Dear Prof Farahnaz Khosrawi, dear reviewers

We thank the editor very much as well as the two reviewers for their very detailed and helpful comments. We have addressed all comments and are submitting a manuscript taking into account all comments as explained in the answers to comment below.

Many thanks again for your help and work on this manuscript,

Amaëlle Landais and Cécile Agosta on the behalf of all co-authors

Editor comments on egusphere-2023-617

General comments:

The quality of this draft is above standard and the subject is suitable for publication in ACP. However, I think it may not be easy for every reader to understand this manuscript. Especially for those who are not experts on water isotope processes. I recommend major revisions of the manuscript so that the contents and results of this study become better understandable for any potential reader. Further, the graphical presentation is rather of low quality. Most of the figures should be improved (increasing the size of the figure itself as well as the font size and the line thickness). Some of the rather important figures of this study are in the supplement and may be moved to the main text or in an appendix to the main text (so that these do not appear in an extra document).

>> Many thanks for this suggestion. Indeed, the idea of an appendix to present the figures is a very good one and we now use it preferentially than the supplementary material for some figures. We also include the figures for the back trajectories in the new manuscript and did an effort to better explain some aspects of the paper following your comments.

The usage of terms etc. should be done more consistently one way or the other. In some occasions the term "vapor mixing ratio", "mixing ratio", "water mixing ratio" or "water vapor mixing ratio" are used making reading this manuscript very confusing. Then the notation "v" as subscript is used in some occasions, but in many others not. I would suggest to use "v" and "p" to differentiate between vapor and precipitation water vapor and water vapor isotopes throughout the manuscript.

>> This is indeed again a good suggestion and we will use the p and v subscripts and use the common term « water vapor mixing ratio » in the new version of the manuscript.

Specific comments

Abstract: The abstract is quite confusing and some transitional sentences, e.g. the first and second paragraph as well as a sentence stating that you use two models for comparison are missing. What is the main focus of your study ? The two paragraphs feel like two independent abstracts. One describing the measurements and the conclusions you derive from these and then the measurement-model comparison and the according results of this part of the study. The problem with the focus of study continues throughout the entire study. Additionally, several technical issues in the abstract need to be corrected (see below the list of technical corrections). Since there were so many issues I provide you here a corrected/improved version of your manuscript as a suggestion how it would read much better:

>> Many thanks for your suggestion. We propose the following abstract:

" In order to complement the picture of the atmospheric water cycle in the Southern Ocean, we have continuously monitored water vapor isotopes since January 2020 on Amsterdam Island in the Indian Ocean. We present here the first 2-year-long water vapor isotopic record on this site. We show that the vapor water isotopic composition largely follows the water vapor mixing ratio, as expected in marine boundary layers. However, we detect 11 periods of a few days where there is a strong loss of correlation between water vapor $\delta^{18}O$ and water vapor mixing ratio. These periods are associated with abrupt negative excursions of water vapor $\delta^{18}O$, often occurring toward the end of precipitation events. Six of these events show also a decrease in gaseous elemental mercury suggesting subsidence of air from higher altitude.

Our study aims at further exploring the mechanism driving these negative excursions in water vapor δ^{18} O. We used two different general atmospheric circulation models to provide a datamodel comparison over this 2-year. While the European Centre Hamburg model (ECHAM6wiso) at 0.9° is able to reproduce most of the sharp negative water vapor δ^{18} O excursions, hence validating the physics process and isotopic implementation in this model, the Laboratoire de Météorologie Dynamique Zoom model (LMDZ-iso) at 2° (3°) resolution only reproduces 7 (1) of the negative excursions highlighting the possible influence of model's resolution for the study of such abrupt isotopic events. Based on our detailed model-data comparison, we conclude that rain-vapor interactions associated with subsidence at the rear of a precipitation event are the most plausible explanations for such isotopic excursions. "

>> In this manuscript, we mainly look for controls on negative isotopic excursions. This is done by using models to unravel processes and while suggesting a mechanism, we could also point on limitations of the performances of some of them, which may support future use of mesoscale isotope models to go further. We propose to add a sentence making the link between atmospheric components of Earth system models and water isotopes and introducing in l. 61 the importance of the implementation of water isotopes in the atmospheric components of atmospheric general circulation models and to confront them with the models as:

"They can also be used as additional tools to test the performance of atmospheric components of some Earth system models in which water isotopes have been added (Risi et al., 2010; Schmidt et al., 2005; Werner et al., 2011). «

P5, L113-114: The Dumont d'Urville and Concordia stations in Antarctica are not really in the Indian sector, but rather in the Pacific sector. Thus, how these are suitable for understanding the atmospheric water cycle over the Indian basin of the Southern Ocean does not become clear.

>> We now make a more broader statement about South Indian Ocean being a significant moisture source for Antarctic precipitation, notably in the region encompassing Dumont d'Urville and Concordia stations. We add references to this statement, including Wang et al. (2020) and Jullien et al. (2020) (see figures R1 and R2 below),

P2, L51: Also in the introduction still the question remains what the purpose of your study is. Is it to confirm/better understand the measurements or to test the capability of the isotope enabled models to reproduce the isotopic processes?

References :

Wang, H., Fyke, J. G., Lenaerts, J. T. M., Nusbaumer, J. M., Singh, H., Noone, D., Rasch, P. J., and Zhang, R.: Influence of sea-ice anomalies on Antarctic precipitation using source attribution in the Community Earth System Model, The Cryosphere, 14, 429–444, https://doi.org/10.5194/tc-14-429-2020, 2020.

Jullien, N., Vignon, É., Sprenger, M., Aemisegger, F., and Berne, A.: Synoptic conditions and atmospheric moisture pathways associated with virga and precipitation over coastal Adélie Land in Antarctica, The Cryosphere, 14, 1685–1702, https://doi.org/10.5194/tc-14-1685-2020, 2020.



<u>Figure R1</u> : Spatial distribution of fractional contribution (%) to annual mean precipitation at the surface (right) from individual source regions in the mean case (left). Left panel is taken from Wang et al. 2020, Fig. 2 ; right panel is taken from Wang et al. 2020, Fig. 6



<u>Figure R2</u>: Composite maps of moisture uptakes for precipitating air parcels over Dumont d'Urville from Jullien et al. (2020) Fig. 10.

We add the following information in the revised version:

"Amsterdam Island is the only oceanic observatory dedicated to atmospheric studies in the southern hemisphere. This region in the Southern Indian Ocean is a significant moisture source for Antarctic precipitation, notably in the region encompassing Dumont d'Urville and Concordia stations (Wang et al., 2020; Jullien et al., 2020) " Figure 1 caption: What is meant with Magand? Add a link or reference? >> We explain better with "photo taken by O. Magand"

P8, L223: What do you mean here with "low altitude"? That the observatory is located at low altitude? Or that the air from higher altitude is transported down to lower altitudes?

>> Indeed, this sentence if not clear. We propose to modify as:

"The identification of such observational processes (lower GEM concentrations in high-altitude air masses versus marine boundary layer ones) is used here to help indentifying possible intrusions of high-altitude air masses down to the surface in Pointe Benedicte Observatory."

P8, L228: Here now subscripts "v" used, but before not.

>> Actually, when we were writing either "water vapor δ^{18} O" or simply " δ^{18} O_v" all along the manuscript but we will simplify and put systematic δ^{18} O_v, q_v and δ D_v everywhere.

P9, Figure 2 caption: Why is here the anomaly used?

>> This is the standard way to correct the Picarro $\delta^{18}O_v$ and δD_v measurements as detailed in the references given in the manuscript « (Tremoy et al., 2011; Leroy-Dos Santos et al., 2020)".

P9, L247: What exactly are these standards and how are these derived? Not clear! Are these typical relationships between these species? Are these documented somewhere else?

>> This is very classical when using isotopic composition of water vapor but we agree that it is difficult to follow when not working with this tool. We thus propose to explain better with the modified sentences:

"Here, we introduced two different water standards, EPB-AMS obtained from tap water at LSCE and GREEN-AMS obtained from melting Greenland surface snow. Theses standards are calibrated versus international water standards provided by IAEA. The water isotopic composition of these two standards lie on the global meteoric water line (Craig, 1961): they have respective values of (-5.66 ‰, -47.31 ‰) and (-32.65 ‰, -263.76 ‰) for the couple (δ^{18} O, δ D) which encompasses the range of isotopic values of δ^{18} O_v and δ D_v in Amsterdam Island. "

P9, L247: What exactly is denoted by these numbers? The data range?

