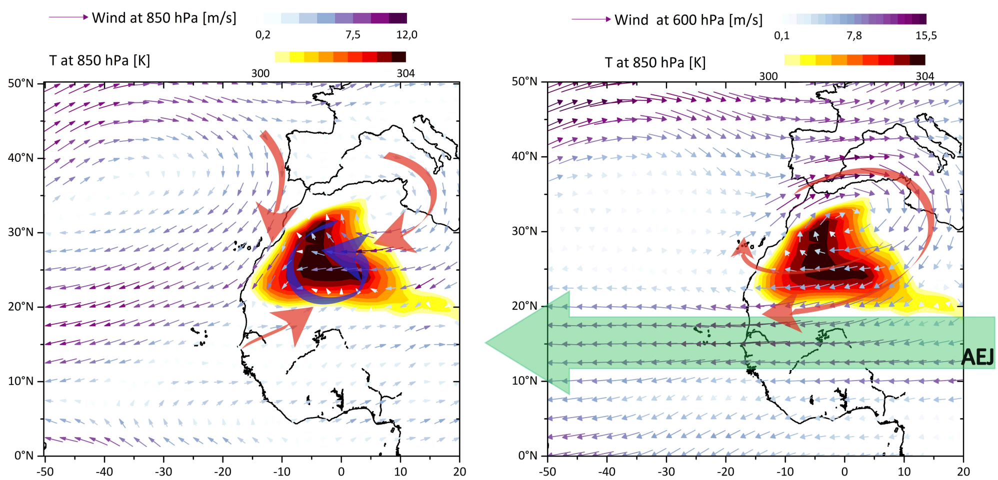

Figure 3. Low level (left panel) and mid-level (right panel) circulation associated with the SHL. The location of the SHL is indicated by the temperature field at 850 hPa above 300 K on both panels. The arrows represent wind fields at 850hPa (left panel) and 600hPa (right panel). On the left panel, the red arrows indicate the different sources converging in the depression and the blue arrow represents the anticyclonic circulation associated with the SHL. On the right panel, the red arrows shows how the anti-cyclonic circulation is divided into a part strengthening the AEJ.

taneously the south westerly monsoon flow east of its center and the north-easterly Harmattan flow to the west. The near-surface convergence generates enhanced rising motion in an environment prone to dry convection, leading to the formation of deep well mixed boundary layers, whose top often reach 600 hPa or higher. The divergent circulation at the top of the SHL generates an anticyclonic circulation in the middle troposphere, which contributes to the intensification of the African easterly jet (AEJ) further south (Thorncroft and Blackburn, 1999) and which is responsible for the horizontal transport of continental air masses above the Atlantic. The middle tropospheric circulation for July 2012 represented in Figure 3 shows how the anti-cyclonic circulation contributes to strengthening the AEJ and to bringing mid-level air masses from Northwest Africa over the Northeastern Atlantic.

The development of a heat low due to the surface heating has several consequences on the atmospheric dynamics. In the summer, ~~before the displacement of the West African Heat Low (WAHL) over the Western Sahara, the Saharan atmospheric boundary layer (SABL) depth is on the order of 5 km, which it one of the deepest ABL over the globe in summer. Once the~~ once the SHL settles in, the top of the ~~SABL can reach an even higher~~ Saharan air boundary layer (SABL) can reach altitude of 6 to 7 km (∼550 hPa) ~~during the months of July and August~~ due to the enhanced near-surface convergence in the SHL region. The SABL is fully developed in the late afternoon (Chaboureau et al., 2016). During most of the day, while the mixed layer is developing, the characteristics of air masses in the upper part of the SABL (the residual layer) are controlled by advection from the East

(Flamant et al., 2007, 2009; Chaboureau et al., 2016). As the warm and dry air moves off the African coast, the SABL rises and becomes the Saharan Air Layer (SAL) undercut by the cool and moist marine boundary layer.

The intensity of the SHL also has an influence on deep convection over the Sahel (Lavaysse et al., 2010) and hence on the precipitation over the area. Lavaysse et al. (2010) have indeed shown that during anomalously warm SHL phases (SHL more intense than on average) the southwesterly monsoon flow is reinforced over the Central and Eastern Sahel, leading to enhanced deep convection. At the same time, deep convection is reduced over the Western Sahel. Evan et al. (2015) and Lavaysse et al. (2016) have further shown that the SHL intensity has increased (due to the warming of the Sahara) in the 2000's with respect to the 1980's, explaining the increase of precipitation observed in Central and Western Sahara in the recent years (Panthou et al., 2014). At inter-annual scales, the intensity of the SHL can significantly modulate deep convection over the Sahel. Furthermore, the diverging anti-cyclonic circulation at the top of the SHL influences the intensity of the AEJ and the westward transport of moisture.

3.3 Relationship between the SHL and the summer enrichment over the Atlantic

To gain insights into the processes leading to the seasonal evolution of the isotopic composition of water vapor shown in Figure 2, we analyse the monthly δD-q diagrams from March to October 2012 in Figure 4-a using Rayleigh and mixing models ~~.~~ (the same models than in Figure 1 are shown for simplicity) and using all the IASI observations. The colour indicates the gradient of temperature computed between 5.5 and 1.5 km. In parallel, we provide monthly analyses of airmass trajectories reaching ~~Izana~~ the CAR from HYSPLITT backward trajectory analyses (Figure 4-b). Air masses arriving at ~~Izana~~ the CAR from lower altitudes are identified by dark to light orange lines while air masses coming from higher altitudes are identified by dark to light purple lines. The bottom panels of Figure 4 (Figure 4-c) show the monthly temperature at 850 hPa over the domain, this variable being a proxy of the SHL location over the Sahara.

The δD-q diagrams in the top panel of Figure 4 show a clear change in the repartition of the δD-q pairs from May to June. In May, air masses are characterized by low δD values and a wide range of specific humidity. These observations are localized close to or below the Rayleigh model or close to the dry end of the mixing model. The measurements below the Rayleigh model are associated with air masses coming from the tropics and from lower altitude and can thus be explained by convective processes (e.g. Worden et al. 2007). In June there is a clear separation of the measurements in two clusters, with the cluster of enriched air masses close to the mixing model and the cluster of depleted air masses located between the Rayleigh model and the mixing model. The two clusters also indicate a clear change in the temperature gradients highlighting two different atmosphere. In July, only the enriched cluster is seen with high temperature gradients; it is associated with larger specific humidity values. The same applies in August with slightly more depleted values. In September

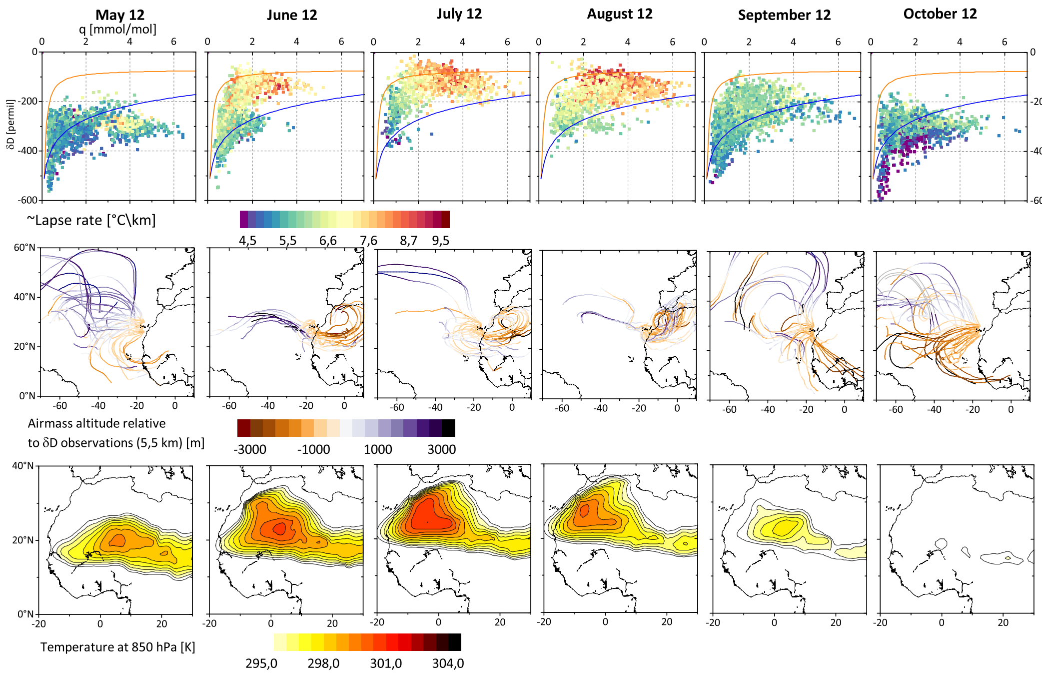

Figure 4. Top panels: δD and q ~~daily~~ variations from May to October 2012. All IASI individual observations within the box are shown. The mixing (orange line) and Rayleigh models (blue) are also shown. The temperature lapse rate is given by the colour scale. Middle panels: backward trajectory analyses of the air parcels arriving at ~~Izana~~ the CAR from May to October 2012. The altitude of the air parcels are indicated relatively to the observation altitude (5.5 km). Bottom panels: monthly temperatures at 850 hPa from May to October 2012.

and October, the observations are located at the left corner of the diagram (more depleted and less humid). The distribution of the δD-q observations in October 2012 is similar to that of May 2012. This situation then persists until the end of the year (not shown here). In summer, the position of the δD-q pairs along the mixing model suggest important mixing between dry and depleted air parcels with moist and enriched ones. Additionally, the fact that they are localized on the moist branch of the mixing model indicate a mixing for which the proportion of the moist term is quite important; this is surprising at an altitude of 5.5 km.

The backward trajectories shown in the middle panel of Figure 4 highlight that the shift in the cluster of δD-q pairs towards higher δD values (from -300 to -100 ‰) corresponds to a change in the origin of the airmass. During most of the year, the main origin of the air masses arriving at ~~Izana~~ the CAR is the upper troposphere above the Atlantic Ocean. Conversely, from June to August, the air masses come from lower altitudes and from a more localized area in the western Sahara. The situation in June, which is characterized by a HDO-enriched δD-q cluster and an HDO-depleted one, is particularly noteworthy and can be explained with the trajectories analysis: the depleted cluster would there be associated with air masses coming from the Atlantic and from higher altitudes while the enriched cluster would correspond to the air masses coming from the African continent and from lower altitudes. Such differences in the δD-q distributions due to different origins of air masses have already been reported from in situ measurements in González et al. (2016).

The bottom panels of Figure 4 indicate that the change in airmass trajectories is concomitant with the onset of the SHL, i.e. the installation of the SHL over the Western and central Sahara (Lavaysse et al., 2009). The concomitant shift of δD-q pairs clusters from a δD value of ∼-300 ‰ to a value of ∼-100 ‰, with the change of airmass trajectories and the installation of the SHL above the Western Africa shown here for the year 2012 is observed for the entire 2009-2013 period with some differences that are discussed in Section 4. Therefore, it appears that the isotopic signal in the water vapour above the Atlantic is strongly influenced by the development of the SHL.

The Figure 5 shows different diagnostics of the state of the atmosphere associated with different combinations of isotopic composition and water ~~vapour~~ vapor content. We plot in Figure 5-a the daily averages (not the individual observations as in Figure 4) δD-q variations for the period 2009-2013 and select four boxes defining contrasting conditions: dry and depleted (orange box), dry and enriched (cyan box), humid and depleted (purple box) and humid and enriched (green box). The green box corresponds to the situation found in July-August with air masses coming from the Sahara, the purple box corresponds to depleted values associated with air masses coming from the tropical Atlantic at the end of the summer while the cyan and orange boxes correspond to most of the daily values found during most of the year and are generally associated with air parcels coming from the North Atlantic. The number of days per month corresponding to each box is plotted in Figure 5-b. The average precipitation amount (computed from the re-analysis at a time step of 3 hours) along the backward trajectories is also shown in Figure 5-c. For each box we plot the composite profiles of the temperature (Figure 5-d), specific humidity (Figure 5-e), relative humidity (Figure 5-f), Richardson number (Figure 5-g)~~and vertical velocity (Figure 5-h)~~.