>> These are the δ^{18} O and δ D values has explained in the sentence above (answer to previous comment)

P9, L254ff: I could not follow you. Why does the data need to be corrected? What did you find here in the relationship that is not as it should be?

>> From Figure 2 of the manuscript, you can see that the value of $\delta^{18}O_{measured} - \delta^{18}O_{standard}$ is not flat as it should be when we inject the same standard at different humidity. This is a well known artefact of the water vapor analyzer which needs to be corrected as in all previous studies dealing with such kind of measurements.

We suggest the modified text:

"While we expect a constant null value for $\delta^{18}O_{measured}$ - $\delta^{18}O_{standard}$ on Figure 2 because we always inject the same water standards at different humidity, the $\delta^{18}O$ measurements of both EPB-AMS and GREEN-AMS standards decrease with increasing humidity with the same amplitude. $\delta D_{measured}$ - $\delta D_{standard}$ displayed on Figure 2 also shows variations but in contrast to the relative evolution of $\delta^{18}O$ with respect to water vapor mixing ratio, the δD measurements of both EPB-AMS and GREEN-AMS standards exhibit different behavior: δD of EPB-AMS increases by 1.5‰ and δD of GREEN-AMS decreases by 2.5‰ over the same 6,000-24,000 ppmv range for water vapor mixing ratio q_v . "

P15, L402: How do you know that these peaks occurred during a cold front? No analyses ofmeteorological parameters indicating a front passage are shown or discussed here.

>> Indeed, we simply wanted to say that these $\delta^{18}O_v$ excursions occurred near the presence of a cold front as observed on weather synoptic maps for each excursion (see answer to reviewer 2, Figure R3). But as explained in the answer of reviewer 2, many cold fronts are also not associated with $\delta^{18}O_v$ excursions and we refrain now from referring to much to the cold fronts in the revised version. We agree that this was confusing and we explained in the answer to comments of reviewer 2 how we dealt with this issue.

P14, L403-404: You only picked the 11 excursions with low correlation coefficient between d18Ov and qv. However, according to Fig. 3, there are other negative excursions of d18O besides these 11 cases. If the goal of this study is to show the isotopic features associated with the passage of the cold fronts, the authors should rather pick up the events from the weather chart showing a cold front passage, not from the low correlations alone.

>> As mentioned above, we concentrate on water vapor $\delta^{18}O_v$ excursions and not cold fronts. So we will still pick the events using low correlation coefficient between water vapor $\delta^{18}O_v$ and q_v and the occurrence of $\delta^{18}O_v$ excursions. As explained in our answer to reviewer 2, we give the followiong precisions in the revised manuscript :

« The 11 most abrupt events occurring when correlation coefficient R between $\delta^{18}O_v$ and d-excess_v is larger than -0.5 are associated with $\delta^{18}O_v$ negative excursion larger than 3 ‰ (at 6h resolution) on a total length smaller than 24 h (as taken between the mid-slopes of the decrease and increase of the $\delta^{18}O_v$ respectively). The 11 selected negative excursions occur at a rate larger than -0.5‰.h⁻¹ and the $\delta^{18}O_v$ increase at the end of each excursion has an amplitude larger than half the amplitude of the corresponding initial decrease. »

Note that some excursions were also discarded because of a too large interruption in the water isotopic record (21st March 2020).

P16, Figure 4: Add a legend so that we can understand which color indicates which data just by looking at it.

>> This has been added, see answers to comments to reviewers 1 and 2.

P17, L454-455: Sentence not clear since it is grammatically incorrect. Please rephrase.

>> We propose this new sentence "Such mismatch makes the understanding of the processes at play during these events particularly important to test to further improve the performances of atmospheric general circulation models equipped with water isotopes."

P18, L475: I still have not seen how you can be sure that there was a cold front passage. How have the cold fronts been detected?

>> You are right. This sentence leads to confusion since we wanted to focus on excursions only. We modify this sentence as:

"Several hypotheses can be proposed to explain the negative excursions of $\delta^{18}O_v$."

Line 504-506: Rain evaporation occurs under the cloud base, moistening the boundary layer. So, the authors should not underestimate the role of rain evaporation because of the high relative humidity near the surface.

>>> This is true, we actually can not rule out the effect of rain evaporation and we propose to be more cautious by stating :

"Surface relative humidity remains relatively high during these events (values given in Table 1 compared to a mean value of 77 %) which favors rain-vapor diffusive exchanges."

P19, Figure 6: This figure is not clear at all and needs more explanation. The plot represents the observations in the boundary layer, but the theoretical curves are the isotopic changes in the free troposphere? At least Noone (2012) used them to investigate processes in the tropical mid troposphere. What exactly is meant with "inspired" by Noone (2012)? How have these curves been derived? Do you take these from the Noone (2012) paper? Have you calculated/estimated these yourself?

>> The aim of figure 6 is to look at simple relationships between water vapor $\delta^{18}O_v$ and q_v using simple relationships. Calculation of the curves has been done using the formulas given in the Noone (2012) paper. These formulas are quite simple and could be used for different applications. Of course, it is better adapted to studies in the free troposphere but it has also been used in the past for studies of the isotopic composition of water vapor near the surface (e.g. Guilpart et al., JGR, 2017). We explain in the revised manuscript that this simple representation is a first order approach for discussing the mechanisms. But because it is not enough, we then use atmospheric general circulation models equipped with water isotopes in a second part of the discussion.

We hence propose to add the following explanation to introduce our analysis:

" First, to test the hypothesis of vapor-droplet interactions, we looked at the $\delta^{18}O_v$ vs q_v distribution following the approach already used by Guilpart et al. (2017) (Figure 6). We acknowledge that our approach is simple and should be taken as a first order approach since we can only look at the water vapor $\delta^{18}O_v$ vs q_v distribution in the surface layer while it may be more adapted to look at this relationship in the free troposphere. "

>> We also changed the word "inspired" in the caption of Figure 6 and propose:

"The solid lines are theoretical lines whose equations are detailed in Noone (2012) "

<u>Reference</u> :

Guilpart, E., Vimeux, F., Evan, S., Brioude, J., Metzger, J. M., Barthe, C., ... & Cattani, O. (2017). The isotopic composition of near-surface water vapor at the Maïdo observatory (Reunion Island, southwestern Indian Ocean) documents the controls of the humidity of the subtropical troposphere. *Journal of Geophysical Research: Atmospheres, 122*(18), 9628-9650.

P20, L523ff: Since subsidence is an important aspect of this study and you use Figure S1-S3 for the discussion, I don't understand why these figures are in the supplement instead of in the main part of the manuscript.

>> You are right and the modified figures according to the corresponding answer to reviewer 2 are now added in the main text (Figure 7) and appendices (figure A3 and A4).

P21, L623: I still haven't seen any proof that there has been a cold front passage.

>> As mentioned in other parts, this has been modified. Not all cold fronts are associated with $\delta^{18}O_v$ excursions (even if all water vapor $\delta^{18}O_v$ excursions occur in the vicinity of a cold front, Figure R4). We removed the expression "associated with cold fronts" here.

P25, L640: It is still not clear what the function of the models are. Are these only used to be evaluated or are these also used to understand the processes behind the peaks in the d18O time series?

>> Indeed, in this conclusion, we explain that we used the models for the two applications. In the second paragraph (l. 629 - 634), we say that the model-data comparison permits to explain the mechanism at play to explain the $\delta^{18}O_v$ excursion. And in the last paragraph (l. 635 - 640), we say that the kind of water isotopic records provided can be useful to evaluate model equipped with water isotopes.

We propose a new version of the last paragraph to improve clarity:

"We have shown that the isotopic composition of water vapor measured at the surface is a useful tool to identify aspects to be improved in the atmospheric component of the general circulation models. In our case, we used it to test different horizontal resolutions which influence the representativity of the vertical dynamics and have important implication in the simulation of surface variations of water vapor $\delta^{18}O_v$. Our study highlights the importance to have high-resolution models (e.g. mesoscale models) equipped with isotopes to further study such abrupt isotopic events.«

P26, L682: What do you mean with "of the s"?

>> Changed to "realized most of the simulations"

Supplement: The supplement contains too many figures. I think not all of them are really necessary and the number could be reduced. Further, the formatting should be the same as for the ACP paper, that means no underlined headers and the same style for the figures (no underline of the figure caption title and no italic text for the caption text.)

>> We will indeed reduce the numbers of figures and put some of them in the appendices.