For all variables, the ones corresponding to the green and purple boxes strongly deviate from the majority of the observations (characterized by the blue and orange boxes where most of the δD-q pair lie). The humidity profiles corresponding to the purple box show high humidity (relative and specific) values, which strongly suggest convective processes at play, furthermore confirmed by the precipitations found along the backward trajectory (Figure 5-c). In the case of the enriched and humid values, corresponding to the summer enrichment (green box), the different profiles characterize a very particular atmosphere. The specific humidity profile has very high values close to the surface, which rapidly decrease with altitude; the relative humidity profile shows also very high values in the first layer of the atmosphere than very dry values up to around 550 hPa; the temperature profile presents an inversion layer followed by a warm layer; the Richardson number profile indicates very stable values within a thick layer between 900 to 550 hPa. All these features are in fact characteristic of the thermodynamical structure of a deep Saharan air layer (SAL) moving above a thin marine boundary layer. In Figure 6 we show the development of the deep SABL during the year. One can

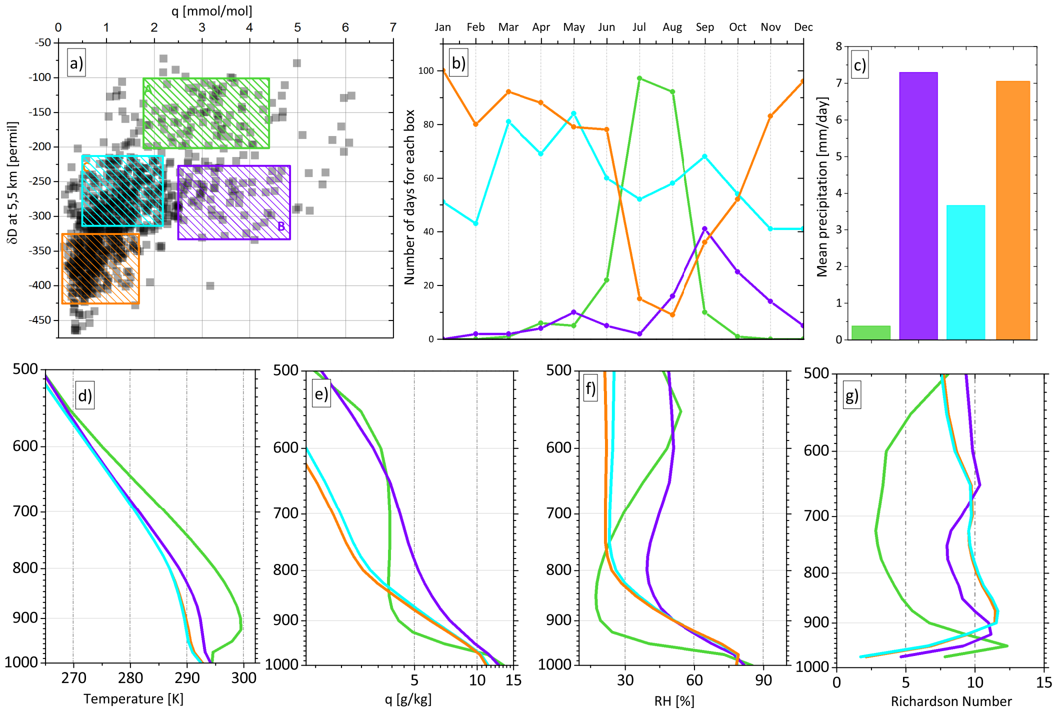

Figure 5. Dynamical characterization of the δD-q space. a) Daily variations of δD and q for the entire 2009-2013 period with different boxes where composite profiles have been derived from different geophysical parameters (see text for details). b) Number of days per month found within each box, c) mean precipitation along the backward trajectories, panels d) to h) show composite profiles of temperature (K), specific humidity (g/kg), relative humidity, richardson number ~~, and vertical velocity~~(from MERRA re-analysis).

see that the height of the boundary layer reaches the altitude of IASI sensitivity from June to the end of August. It is however only by the end of June when the air masses originating from the upper part of the ABL above the Sahara are transported above the Atlantic that the δD signal above ~~Izana~~ the CAR rapidly increases. The seasonality of δD observed in the free troposphere above ~~Izana~~ the CAR can thus be explained by the development of deep ABL associated with the settling of the SHL over the Western Sahara. The latter acts to efficiently mix dry upper tropospheric air with air from the boundary layer.

Noteworthy, the four boxes delimiting the different δD-q signatures (here defined arbitrarily) are very similar to the dissociation of in situ δD-q measurements by González et al. (2016) based on the temperature of the last condensation of air parcels.

3.4 Spatial extent of the SHL influence

As the high seasonality in the water isotopic composition observed at ~~Izana~~ the CAR is closely associated with the activity of the SHL, it can serve as a diagnostic to evaluate the spatial influence

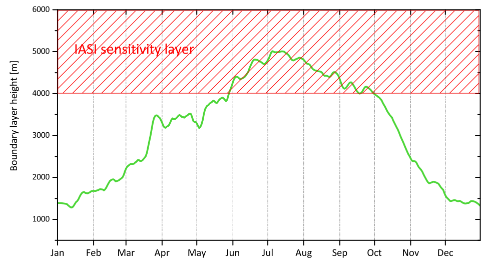

Figure 6. Development of deep boundary layers during summer above the Sahara (7°W-5°E,20°N-30°N). Boundary layer heights are extracted from ECMWF ERA re-analysis and are averaged from 2009 to 2013.