Technical corrections

>> All technical corrections will be taken into account. Many thanks again for your careful reading

Review 1

I would like to compliment the authors for having prepared such a well-written paper and I strongly recommend the paper for publication after a very few adjustments/corrections. In general, the interpretation and the discussion of results is sounding and easy to follow. All figures are clear (see my only comment on Fig.4).

>> Many thanks for this general comment

I only have one comment/question about the interpretation of the results. Why d-excess has been (almost) left out of the discussion? The authors clearly state that during the depletion events both $\delta^{18}O_v$ -q and $\delta^{18}O_v$ -dexcess correlations break down. But how d-excess signal looks like during the event? If the d-excess doesn't change much, it would provide support to the hypothesis of rain-vapor interaction close to equilibrium than to rain-evaporation and to atmospheric subsidence, since evaporation of raindrops and free tropospheric air are associated with high d-excess.

>>> d-excess of water vapor is indeed not changing much over the $\delta^{18}O_v$ excursion. We thus agree that it may not be explained by strong rain drop evaporation at the surface (even if rain drop evaporation is mostly affecting the rain) and it is also in agreement with the relatively high relative humidity at the surface. We thus agree that it may then support rain-vapor interaction close to equilibrium for the acquisition of this signal. Such rain-vapor interaction is indeed indicated on our summary on Figure 9 and we added in the manuscript that the stable d-excess signal supports this interpretation.

New sentence in the section 4.1 of the discussion:

« Such interpretation is also supported by the stable d-excess, during these events."

Minor comments:

L149 Please rearrange number of gasses (4) and monitoring years (3).

>> We have simplified this sentencev to keep only what is useful for our study as:

" Hg species have been continuously measured since 2012 »

- L187 STP conditions: 273.15 K
- >> Thank you, this will be changed.

Figure 4 Including a legend and reducing the size of the caption could improve readability.

>> Thank you, we propose the updated figure:



<u>Figure 4</u>: Data model comparison (January – March 2020); a- water vapor d¹⁸O (light blue for data on hourly average, dark blue for data resampled at a 6-hour resolution); b- mixing ratio from our data set; c- vertical velocity; d- Precipitation amount. The grey shaded areas highlight the negative d¹⁸O excursions (note that in this figure the excursions of the 3rd and 9th of January 2020 are distinct while the distinction could not be done on Figure 3 because of the scale).

Review 2

This manuscripts presents a unique time series of stable water isotopes in water vapour in the undersampled area of the Southern Indian Ocean. The calibration of the water vapour isotope data follows established standards in the community. While the dataset presented here is of unique coverage, the relevance of the analysis and the conclusions is not convincing. Furthermore, the figures presenting the analysis are often difficult to read and many important information is in the supplement figures. In its current state, the manuscript lacks a motivation for the presented analysis and how it improves the understanding of the atmospheric water cycle. It might be worth considering a submission to Earth System Science Data (ESSD) instead of ACP.

>> The reason why we chose to submit to ACP is because we aim at combining for the first time water isotopes (data + 2 different models) and atmospheric species, here gaseous elemental mercury, over a long time series. The observation of concomitant water isotopes and Hg excursions suggests subsidence of air from high altitude.

We believe that water isotopes and elemental mercury records present an added value to the understanding of the dynamic of the atmospheric water cycle since they provide a surface diagnostic of processes happening higher in the atmosphere.

We agree with the reviewer that this objective was not clear enough in the manuscript, nor the added values of combining water isotopes, Hg and models and this is now hopefully better explain in the new version. Also, we worked on improving the readibility (and colorscale) of the figures and moved some figures (backtrajectories for example) from the supplement to the main text and in appendices as suggested by the editor.

Major comments: • Model-measurement comparison

The comparison of ECHAM6-wiso and LMDZ-iso with the measurements at Amsterdam Island leads to the main conclusion that "the isotopic composition of water vapor is a powerful tool to identify aspects to be improved in the general circulation models, such as the horizontal resolution which may influence the representativity of the vertical dynamics." Why are *isotope-enabled* models needed to illustrate that horizontal resolution influences vertical dynamics in GCMs? This is, for example, already evident when

comparing the vertical wind fields in Fig.7. Which "aspects to be improved", other than the horizontal resolution, were identified in this study?

Our study aims at exploring the mechanism driving the variations of water vapor $\delta^{18}O_v$ and especially the 11 events identified in the water vapor $\delta^{18}O_{v}$. With the aim to understand the associated processes, the comparison of the data series with outputs of atmospheric components of AGCMs equipped with water isotopes is a useful tool to evaluate potentiel biais in models. In addition to propose a mechanism for the water vapor $\delta^{18}O_v$ excursions through our model – data comparison, we could also propose some ideas to explain why some models could not reproduce the water isotopic excursions at the surface. Isotope implementation is quite similar in the 2 models and is probably not the reason for the differences, a result that we now highlight in the revised version of the manuscript. Nudging is slightly different between the two models (vorticity nudging in ECHAM6-wiso, shorter relaxation time in LMDZ-iso) but as the reviewer rightly points out, the conclusion is that if isotopes are controlled during these events by vertical dynamics at the scale of a few tens of kilometers, then it is probably necessary to have high-resolution models to unravel the mechanisms. As detailed in the comments below, we added in the discussion and conclusion a couple of sentences saying that our model-data comparison validates the physics processes within the ECHAM6-wiso model as well as the implemented physics of water isotopes. Moreover, we stress in the conclusion the importance to have higher resolution models (mesoscale models) equipped with isotopes if one wants to study such events.

Finally, vertical wind fields from initial figure 7 are from reanalyses, themselves based on model. They were not measured. So we believe that our study, unique in this region, still has an added values by comparing a new measured data series (the records of water vapor $\delta^{18}O_v$ at the surface) with model outputs.

Cold front analysis

The analysis is based on d18Ov excursions which are described as cold front events. The selection and analysis of these excursion with respect to cold frontal dynamics on a synoptic scale is missing. It is therefore difficult to interpret the described events with respect to the large-scale dynamics. In detail: • The analysis focuses on 11 events of cold fronts. These fronts and their spatial structure is not described in the manuscript and there are no synoptic figures describing a typical situation during a cold front passage, except for a few weather analysis charts in the supplementary information. How are the cold fronts identified? What are the properties of the cold fronts? How much of the annual precipitation is represented by cold frontal precipitation?

>> This comment is very sound. Our aim was actually not to focus on cold fronts but on the isotopic record and especially on periods when the isotopic record is providing an added value to understand features associated with the atmospheric water cycle, that is to say a different signal than the meteorological signals (humidity, temperature). This is why we concentrate on the abrupt negative water vapor $\delta^{18}O_v$ excursions which are not seen in the humidity signal. The periods during which we observed water vapor $\delta^{18}O_v$ negative excursions are associated with a cold front but there are probably many cold fronts that are not exhibiting any water vapor $\delta^{18}O_v$ excursions. Because this is not the subject of this analysis, we better avoid now to refer to cold fronts when unnecessary as it was indeed misleading.

In the previous manuscript, we had identified the cold fronts from the synoptic figures. All weather charts corresponding to the 11 water vapor $\delta^{18}O_v$ negative excursions are shown below:

	Day-1 of event at 00:00 UTC	Day of event at 00:00 UTC	Day+1 of event at 00:00 UTC
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10/05/2020	Note: Soft Vrate Soft Vrate NS12 Analysis (bp.) Soft Vrate Soft Vrate Valid bood Valid Soft Bood Vrate Soft Vrate Valid bood Valid Soft Bood Vrate Soft Vrate Soft Vrate Valid Soft Bood Vrate Soft Soft Soft Vrate Soft Soft Soft Soft Soft Soft	Mail Set Tool Well Analysis (DPa) Set Vell Set Tool 1003 Set Tool 101 Set Set 1023 Set Set 1024 Set Set 1025 Set Set 1026 Set Set 1023 Set Set 1024 Set Set 1023 Set Set 1024 Set Set 1025 Set Set 1021 Set Set	Mill Analysis (Drv) Vide 2000 VICT 1 May 2020 1000 A2TT 1 May 2020 H 1023 H 025 H 05 H 0



Figure R3: Weather analysis charts provided once a day at 00:00 UTC by the Analysis Chart Archive service of the Australian Government Bureau of Meteorology <u>http://www.bom.gov.au/australia/charts/archive/index.shtml</u>. Red dot on weather charts displays Amsterdam Island location.