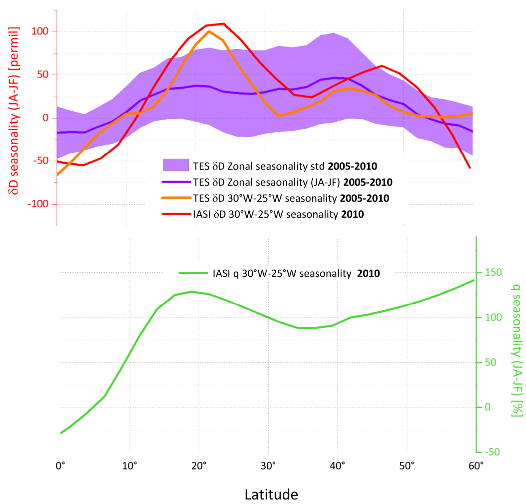

Figure 7. Top panel: Global δD zonal seasonality (JA-JF) from 0 to 60°N (purple line) with its associated standard deviation (shaded area) and seasonality found offshore of West Africa (30°W-25°W,0-60°N) (orange line) observed by TES for the 2005-2010 period ~~. Middle panel: same~~ and as ~~the top panel but where~~ observed by IASI seasonality ~~(30°W-25°W,0-60°N) is shown~~ for 2010 (red line). Bottom panel: IASI specific humidity seasonality.

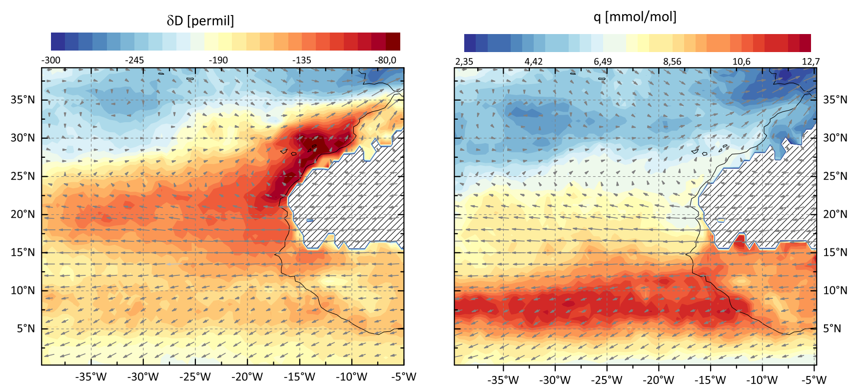

Figure 8. δD (left panel) and q (right panel) distributions from IASI at 4.5 km for July 2012 together with the averaged wind fields at 600 hPa.

of the SHL. In Figure 7-a we first present the seasonality in δD signal (defined as $\bar{\delta}\mathrm{D}_{\mathrm{July-August}}$ - $\bar{\delta}\mathrm{D}_{\mathrm{January-February}}$) observed from the TES instrument. We use the TES δD data here as they are available over a longer time period than the IASI dataset considered here and at a global scale. We plot the seasonality of δD as a zonal mean with its associated standard deviation, calculated for the 2005-2010 period. The seasonality observed off the Western African coast, on an entire latitudinal band of narrow longitudinal extent (30°W-25°W), is also drawn in orange. The latter exhibits a sharp maximum around 22°N, which exceeds values found globally. We attribute this high seasonality as the result of the SHL activity and therefore suggest its influence on the isotopic budget of water vapor extends over a large part of the North Atlantic. Figure 7-b shows the same as Figure 7-a but for IASI data in 2010. These also show the enrichment in July August from 15° to 30 °. The bottom panel of Figure 7, which shows the seasonality for the specific humidity (in percent), reveals a different behaviour~~and this~~. The observed maximum in δD which does not correspond to the maximum of humidity can also be interpreted as the signature of the SHL, as mixing processes produce a stronger isotopic signal for a given specific humidity than any other hydrological processes (Galewsky and Hurley, 2010; Noone, 2012).

The differences between humidity and δD are also clearly visible in the spatial distributions of δD and q for July 2012 shown in Figure 8. The water vapour distribution strongly differs from its isotopic composition as the maximum in Deuterium enrichment does not appear along the ITCZ, where high δD values are generally associated with high humidity in convective areas (Risi et al., 2012), where convection act to bring enriched air masses at higher altitudes. Instead, we find the maximum of enrichment further North, around 20°N at the northern edge of the AEJ, for a wide range of specific humidity values.

The spatial pattern drawn by the high seasonality of δD can be linked to the dynamics of the SAL. Tsamalis et al. (2013) have shown that the SAL displays clear seasonal cycle (both in latitudinal

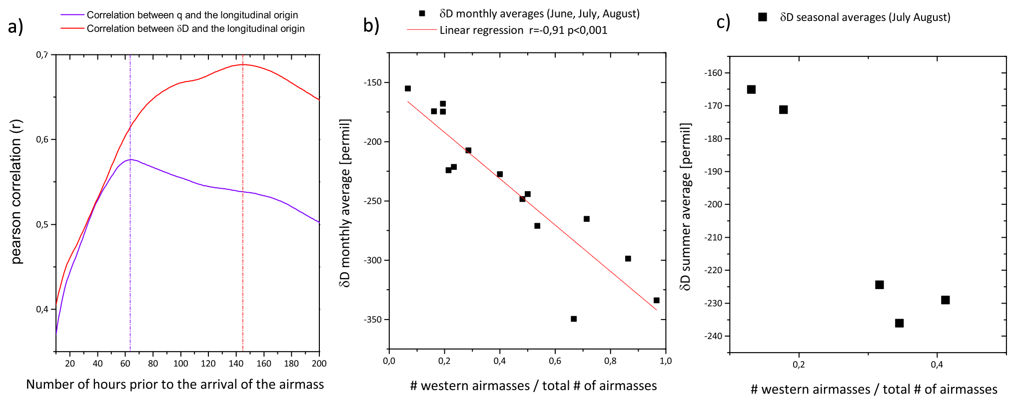

Figure 9. a) Correlation between δD (red curve) and q (purple curve) daily variations and the longitudinal origin of the airmass at various time steps (number of hours prior the arrival of the airmass at ~~Izana~~the CAR). b) δD monthly averages (June, July and August) as a function of the ratio of the number of air masses arriving from the Atlantic (Longitude<-20°W) ~~and~~ to the total number of ~~air masses arriving from the African continent~~air-masses. c) Same as b) but for summer averages (July, August).

extent and in vertical structure), using 5 years of data from the space-borne Cloud-Aerosol LIdar with Orthogonal Polarization (Winker et al., 2010). The SAL occurs at higher altitudes and farther north during the summer than during winter. Near the African coastline, the SAL is found between 5°-30°N in summer, its northern edge being observed just north of the Canary Islands. The northern edge of the SAL migrates to 15°N during the winter, and is generally observed to be south of the Canary Islands from September to May (see Figure 2 of Tsamalis et al. 2013). During the summer, the SAL is found to be thicker and higher off the coast of Africa, between 1 and 5 km above mean sea level, while it is observed between 1 and 3 km during the winter (see Figure 4 of Tsamalis et al. 2013), i.e. below the altitude of maximum sensitivity of IASI-derived δD products.