The idea was to check for the presence of a cold front in a distance of 100 km around Amsterdam Island in a 48h period covering the time of the event. We indeed see that we systematically have cold

fronts in the vicinity of the Amsterdam Island at the time of the water vapor $\delta^{18}O_v$ excursions. Still, in some cases, such as the 06/12/2021, there are cold fronts identified on the weather charts in the vicinity of the Amsterdam Island 1 day before and 1 day after the excursions but no clear cold front over Amsterdam Island at the exact time of the water vapor $\delta^{18}O_v$ excursion.

We explain now very simply the link to cold fronts as :

«These negative $\delta^{18}O_v$ excursions always occurred during low pressure periods (atmospheric pressure below 1005 mbar) and we observe the presence of a cold front in a distance of 100 km around Amsterdam Island in a 48h period covering the time of the event (Supplementary Material). »

To address the occurrence of cold fronts in a more detailed manner, we propose here a second synoptic analysis with the frontal passage, computed as the maximum gradient of 850 hPa potential temperature, when this gradient is greater than 2 K/100 km, following Schemm et al. (2015). These results are displayed in the figure below which is now part of the supplementary material (2 first columns only):







Figure R4: synoptic analysis using hourly ERA5 fields at the time of observed minimum $\delta^{18}O_v$ correspondint to the 11 events identified in the manuscript: (a) air temperature at 850 hPa, (b) precipitation, and (c) vertical velocity at 850 hPa. White and black lines represent frontal passage, located at the maximum gradient of 850 hPa potential temperature. Front is computed as the zero-line of the gradient of the magnitude of the gradient of 850hPa air temperature, when the gradient of 850hPa air temperature is greater than 2 K/100 km, following Schemm et al. (2015).

<u>Reference :</u>

Schemm, Sebastian, Irina Rudeva, et Ian Simmonds. « Extratropical Fronts in the Lower Troposphere–Global Perspectives Obtained from Two Automated Methods ». Quarterly Journal of the Royal Meteorological Society 141, no 690 (2015): 1686-98. https://doi.org/10.1002/qj.2471.

 \circ The 11 events are chosen using the following criteria: "The green rectangles indicate the period with (1) correlation coefficient >-0.5 between d-excess and d18O of water vapor and (2) occurrence of a negative excursion in water vapor d18O." There are (from eye) other events that could fall into these criteria. For example, before the event in ~March 2021 (6th green rectangle in Fig. 3), there is an event agreeing with criteria (1) and showing a strong decrease in d18Ov. Why are other events not included? And how is a negative excursion in d18Ov defined?

>>> Many thanks for this comment and we agree that the definition of a negative excursion was not clear enough and we agree that it may be a bit subjective since we took only the 11 most prominent events. The reason why the event in March 2021 has not been selected is its slower d180v decrease at the start of the excursion (less than 0.5 permil.h-1) and the fact that, at the end of the excursion, the $\delta^{18}O_v$ does increase with an amplitude which is less than half the amplitude of the initial decrease (2.4 permil to be compared to 6 permil). This criterium is now added in the new version of the manuscript.



Figure R5 : Focus on the event of the 19-20 February 2021, not selected for our 11 most prominent events because one criterium in the evolution of the $\delta^{18}O_v$ was not fulfilled. Still, this event shares many of the main characterictics of the selected events.

We now precise in the revised manuscript that :

« The 11 most abrupt events occurring when correlation coefficient R between $\delta^{18}O_v$ and d-excess_v is larger than -0.5 are associated with $\delta^{18}O_v$ negative excursion larger than 3 ‰ (at 6h resolution) on a total length smaller than 24 h (as taken between the mid-slopes of the decrease and increase of the $\delta^{18}O_v$ respectively). The 11 selected negative excursions occur at a rate larger than -0.5‰.h⁻¹ and the $\delta^{18}O_v$ increase at the end of each excursion has an amplitude larger than half the amplitude of the corresponding initial decrease. »

Note that some excursions were also discarded because of a too large interruption in the water isotopic record (21 March 2020).

• The analysis of the 11 events is mainly qualitative, and it is difficult to follow the description of the 4 events from 01-03/2020. All map plots and most of the vertical cross sections of these events are in the Supplementary Information, which makes it difficult to understand the synoptic situation during the events and the model performance. Further, it is not clear why these four events where chosen for a detailed description. Also, the description of the events is scattered in different paragraphs of Sections 3 and 4. The paragraphs should be better structure to lead the reader through the evolution of the cold front events.

The reason why we chose to show only the events from 01-03/2020 in the main text is simply to be able to read the figures easily and avoid too many repetitions. This period was favored because it encompasses several events which were then easily to see on a graph covering 3 months. As we wrote, « This period has been selected for display because it encompasses 4 out

the 11 negative excursions of $\delta^{18}O_v$, but the extended comparison over the whole 2 years period is displayed on Figure S1. » and the same conclusions can be drawn if we consider the 11 events (all discussed in Table 1). To support this assumption, we now provide in the appendices the figures corresponding to each event (such as current Figure 5 for the events of January 2020).

Following this comment, we have reorganised the section 4 with addition of subtitles and reduction of some paragraphs (sections 3 and 4) to improve readibility. We also added more figures from the supplement in the main text (backtrajectories) or appendices (Figure S1 has been moved in appendices, figure A1, and we have added a focus on each event as Figure A2).

Further general comments:

• The section headings are not very specific. While Section 3 "Data description" has many short subsections, Section 4 "Discussion" has no subsection while introducding a lot of information and new analyses.

>>> We have included subsection titles in the discussion (4.1 to 4.5) so that it will be easier to follow the argumentation. Some paragraphs have also been rewritten for clarity as explained in answer to specific comments listed below.

• Water isotope measurements:

Various information is missing in the description of the water isotope measurements: • What was the material and length of the inlet line to the Picarro instrument? Was the inlet line heated ?

>>> The inlet line was indeed heated (40°C) and the 5 m inlet tube was of PFA. This is explained in the new manuscript.

« The instrument has been installed in a temperature-controlled room at the observatory on the Amsterdam Island and the sampling of water vapor is done outside at ~ 6 m above ground level (or 76 m above sea level) through a 5 m long inlet tube made of PFA (perfluoroalkoxy alkanes) and heated at 40°C."

\circ Was the water vapour mixing ratio measured by Picarro calibrated? How does it compare to other humidity measurements on the Island?

>>> The calibration of water vapour mixing ratio was done in the laboratory before sending the instrument and this protocol is valid as this calibration depends only on the laser cavity configurations. On the field, we found an excellent agreement between mixing ratio measured by the Picarro and by the weather station (the difference between the two records always stays below 2% and there is no systematic shift between the two records). This is added in the revised version :

« The calibration of water vapour mixing ratio was performed in the laboratory before sending the instrument. On the field, we found an excellent agreement between mixing ratio measured by the Picarro instrument and mixing ratio measured by the weather station (the difference between the two records always stays below 2% and there is no systematic shift between the two records). " $^\circ$ When was the humidity-isotope dependency calibration done and what kind of calibration device was used?

>>> The humidity-isotope dependency calibration was checked every year and the calibration device is the standard delivery module by Picarro. These explanations are given in the new manuscript.

« The calibration of the water isotopic data is performed in several steps following previous studies (Leroy-Dos Santos et al., 2020; Tremoy et al., 2011) and using a standard delivery module by Picarro. »

« This calibration step has been performed every year over the whole range of mixing ratio

values and provided very similar results from one year to the other."

Detailed comments:

Lines 68-69: What is (18O/16O) and (D/H)? Does this represent the isotopic ratio?

>> Indeed, these are isotopic ratios between heavy and light isotopes. This is now added.

Lines 90-93: "For this objective, several instruments have been installed either in observatory stations ... or on boat ...".

Is there a reason that this summary omits aircraft measurements?

>>> Many thanks for this comment, we now also mention aircraft measurements adding a reference to the following study as an example :

Henze, D., Noone, D., and Toohey, D.: Aircraft measurements of water vapor heavy isotope ratios in the marine boundary layer and lower troposphere during ORACLES, Earth Syst. Sci. Data, 14, 1811–1829, https://doi.org/10.5194/essd-14-1811-2022, 2022.

Lines 97-99: "Such data comparison enables one to test the performances of the models either in the simulation of the dynamic of the atmospheric water cycle or in the implementation of the water isotopes."

I agree with this statement but I don't see how this study adds any new knowledge on model performance or isotope parametrisations. Can you elaborate further?

>>> In the two models presented here, a very similar approach has been followed for water isotopes implementation. Because one model is able to reproduce well the observed water vapor $\delta^{18}O_v$ excursions, we can conclude that the isotopes parameterisation is appropriate (at least in this region). It means that the reason why the other model is not able to reproduce the excursion is not due to isotopic parameterisation but to the modeled atmospheric dynamics whose horizontal resolution is likely too coarse. We have rewritten some part of the manuscript to adress this comment in the discussion section:

Discussion :

« Still, the fact that at least ECHAM6-wiso is able to reproduce every negative $\delta^{18}O_v$ excursions (whether they are associated or not with subsidence or rain-water vapor reequilibration) shows that not only the patterns of atmospheric water cycle are correctly reproduced (a validation which can also be performed using humidity and precipitation data) but also that the isotopic processes are correctly implemented in this model. Such abrupt $\delta^{18}O_v$ events can hence be used as a test bed of the skills of water isotopes enabled general circulation models. "

Conclusion :

"The good agreement between modeled and measured $\delta^{18}O_v$ when using ECHAM6-wiso validates the physics processes within the ECHAM6-wiso model as well as the implemented physics of water isotopes.

[....]

This study highlighted the added value of combining different data from an atmospheric observatory to understand the dynamics of the atmospheric circulation. The 2-year records are also a good benchmark for model evaluation. We have especially shown that the isotopic composition of water vapor measured at the surface is a powerful tool to identify aspects to be improved in the atmospheric component of the general circulation models. In our case, we used it to test different horizontal resolutions which influence the simulation of the vertical dynamics and have important implication in the simulation of surface variations of water vapor $\delta^{18}O_{v}$. Our study highlights the importance to have high-resolution models (e.g. mesoscale models) equipped with isotopes to further study such abrupt isotopic events."

Line 101-102: "This region is poorly documented with present-day observations despite its primary importance in governing CO2 sinks" Do you have a reference for this statement?

>>> This sentence has been removed for simplicity, cf next comment.

Line 102-105: "Moreover, we lack precise descriptions of atmospheric processes associated with cloud microphysics and surface-atmosphere exchange in polar regions, and the evolution of westerly wind locations and strength (Fogt and Marshall, 2020)." Why is this relevant for the presented study? The study site lies in the mid-latitudes.

>> We agree that this sentence is more general and not really adapted for this study in particular (it mostly refered to our other studies in Antarctica). For simplification, we propose to remove « This region is poorly documented with present-day observations despite its primary importance in governing CO_2 sinks. Moreover, we lack precise descriptions of atmospheric processes associated with cloud microphysics and surface-atmosphere exchange in polar regions, and the evolution of westerly wind locations and strength (Fogt and Marshall, 2020)."

Line 134: "Climate is temperate, generally mild with frequent presence of clouds." What do you mean with frequent presence of clouds?

>> We precise by stating that « the average total sunshine hours is 1581 hours per year from the period 1981 – 2010 » (https://donneespubliques.meteofrance.fr/FichesClim/FICHECLIM_98404002.pdf)

Lines 149-150: "CO₂, CO, CH₄ and Hg species have been continuously measured since 1980, 2014, and 2012 respectively."There are four species but only three years are mentioned. It is not evident which species belongs to which year.

Because of the need to simplify this section (cf other comments) and keep only what is necessary for our study, the paragraph has been simplified as :

« Numerous atmospheric compounds and meteorological parameters are and were continuously monitored at the site since 1960 (Angot et al., 2014; El Yazidi et al., 2018; Gaudry et al., 1983; Gros et al., 1999, 1998; Polian et al., 1986; Sciare et al., 2000, 2009; Slemr et al., 2015; Slemr et al., 2020). In particular, the Amsterdam (AMS) site hosts several dedicated atmospheric observation instruments notably at the Pointe Bénédicte atmospheric observatory (70 m above sea level) where greenhouse gases concentrations and mercury (Hg) are monitored. . Hg species have been continuously measured since 2012. "

Wind speed and direction, atmospheric pressure, surface temperature and relative humidity data are currently obtained at a minute resolution. Another meteorological station is based on the island and is operated by Météo France at Martin-de-Viviès life base around 27 m above sea level, about two kilometers east from the Pointe Bénédicte observatory collecting air temperature, humidity, precipitation, wind speed and direction, pressure and solar radiation "

Line 165: What is IGE?

>> IGE is the "Institut des Geosciences de l'Environnement", it has been removed in the new version (for simplification as requested in previous comment)

Line 187: What is STP?

>> It means "Standard temperature and pressure" and is written as such in the new manuscript. Also there was a mistake in the first manuscript since the temperature should be 273.15 K.

Line 202: "high altitude air masses (lower/ upper troposphere, or even above)" This is very unspecific. What do you mean with lower/upper troposphere?

>> There is not a precise altitude above which the GEM profile shows a decrease and is replaced by Hg oxidized species. The observations show that when we are above the free troposphere (in general above 5-6 km) and in the low stratosphere and when there is no biomass burning transportation from Africa, the GEM concentrations decrease with height (and this is the inverse for oxidized species). We thus propose to replace the following text:

"In this study, atmospheric GEM is used as potential tracer of intrusion and/or subsidence of high altitude air masses (lower/ upper troposphere, or even above) that may possibly impact the atmospheric records in Pointe Benedicte Observatory which collects marine boundary layer most of the time"

by:

"In this study, and even if long-range transport and a variable tropopause height may modulate, atmospheric GEM is used as potential tracer of stratosphere-to-troposphere intrusion and/or subsidence of upper troposphere (above 5-6 kms) that may impact the atmospheric records in Pointe Benedicte Observatory which collects marine boundary layer most of the time"

Lines 204-205: "As mentioned above, mercury in the atmosphere is detected in three defined forms:"This has not been properly introduced earlier.

>>> We propose to replace the first sentence of section 2.2 by:

"Mercury (Hg) in the atmosphere consists of three forms: gaseous elemental Hg (GEM as defined above), gaseous oxidized Hg and particulate-bound Hg. »

Line 233: "outside at ~ 6 m above ground level." What is this relative to m agl/asl, i.e. compared to the other measurements?

>> See previous answers to comment. We now precise :

« . The instrument has been installed in a temperature-controlled room at the observatory on

the Amsterdam Island and the sampling of water vapor is done outside at ~ 6 m above ground level (or 76 m above sea level)."

Line 279: What do mean with "quadratic error"?

>> Because d-excess is defined as d-excess = δD -8× $\delta^{18}O$, the "quadratic error" on d-excess (σ^2) is calculated as the root square of ($\sigma_{\delta D}^2$ +64× $\sigma_{\delta 18O}^2$). The term "quadratic error" is confusing and is replaced by "uncertainty" as for the $\delta^{18}O$ and δD uncertainties given above.

Line 290: Why are you starting the trajectories at 100m a.s.1.?

>> Because our instrument is at the surface and the Pointe Bénédicte observatory is 70 m above the ground, we chose the 100 m level. But you are right that we could also have taken a lower level. We have checked that we obtain exactly the same pattern when starting the back-trajectories at 100 m or 50 m high.

Line 324: "high spatial": 0.9° horizontal resolution is high compared to the LMDZiso simulation of this study but low compared to convection permitting climate simulations. I would therefore skip "high".

>> OK

Line 389: "d-excess of the precipitation". I don't see this in Fig. 3.

>> Indeed, we did not show this record because it does not add much to the study. We precise in the new version « No significant seasonal variations are observed in the record of d-excess of precipitation (not shown). »

Line 395, 395, 399: What is R₂? The correlation coefficient R to the power of 2? What kind of correlation are you calculating? R₂ is used before R is introduced.

>> R2 is the coefficient of determination for a linear regression between the time series at hourly resolution. We now precise "with R2 being the coefficient of determination for a linear regression".

Line 399-400. "...(R is calculated continuously from hourly records in 8 consecutive days)..." Do you mean that you used an 8-day moving window?

>> Yes, this is correct and the sentence has been changed

Line 402-403: "d-excessy" has not been introduced.

>> It is now introduced as "d-excess_v = δD_v - $8 \times \delta^{18} O_v$ "

Lines 434 - 436: "...the agreement with measured precipitation amount is better for ECHAM6-wiso (R2 = 0.45) than for LMDZ-iso (R2 = 0.08 - 0.13 for VLR - LR)..."

The correlation of LMDZ-iso with the measurements is close to zero, i.e. there seems to be nearly no agreement. The statement that the agreement with measured precipitation is better for ECHAM6-wiso than for LMDZ-iso seems weak in this context.

>>> We changed the wording and wrote that the correlation for LMDZiso is close to zero while there is a correlation for ECHAM6-wiso.