4 Inter-annual variations above ~~Izana~~the CAR

Figure 2 shows that there is significant inter-annual variations observed in δD signal at ~~Izana~~the CAR. In this section, we investigate the reasons that could explain this variability.

4.1 Control of the zonal transport

As explained previously, the δD variations are sensitive to the source of water vapour. During this time, the air masses reaching ~~Izana~~ the CAR have contrasting isotopic signatures and water vapour content: the air masses from the Atlantic are dry and depleted, while conversely, the air masses from the African continent are wet and enriched. Schneider et al. (2015) already documented

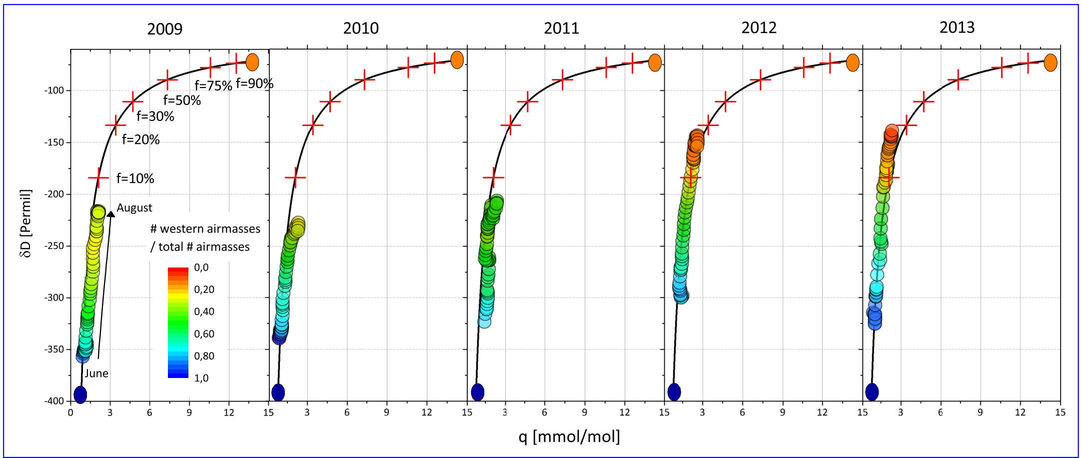

Figure 10. Summer enrichments (from June to August) observed at ~~Izana~~ the CAR from 2009 to 2013 along a mixing model defined by the mixing between upper tropospheric water vapor (UT) and boundary layer vapour above the Mediterranean (MBL). IASI daily observations at 5.5 km are smoothed on a 30 days moving average filter and the corresponding ratios of western air-masses to total number of air-masses is shown in colour. The 10, 50, 75 and 90% fractions (f) of MBL of equations 2 and 3 are indicated with the red crosses.

the link between high δD values and the continental origin of the airmass by coinciding observations of dust concentrations. Thus, the origin of the air masses must control δD.

In this section we translate the control of ~~western to eastern~~ the airmass origins in terms of mixing fraction of the mixing. Indeed, assuming that the monthly average isotopic composition at ~~Izana~~ the CAR in summer is the result of mixing between upper tropospheric air and boundary layer air, the control of the ratio of the number of the western air-masses to the eastern airmasses can be understood in terms of the mixing fraction of the humid source (f in equationvery similar values and only the specific humidity varies depending on the latitudinal position of the sea. Hence the humid term of the mixing model (in orange in Figure11) could be associated with tropical boundary layer water vapor or boundary layer water vapor from the Mediterranean region (drier) both being potential sources of water vapor fed into the SHL (so called SHL ventilation from south or north, respectively, ~~Lavaysse et al. 2009~~Lavaysse et al. (2009)). This means that moisture transported at low-level from the oceans and the seas surrounding the continent towards the SHL contribute to the moistening of the free troposphere over the Northeast Atlantic. The SHL is a key player in this process as the relatively moist and enriched air masses are mixed vertically over the depth of the Saharan ABL before being transported over the Ocean due to the divergent, anticyclonic circulation at the top of the SHL. In Figure 10, the colours indicating the ratio of the number of western air-masses to the total number of air-masses show that from June to August, as δD increases along the mixing model, the ratio progressively decreases. Assuming constant dry and humid terms, this displacement along the mixing

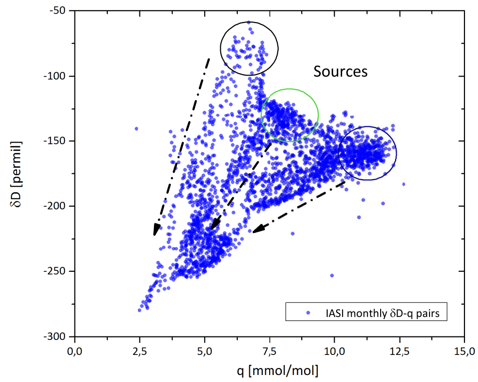

Figure 11. δD-q monthly averages obtained from IASI for July 2012 (see also Figure 8). Each point represents one location over the North Atlantic. Three sources (circles) and different dehydration pathways (black arrows) are identified. See text for details.

model can be explained by an increase of the mixing fraction (f) in equations 2 and 3. The ratio of the number of western air-masses to the total number of air-masses acts thus like the mixing fraction in controlling the δD composition of water vapour. The magnitude of the enrichment is important because in the dry member of the mixing model, a small increase of the fraction of boundary layer air acts to significantly enrich the resulting mixed air-mass (mixing fractions are indicated by the red crosses in Figure 10), conversely, the specific humidity increase is small. The summer enrichment observed at ~~Izana~~ the CAR can therefore be interpreted as the progressive increase of the boundary layer air fraction in the mixing as the SHL acts to efficiently blend boundary layer air over increasing depths from June to August, bringing moisture to altitudes where only upper tropospheric air is observed during the rest of the year. Figure 10 also nicely shows the inter-annual variations observed from 2009 to ~~2103~~ 2013 as the main origin of the air-masses varies. Assuming constant end members of the mixing, the observations suggest that f reaches up to 20% in 2013 while it is below 10% from 2009 to 2011.