« The correlation between modeled and measured precipitation is close to zero for LMDZ-iso ($R^2 = 0.08 - 0.13$ for VLR - LR) while there is a better agreement when comparing measured precipitation amount to outputs of ECHAM6-wiso ($R^2 = 0.45$). »

Lines 437-439: "...they are in general more strongly expressed in the data series than in the model series which is only partly due to the hourly resolution of the d18Ov record compared to the 3h and 6h resolution of the outputs of the LMDZ-iso...)." What is the basis of this conclusion?

>> Our idea is simply to see if part of the disagreement between data and model is linked to the fact that the models have only a 3h or 6h resolution while the data were displayed at 1h resolution. To test this, we have reinterpoled our data at 6h resolution in the figure 4 and calculated the amplitude of the water vapor $\delta^{18}O_v$ excursions with data resampled at 6h resolution. We will better explain that our aim was here to look if the difference resolution between the models and between models and data can explain the bad agreement between model outputs and data for the LMDZ-iso model.

We propose this new formulation :

« Part of this disagreement can be explained by the fact that the $\delta^{18}O_v$ record has a higher temporal resolution (1h) than the model outputs (3h for LMDZ-iso and 6h for ECHAM6-wiso). However, when interpolating the $\delta^{18}O_v$ record at a 6h resolution, the negative excursions are still clearly visible which cannot be captured by the LMDZ-iso model (Figure 4 and Table 1). »

Linbes 450-451: "They always occurred during low pressure periods (atmospheric pressure below 1005 mbar)." What is the synoptic situation leading to this low pressure and cold fronts?

>> We have removed the numerous references to cold fronts because it was confusing and we actually wanted to focus on the water vapor $\delta^{18}O_v$ excursions. We thus do not discuss the synoptic situations which is not covered by our study. We rewrote this part as :

« «These negative $\delta^{18}O_v$ excursions always occurred during low pressure periods (atmospheric pressure below 1005 mbar) and we observe the presence of a cold front in a distance of 100 km around Amsterdam Island in a 48h period covering the time of the event (Supplementary Material Figure S1). »

Lines 501-503: "However, for the 11 events highlighted above, the d18Ov vs qv evolution follows an evolution characteristic of remoistening processes, i.e. a curve standing below the curve of the d18Ov vs qv evolution observed for the rest of the series..."

The single events show a much steeper evolution in the d18Ov-qv diagram than the remoistening curve. Why is this?

>> You are right. As mentioned as answer to the editor, this figure is a "first order" approach following previous study of Guilpart et al. (2017) but is not 100% appropriate since the simple modeling curves are idealized $\delta^{18}O_v$ vs q_v trajectories. Even if we have adapted our initial conditions to the observation of the surface isotopic composition of the water vapor, we do not expect the curves to be aligned with our events and we simply use them at first approach to show that enhanced remoistening or water-rain interactions may be a good candidate to explain the $\delta^{18}O_v$ vs q_v relationship during the events.

We propose to better explain as:

" Even if the water vapor $\delta^{18}O_v$ vs q_v evolution is rather steep, there is some resemblance with the idealized theoretical curve for remoistening initially calculated for the free troposphere (Noone, 2012) and adapted here with initial conditions corresponding to the surface water vapor isotopic commposition. Even if the analogy between our measurements and this simple modeling approach should be taken with cautious, the fact that the water vapor $\delta^{18}O_v$ vs q_v evolution lies below the idealized curve for condensation processes supports the depleting effect of vapor-rain interactions for our negative water vapor $\delta^{18}O_v$ excursions (Noone, 2012; Worden et al., 2007). "

Lines 504-506: "Since relative humidity is relatively high during these events (values given in Table 1 compared to a mean value of 77 %), it more likely reflects rain-vapor diffusive exchanges than rain evaporation." Are you referring to the relative humidity at the surface? How about relative humidity above that will also influence the interaction of the rain with its surroundings?

>> This is correct, we were referring to relative humidity at the surface. Actually, high relative humidity will favor the diffusive exchange between rain and vapor but we can not exclude reevaporation at the surface nor in the upper atmosphere since both can have an influence as shown in Figure 8 and in 1. 585 – 588 of the previous manuscript. We thus propose the new text :

« Surface relative humidity remains relatively high during these events (values given in Table 1 compared to a mean value of 77 %) which favors rain-vapor diffusive exchanges. »

Line 519ff.: As the trajectories are only shown in the supplement, it is difficult to follow this paragraph. The beginning of the paragraph leads to think that the trajectories indicate that subsidence is import but in the end the conclusion is that "back trajectories are however not supporting systematic subsidence for other cases".

>> Indeed, the backtrajectories do not evidence systematic air subsidence for the water vapor $\delta^{18}O_v$ excursions and this is the reason why the figures were initially put in the supplement but we agree that it is better to have some of them in the main text. Actually, when looking in details the atmospheric dynamic, it is clear that the $\delta^{18}O_v$ excursions occur at the transition between ascendence and subsidence and this is probably the reason why we could not easily detect it from 10 days back-trajectories. This will be better explained in the text with back-trajectories of 5 days only (cf next answer to comment).

Lines 526-527: "... the maximum altitude of the envelope of the back trajectories increases from 5,000 to 8,000 m..."

What it the mean/standard deviation of the maximum trajectory height? How many days before arrival are the trajectories at their maximum height? How relevant is this for the isotopic composition upon arrival? E.g. if the trajectories descend over the ocean and take up moisture, their maximum height before the moisture take-up is less relevant for the isotopic composition at Amsterdam Island. If you are using the full 10-days backward trajectories to calculate the maximum altitude, I don't think that the maximum altitude is a good measure of subsidence in front of the cold front.

>> Actually, what we wanted to test initially was if there is a change of the origin of the air mass during the excursion which may explain different isotopic signature of the water vapor. To answer the questions listed here, we have developed a new representation of the back-trajectories on only 5 days and with the indication of the location of the average (using humidity weighting) backtrajectory. We propose also to better explain that back-trajectories are used to mainly study the change of air origin.

The new paragraph in the main text reads :

« To further explore the processes leading to the decoupling of humidity and $\delta^{18}O_v$ as well as sharp negative excursions of $\delta^{18}O_v$ during the 11 events identified here, we also use information from the ERA5 reanalyses. In particular, the influence of atmospheric circulation (vertical and horizontal advection) and moisture origin can be studied through back trajectories. The back trajectories performed over the 5 previous days (Figures 7 and A3) confirm the information from wind directions that there is no systematic change in the horizontal origin of the trajectories for the different events. No systematic pattern is also identified in the vertical advection even if we note that for the event of the 3rd of January, the average altitude of the envelope of the 5-day back trajectories increases when comparing the situation before the excursion and the situation during the most negative water vapor $\delta^{18}O$ value. This observation may support the occurrence of air subsidence indicated by the GEM record on this particular event (Figure 7). "

Figure 7 is the following:



Figure 7 : FLEXPART footprints of 5-day back trajectories for the event of the 3rd-4th of January. (a) Latitude-longitude projection of the FLEXPART back trajectory footprint for the 3rd of January 2020 at 13h30. The yellow to green colors on each grid point of these projections represent the density of particles. The white to blue colors indicate the water vapor mixing ratio on the humidity weighted average back-trajectory. Each red point indicates the location of the average back-trajectory for each of the 5 days before the date of the considered event. (b) Same as a for the 3rd of January 2020 at 22h30. (c) Top shows the evolution of the water vapor mixing ratio of the back trajectories for the 3rd of January 2020 at 13h30; bottom shows the altitude evolution of the back trajectory for the 3rd of January 2020 at 13h30. (d) same as (c) for the 3rd of January 2020 at 22h30. »

Lines 543-555: "There is no evidence for changes in the horizontal advection of air over the 11 particular events from the observation of wind direction around these cold front events."

How is the cold front identified? Does it divide different air masses? A cold front normally implies a horizontal transport of air, why is this not the case for these cold fronts?

>> The cold front were initially identified with the synoptic weather charts as explained in answers to other comments. As mentioned elsewhere, the reference to these cold fronts are muted in the new version and we have removed « around these cold front events » from this particular sentence.

Lines 556-558: "Such abrupt d18Ov events can hence be used as a test of the performances of general circulation models equipped with water isotopes."

What was learned about the performance of the GCMs involved in this study? Was it necessary to include d180 in such a performance test instead of just using traditional humidity variables (e.g. relative humidity, specific humidity, precipitation)?