5 Dehydration pathways in the North Atlantic

Finally, we have analysed the information contained in the isotopic composition of water vapour over the North Atlantic for July 2012. In Figure 11, we present the δD and q distributions previously shown in Figure 8 in the δD-q space. The IASI δD-q pairs occupy very distinct domains in the diagram indicating a wide variety of sources and processes controlling the water vapour above the Atlantic. In this section we aim at disentangling these various sources and processes. To do this, we map geographically the different sources and pathways identified and use simple models describing the isotopic depletion of water vapour (Noone, 2012).

5.1 Sources

The δD-q pairs draw distinctive pathways (Figure 11) corresponding to the progressive dehydration/moistening, depletion/enrichment, from different sources. The latter ~~can easily be distinguished by identifying the humid~~ are identified as the moist members of the different ~~visible pathways~~branches visually identified of the δD-q pairs scatter plot. Three different sources of water vapour have been identified according to their positions in the δD-q diagrams. These sources are shown in Figure 12-a and mapped on the spatial distribution of δD in Figure 12-b.

- S1: This source (black squares) is the most enriched and the driest one. It corresponds to water vapour found in a localized area close to the African coast around 26°N and 15°W. We suggest that this enriched source is the direct result of the SHL activity over Sahara. The dry convective mixing of water vapour from the Mediterranean sea with very dry and depleted air from the upper troposphere - as shown with the mixing model in orange - could indeed produce such high enrichment. Note that the humid term could also be more humid but that this would not affect significantly the mixing model.

- S2: This source (blue diamonds) is the most depleted and the wettest one. It corresponds to water vapour found along the ITCZ between 5° and 10°N. This strongly suggests that this source is the result of the convective uplift of water vapour (with condensation) from lower altitudes. In that case, a Rayleigh model describing the depletion of water vapour evaporated from the tropical ocean can explain the δD-q values observed (Figure 12-a, purple line).

- S3: This source with intermediate δD and humidity contents corresponds to a thin longitudinal band along 20°N. This correspond to the Westward transport of dust and aerosol from Africa along the Northern border of the AEJ, where the AEJ is strengthened by the SHL anti-cyclonic circulation (see Figure 3). Comparatively to S1, if we assume that this source is also produced by mixing and that the humid term is similar in both cases, this would mean that the dry term must be different than the mixing potentially explaining S1. As seen in Figure 12-b, the mixing between MBL air that has distilled from tropical water vapor could explain the position of S3 in the δD-q diagram. We thus hypothesise that S3 is the result of mixing between ascending air from the Sahel (MBL) and air from the AEJ which could be the result of a simple Rayleigh distillation of TBL. Noteworthy, as the S3 exhibit constant δD and humidity values along the northern border of the AEJ, it means that there is no dehydration and no depletion along this longitudinal band, and thus weak mixing along the westward transport above the Atlantic.

In summary, the air masses that circulated within the divergent flow at the top of the SHL show here two different isotopic signatures depending on whether they contribute to strengthen the AEJ over the continent or if they are transported anticyclonically over the Atlantic around the SHL. These two signatures are distinct from the signature of convection found along the ITCZ.

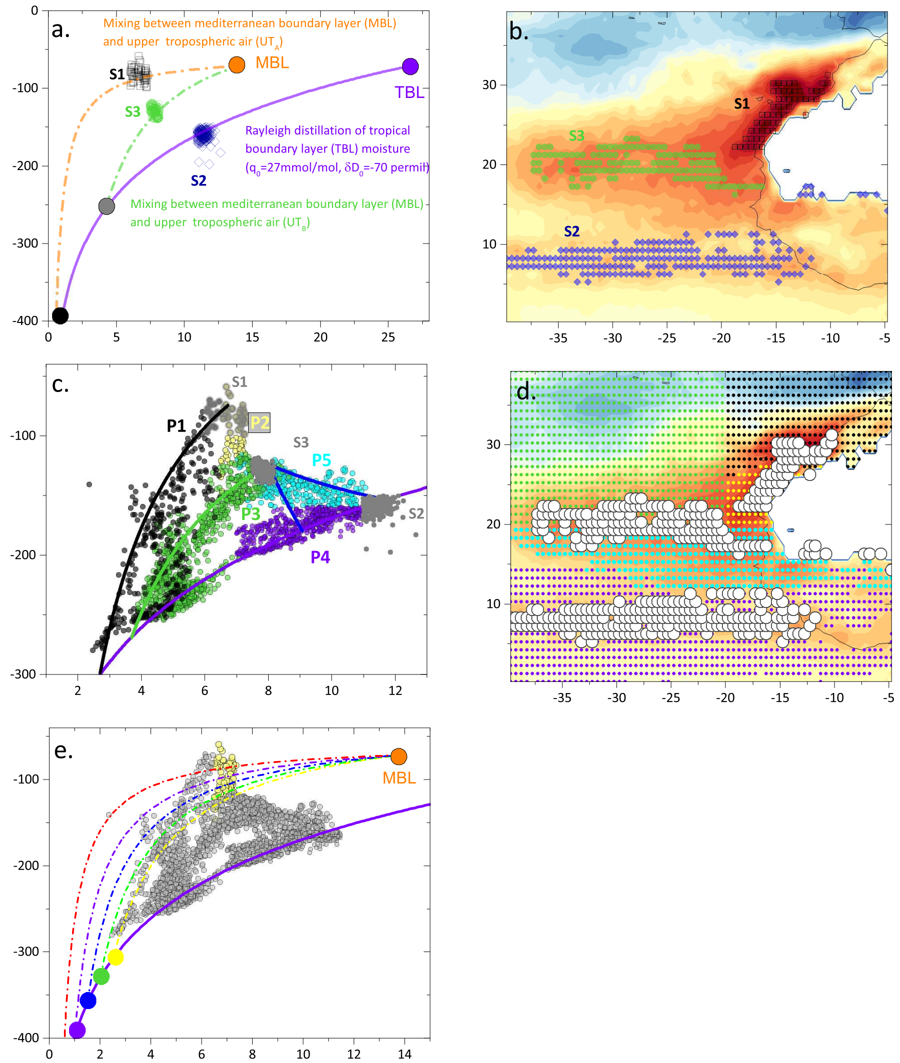

Figure 12. a) δD-q composition of the three main sources identified in the δD-q diagram with a mixing model (orange curve) between upper tropospheric air (black circle) and Meditteranean boundary layer vapor (orange circle); a Rayleigh model from a tropical boundary layer vapor (purple circle); and a mixing model between MBL and water vapor distilled according to the Rayleigh model defined. b) Geographical location of the differents sources identified in a). c) All δD-q pairs for July 2012 together with different mixing models that could explain the variations observed (see text for details), the colours indicate the different pathways identified. The sources identified in a) are shown in grey. d) Geographical location of the different pathways identified in c). e) Illustration of the mechanism explaining the variations observed in yellow (P2): mixing models between a constant humid term (MBL) and a dry term more and more enriched.

5.2 Pathways

Now that the sources have been identified, we analyse the different moistening and dehydrating pathways visible in the δD-q space from the different sources (Figure 12-c). ~~The~~ To dissociate the different pathways we use their position in δD-q space and we also use their geographical position to facilitate the dissociation in both spaces. Note that the sources show quasi constant δD and q values ~~. The~~ while the dehydration pathways, on the other hand, ~~as they~~ show an important variability ~~suggest that the processes responsible of the variations are horizontal~~. The dehydration and depletion is mainly latitudinal.

- P1: This pathway describes the dehydration and the rapid depletion of S1. Figure 12-d shows that this pathway corresponds to the area in the North of Africa, dominated by wind (see in Figures 8 and 3) towards Europe where the air is particularly dry and depleted. A mixing model between S1 and a dry term can be used to describe this pathway.

- P2: This pathway corresponds to a small area in between the Canary Islands (S1) and S3. Coherently, in the δD-q space, these points lie also between S1 and S3. The mixing between S1 and S3 can not explain the observations as a mixing model between these 2 terms would produce a hyperbolic line without reproducing the slope observed. Instead these observations could be explained by the mixing of a constant source such as the MBL with air becoming more enriched and more humid. This could be explained by a stronger influence of the African easterly jet and a weaker influence of the subsidence as we get closer to S3. The Figure ~~11~~ 12-e shows how this mechanism could explain the δD-q pairs observed.

- P3: This pathway corresponds to the western part of the Northern Atlantic where S3 is probably mixed with air from the large scale subsidence as the end member of the mixing model corresponds to the location of the Azores high.

- P4: This pathway describes the depletion of S2 northward and southward of the ITCZ and can be described by a simple Rayleigh distillation model which has the characteristics of the tropical boundary layer moisture.

- P5: This pathway is found between S3 and S2 or corresponds to distilled air parcels from S2. The position of the observations in the δ-q space could be explained again by mixing. However, in that case, the enriched source is drier than the humid source and the corresponding mixing model presents an inverse curvature. The blue curves showing the mixing models computed from S3 and S2 or more distilled terms allows to explain the scatter observed.

The analysis proposed here suggest that the combined observation of water vapor and its isotopic composition can be very useful to identify the different sources of humidity, which are key actors of the hydrological and dynamical cycle of the region, and their interactions. It is however impossible to unambiguously assess the processes responsible of the position of δD-q pairs as combination of different processes can lead to a same δD-q position. Nevertheless, the coherence of our interpretation with the actual understanding of the SHL dynamic suggests that the interpretation is reasonable.

6 Conclusions

In this study we have explored δD-q distributions derived from the IASI sounder above the North Atlantic for different time and space scales with the objective of providing an interpretation on the controls of δD in that region. We have shown that the seasonal enrichment of δD observed at ~~Izana~~ the CAR was closely linked to the installation of the SHL above the Sahara from June to August. By the end of June, the intense surface heating during the summertime period generates deep boundary layers, which can then be transported above the Atlantic within the so-called SAL. The SAL top reaching the altitude of IASI sensitivity, HDO-enrichment is observed over ~~Izana~~the CAR. We have shown that the influence of the SHL expands far off the coast, suggesting a large influence of the SHL on the isotopic budget and thus on the humidity budget. The summertime δ-q distributions at ~~Izana~~ the CAR are mainly the result of mixing processes between dry and depleted upper tropospheric air with humid and enriched boundary layer air from the oceans and seas surrounding the West African continent. In the summer, the SHL acts to efficiently mix these contrasting sources and transport anomalously moist and enriched air masses (when compared to the rest of the year) over the Northeast Atlantic Ocean. Inter-annual variations of δD were also interpreted as the differences in the fraction of western to eastern air-masses arriving at ~~Izana~~the CAR. The combination of δD and q observations from IASI in July 2012, together with the knowledge of the key components of the West African Monsoon system, allowed interpreting the variety of processes driving the water budget over the Northeast Atlantic. More generally this analysis demonstrates the usefulness of δD measurements from IASI as we show it is possible to disentangle the respective contribution of the different sources of water vapor together with their respective interactions.

The demonstrated capabilities of IASI to provide unique observational constraints on the different sources and processes controlling the free tropospheric humidity in the North Atlantic would be useful to evaluate the representation of these sources and processes in isotopes-enabled climate models. In particular, the strong isotopic signature associated with the SHL and its interactions with the monsoon and the AEJ could be used to assess the correctness of its representation in climate models.