>> You are right that many skills could already be tested using only meteorological data. This is the reason why we focused here only on the periods when water vapor $\delta^{18}O_v$ was showing a different signal than the one inferred from humidity to study what we can learn from this signal. Also, the combination with Hg measurements suggesting subsidence was useful to complement the traditional variable records over these excursions. Finally, because more and more models are equipped with water isotopes, we wanted to show an example of the peculiar signal seen in the water isotopes and not in traditional humidity variable to test the skills of these models, both on the implementation of water isotopes and on the dynamic of the atmospheric water cycle.

We propose this new formulation to take into account this comment :

« Still, the fact that at least ECHAM6-wiso is able to reproduce every negative $\delta^{18}O_v$ excursions (whether they are associated or not with subsidence or rain-water vapor reequilibration) shows that not only the patterns of atmospheric water cycle are correctly reproduced (a validation which could also be performed using humidity and precipitation data) but also that the isotopic processes are correctly implemented in this model. Such abrupt $\delta^{18}O_v$ events can hence be used as a test bed of the skills of water isotopes enabled general circulation models.«

Line 562: What is "SOM"?

>> Supplementary Online Material, it is not in the new version.

Lines 559-584: As both isotope-enabled models were nudged to ERA5 dynamics, it is to be expected that the GCMs reproduce the ERA5 reanalysis wind fields rather well with some caveats due to the lower horizontal resolution. This paragraph (and Fig.7) is mostly describing the smoothing of ERA5 due to the coarser resolution of the isotope-enables GCMs. Why do we need isotope measurements to see the effect of a coarser horizontal resolution? What do we learn about the GCM performances by decreasing the horizontal resolution?

>> It is true that we do not need isotopes to show the effect of decreasing resolution and we have removed the sentences which may convey this idea (especially the first sentence of the paragraph). Still, the idea with the use of water isotopes is to have a record at the surface of what is happening above in the atmosphere (without relying on reanalyses only) and this is the reason why we think that they have an added value to a pure comparison with reanalyses.

In addition, testing the different resolutions shows that good horizontal resolution is essential to correctly simulate front dynamics (as was already pointed by Ryan et al. 2000) and associated isotopic variations.

Ryan, B. F., and Coauthors, 2000: Simulations of a Cold Front by Cloud-Resolving, Limited-Area, and Large-Scale Models, and a Model Evaluation Using In Situ and Satellite Observations. *Mon. Wea. Rev.*, **128**, 3218–3235

We propose the modified paragraph :

« To further explore the $\delta^{18}O_v$ data-model comparison and associated processes, we compare the skills of the ECHAM6-wiso and the LMDZ-iso models over the first months of 2020 in term of atmospheric dynamic (the whole series is displayed in Figure A1). First and as expected because of the nudging, the two models reproduce rather well the evolution of the vertical velocity from the ERA5 reanalyses with a stronger ascent for the model predicting the strongest precipitation amount (e.g. LMDZ-iso for 24th of January 2020). The event of the 3rd of January is the only one reproduced by both ECHAM6-wiso and the two versions of the LMDZ-iso model: the three simulations show a clear subsidence over the isotopic event and a clear negative $\delta^{18}O_v$ excursion (Figure 4). For the other events, neither LMDZ-iso nor ECHAM6wiso show a clear signal of subsidence neither at 500 nor at 850 hPa (Figure 4). However, the horizontal distribution of vertical velocity obtained with ECHAM6-wiso and LMDZ-iso are significantly different (Figure 7 for the event of the 9th of January, Figures S5 for the other events). While the LMDZ-iso modelled vertical velocity displays a rather strong homogeneity on the vertical axis, ECHAM6-wiso modelled vertical velocity highlights subsidence of air below the ascending column at the exact location of the negative $\delta^{18}O_v$ anomaly (Figure 7c). This subsidence of depleted $\delta^{18}O_v$ below the ascending column is responsible for the sharp negative $\delta^{18}O_v$ excursion in the ECHAM6-wiso model. The fact that subsidence of air occurs just below uplifted air, at the limit between ascendance and subsidence (Figure 7j), permits to reconcile the GEM data suggesting subsidence and the sign of the vertical velocity of the ERA5 reanalyses at Amsterdam Island. Since the isotope implementation was done similarly in the two models, the reason why the LMDZ-iso model does not reproduce the water isotopic anomaly is its too coarse resolution as also supported by the comparison between skills of the LMDZ-iso model at low resolution and very low resolution for the event of the 24th of January (Table 1 and Figure 4). As already pointed by Ryan et al. (2000), a fine resolution is necessary to correctly simulate front dynamics and we extend this result here to the high resolution temporal patterns of surface $\delta^{18}O_{v}$. "

Lines 586-590: "A rain event is associated with a strong ascending column in which d18Ov is depleted by progressive precipitation during the ascent and by interaction between rain and water vapor. This ascending column is coupled to the subsidence of d18Ov depleted air at the rear of the event which is pushed toward Amsterdam Island through a south west advection of cold air."

How is the isotopic composition of the subsiding air behind the cold front connected to the progressive precipitation during the ascent? Can the ascending and descending column be differentiated in the d18O excursions?

>> This is a very valid question which is not so simple to answer with the model outputs since we do not have water tagging. Still, we did a simple analysis using the outputs of the ECHAM6-wiso model looking at the water vapor $\delta^{18}O_v$ vs q_v in front of the event (ascending column), during the event and at the rear of the event (subsiding column) (Figure R6).



Figure R6: (left) Evolution of the water vapor $\delta^{18}O_v$ vs q_v in ECHAM6wiso, at the surface at the nearest grid cell to Amsterdam island for all time steps between 01/01/2020 and 31/03/2020 (grey dots), at the surface for event 3 (24th of January 2020) at the latitude of Amsterdam and for all longitudes between 50°E and 100°E (black dots), and for three atmospheric columns at the time of the event (plain colored lines). The three vertical atmospheric columns are taken as best representations of the situation before the water vapor $\delta^{18}O_v$ anomaly of the 24th of January 2020, as an example. Ascending column is represented by the vertical atmospheric column at 81.6°E (upstream of / before the anomalous $\delta^{18}O_v$ event, green line); the situation during the event can be visualized as the vertical atmospheric column at 77.8°E, orange line; the vertical atmospheric column after the event (downstream) can be visualized by the column at 73.1°E, blue line. Vertical velocity directed downward is represented by a thick line (only present for the blue line). The black line indicates the distribution $\delta^{18}O_v$ vs q_v for marine mixing and the dashed lines show Rayleigh distillation distributions. (right) as in Figure 7b of the article, but for event 3, and showing the location of the three atmospheric columns with same color lines as in the right.

The period before the $\delta^{18}O_v$ anomaly corresponds in most cases to the end of the rain event. This period is associated with a strong lift of the moist air in which we see a water vapor $\delta^{18}O_v$ vs qv distribution (low $\delta^{18}O_v$ with high humidity, green curve on Figure R6) which looks like an extreme case of remoistening (super Rayleigh as described in the Figure 6 of the main manuscript). We can

interpret this as ongoing rain-steam exchanges. The situation is different from what happens at the surface where water vapor $\delta^{18}O_v$ vs q_v is not showing any anomalous behavior yet.

During the event (orange curve on Figure R6), the water vapor $\delta^{18}O_v$ vs q_v evolution is completely vertical and really difficult to be explained by only remoistening effect. We thus believe that a dynamic aspect (mixing) is also involved in bringing in the surface boundary layer low water vapor $\delta^{18}O_v$ with relatively high humidity.

After the event (blue curve on Figure R6), we are back to a classic Rayleigh situation, so the water vapor $\delta^{18}O_v$ returns to its initial value.

We did not add anything on this analysis in the new version but if the reviewer and the editor thinks that this is an interesting added value, we propose to ad dit in the supplementary.

Line 621: "hours/days". Is there a cold front passage that has a duration of several days?

>> You are right, we do not have cold fronts lasting several days over the Amsterdam Island. As noted in previous answers to comments, we did put too much emphasis on cold fronts which was misleading. We removed "associated with cold fronts" on I. 623.

Lines 635-640: "This study highlighted the added value of combining different data from an atmospheric observatory to understand the dynamic of the atmospheric circulation. The two-year records are also a good benchmark for model evaluation. We have especially shown that the isotopic composition of water vapor is a powerful tool to identify aspects to be improved in the general circulation models, such as the horizontal resolution which may influence the representativity of the vertical dynamics."