Appendix A: Supplemental backward trajectory analyses

In this appendix we show additional trajectory analyses we did to verify that there is no spatial and temporal mismatches in the trajectories due to the large sensitivity layer of IASI and due to the differences of time sampling.

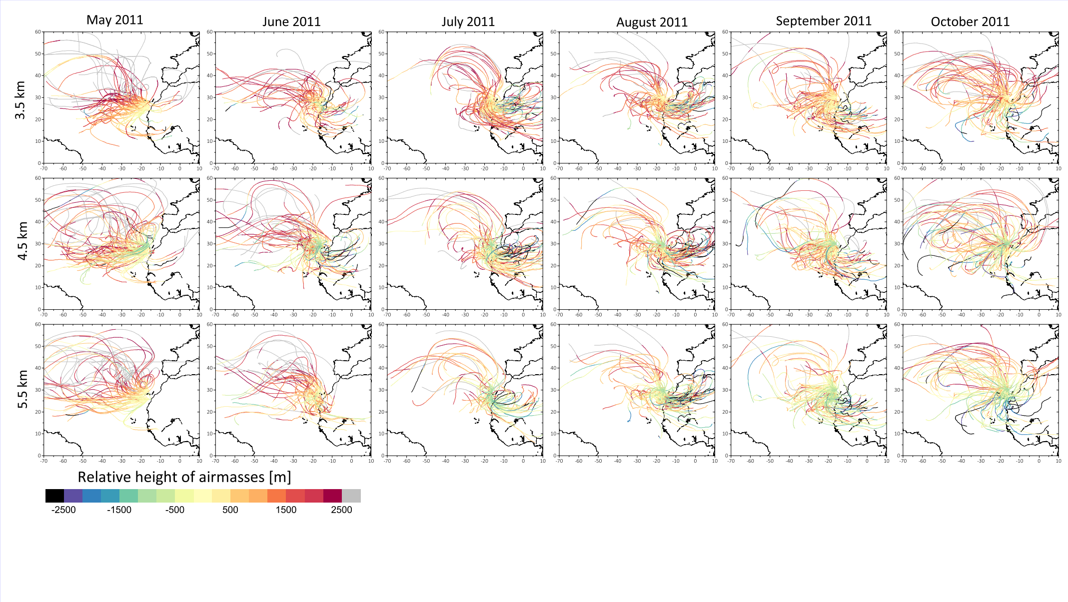

Figure A1. Airmasses arriving above the CAR at three different altitudes (3.5, 4.5 and 5.5 km). The trajectories are initialized from 3 points (26°,28° and 30°N and at the longitudinal center of the box at 12.00).

A1 Coherence of airmass trajectories between 3 and 6 kilometers

IASI δD retrievals are sensitive to δD variations over a large vertical layer. The information mostly comes from the free troposphere between 3 and 6 km. In Figure A2 we show air-masses arriving at different altitudes (3.5, 4.5 and 5.5 km) within this layer for the year 2011. Airmasses arriving at the different altitudes show similar patterns indicating that what we show at 5.5 km is also valid for the 3-6 km layer.

A2 Temporal mismatches

Here, we show additional backward trajectory analysis corresponding to different initialization times (9,12 and 21 UTC). The air parcels show very coherent trajectory in between 9 UTC to 21 UTC.

Author contributions. J.-L. Lacour did the retrievals of δD from IASI spectra, performed the data analysis and prepared the manuscript. C. Flamant provided expertise on the SHL and prepared the manuscript with J.-L. Lacour. Camille Risi provided her expertise on the analysis and has corrected the manuscript. P.-F. Coheur has supervised the first part of this study, in relation to the δD retrievals from IASI. He has corrected the manuscript. C. Clerbaux has supervised the second part of this study and has corrected the manuscript.

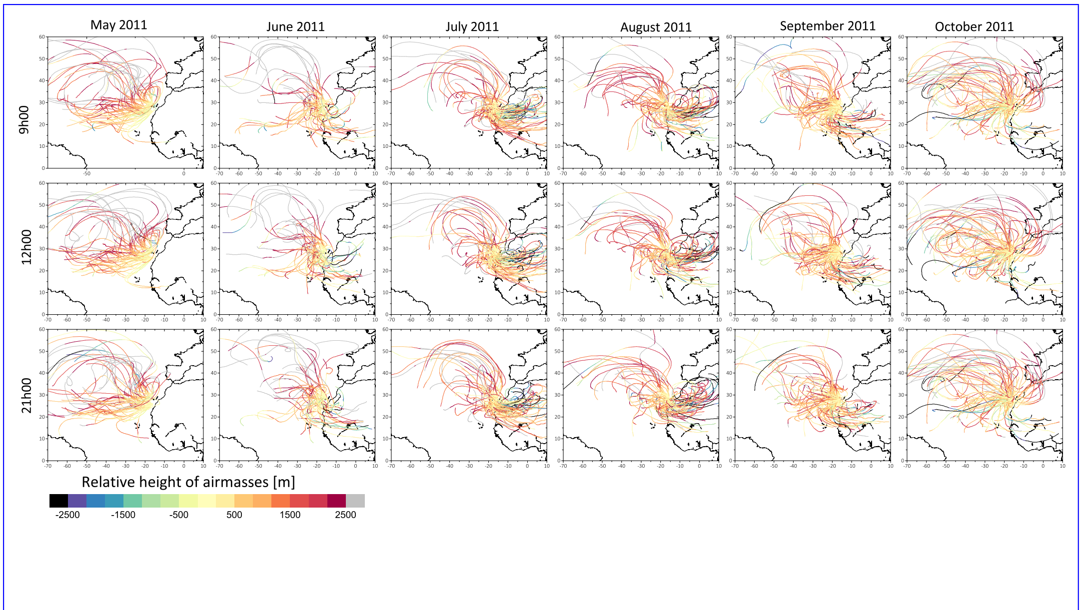

Figure A2. Airmasses arriving above the CAR (4.5 km) at three different times (9, 12 and 21 UTC). The trajectories are initialized from 3 points (26°,28° and 30°N and at the longitudinal center of the box).

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