As also mention above, why are stable water isotopes needed to show that the horizontal resolution may influence the vertical dynamics? The vertical cross sections of vertical wind speed (Fig. 7) illustrates this already quite well.

>> This comment has been addressed in the list of major comments above.

on the Zenodo platform, and the dataset does not include the water vapour mixing ratio.

>>> We will complete the dataset with a version 2 after acceptation of the manuscript including water vapour mixing ratio.

Figures:

General: The figures are often difficult to read, especially the described phenomena are small (e.g. d18O excursions of a few hours in a 2-year or 3 month timeline). Additionally, the colors are not color-blinded friendly and the caption are not concise.

>> We agree that it is difficult to see the excursions on a 2-year timeline and this is the reason why it has been decomposed in several figures of 3-months (now in appendix, see answer to comment above). In addition, some excursions are detailed in the main text (and all are now provided in the

appendix, see comment above). Figure 4 (and corresponding figures in appendix) is now colorblinded friendly and the captions concise (see below for details).

Fig.1: Is this figure needed? Fig.1 is not mentioned in the text.

>> We mention it in the text at first instance.

« In order to complete the picture of the atmospheric water cycle over the Indian basin of the Southern Ocean already measured by these three analyzers, we installed a new water vapor isotopic analyzer in the mid-latitude of the south Indian Ocean on Amsterdam Island (Figure 1) in November 2019. »

Fig.3: x-axis too coarse, light green shading difficult to see.



We change to the following figure:

Fig. 4: Colors red/blue/green are not colorblind friendly. The caption text includes many repetition and should be improved. A legend in the figure could improve the readability.

>> We propose this modified figure (new color code) which will also enable to shorten the caption (since information is on the legend in the figure):



On this panel, we also removed the vertical velocity at 500 hPa from panel c to improve readability and because it did not bring much additional information (the information is given in Table 1).

New caption :

« <u>Figure 4</u>: Model-measurement comparison (January – March 2020); a- $\delta^{18}O_v$ (light blue for data on hourly average, dark blue for data resampled at a 6-hour resolution); b- water vapor mixing ratio from our data set; c- vertical velocity; d- Precipitation amount. The grey rectangles highlight the negative $\delta^{18}O$ excursions (note that in this figure the excursions of the 3rd and 9th of January 2020 are distinct while the distinction could not be done on Figure 3 because of the scale)."

Fig. 6: What is φ=0.025?

This value is indicated together with the "remoistening curve" which has been obtained following the expression in Noone (2012). Basically, the idea is to express remoistening through a modification of the equilibrium fractionation coefficient between water vapor and rain (α_e) so that the effective fractionation factor will be $\alpha = (1+\phi) \times \alpha_e$

This effective fractionation coefficient is then introduced in the Rayleigh distillation equation to deduce the link between $\delta^{18}O_v$ and mixing ratio:

$(\delta - \delta_0) = (\alpha - 1) * \ln(q/q_0)$

We provide this information in the revised version of the manuscript as :

« Remoistening is described through a modification of the equilibrium fractionation coefficient between water vapor and rain (α_e) so that the effective fractionation factor is $\alpha = (1+\phi) \times \alpha_e$. This effective fractionation coefficient is then introduced in the Rayleigh distillation equation to deduce the link between $\delta^{18}O_v$ and mixing ratio as:

$$(\delta^{18}O_{v-}\delta^{18}O_{v,0}) = (\alpha - 1) \times \ln(q_{v/q_{v,0}})$$
 (Eq 8)"

Figure 8: What is SBL? The ascent of air in front of the cold front rises nearly vertical at a constant longitudinal position. As a cold front is moving system (mostly associated with an extratropical cyclone), the ascent does not occur at a constant location (in latitude and longitude). Further, all precipitation seems to fall in front of the cold front, which is unlikely.

SBL is Surface Boundary Layer, it has been changed to Marine Boundary Layer.

We agree that we need to include a more detailed analysis of the synoptic situation during the events. In Figure R4 shown above, we show front location, precipitation and 850 hPa vertical velocities from ERA5 at the time of the events. Precipitation generally falls just ahead of the cold front.

Figure 8 (now Figure 9) scheme is based on the profiles modelled by ECHAM for event of the 9th of January 2020. We show these profiles below (Figure R7). We now make this clear in the new legend to Figure 9 and add these profiles to the Supplement (they share similar patterns for all events).





Figure R7 (Figure S5 of the Supplementary Material): ECHAM6wiso profiles during event of the 9th of January 2020, used to design Figure 8 of the initial manuscript.

New caption for Figure 8 (now figure 9):

« **Figure 9:** Scheme of the mechanism explaining the sharp negative excursion of $\delta^{18}O_v$ recorded at the surface for cold front events associated with precipitation. The scheme is based on the profile modelled by ECHAM6-wiso for event of the 9th January 2020. The top panel show the altitude vs longitude dynamics of air masses with vertical saturated lifting in the center and subsidence at the rear of the lifting. The bottom panel shows the associated evolution of $\delta^{18}O_v$ and precipitations on the same longitude scale than on the upper panel. "

How is the subsidence at 100°E and ascent at 90°E related to the cold front?

The subsidence at 100°E seems to be linked to background conditions, while ascent at 90°E is caused by the cold front moving eastwards, causing precipitations just ahead of the front (Figures R4 and R7)

What does a composite of precipitation and d18Ov for all cold front events look like?

>> We already tried to make a composite before the first version of the manuscript but because the water $\delta^{18}O_v$ anomalies have different amplitude and durations (Table 1) and precipitation amount is very different from one event to the other (some event being associated with no rain, Table 1), a composite does not show any useful additional value.

Can it reproduce the schematic as shown in the "surface box"?

Because it is not possible to make a composite, the surface box of the schematic follows the surface state modeled by ECHAM for event of the 9th of January 2020 (Figure R7, lower right plots).

We propose this new version for the Figure 8:



Technical comments

Generally: there are many abbreviations in the text that are only used a few times. Can you reduce the number of abbreviations?

>> This is true and has been modified in the revised version.

Line 60-61 (and many others): The references are not in chronological order.

>> We changed it.

Line 65-66: "We express the abundance of the heavy isotopes D and 180 with respect to the amount of light isotopes 160 and H in the water molecules..." should be "We express the abundance of the heavy isotopes D and 180 with respect to the amount of light isotopes **H and 160**, **respectively**, in the water molecules..."

>> This has been modified

Line 68: Eq. 1 has strange symbols (squares).*

>> It is a problem from the word to pdf conversion, it can not be changed at this stage but should be possible when finalising the paper for production.

Line 88-89: "water cycle processes such as water cycle processes such as "

>> This has been corrected

Lines 106-109: "Over the previous years, we have installed 3 water vapor analyzers on Reunion Island at the Maido observatory (21.079°S, 55.383°E, 2160m) (Guilpart et al., 2017) and in Antarctica (Dumont d'Urville and Concordia; (Leroy-Dos Santos et al., 2021; Breant et al., 2019; Casado et al., 2016). " Check usage of brackets.

>> This has been corrected

Line 133: "from the nearest lands, Madagascar, and"

>> It has been corrected

Lines 140-141: "...and were continuously monitored at the site since 1960..."

>> This has been corrected

Lines 145-150: The section is very difficult to read, the websites and datasets should better be included as references. Same for link to AERIS on line 178 and 200.

>> datasets are now only included in the section « data availability » and we removed useless reference to program not used in this study.

Lines 180-181: "instrument models (Tekran Inc., Toronto, Canada) (Angot et al., 2014; Slemr et al., 2015, 2020; Sprovieri et al., 2016; Li et al., 2023). "

>>> This is simplified in the new version.

Line 202: "may possibly" Doubling, omit either.

>> We have removed "possibly"

Lines 211-221: This sentence is too long. Can you divide into two sentences?

>>> Sure, we have this new formulation:

"Chemical cycling and spatiotemporal distribution of mercury in the air is still poorly understood whatever atmospheric layer considered (surface, mixed or free troposphere, stratosphere), and complete GEM oxidation schemes remain still unclear (Shah et al., 2021 and associated references). Still, several studies provided evidence that vertical distribution of atmospheric mercury measurements from boundary layer to lower/upper troposphere and stratosphere shows a decreasing trend in GEM concentration with increasing altitude, in parallel with an increase in the concentration of divalent mercury (GOM + PBM) resulting from GEM oxidation mechanisms (Murphy et al., 2006 ; Swartzendruber et al., 2006, 2008 ; Talbot et al., 2007 ; Fain et al., 2009 ; Sheu et al., 2010 ; Lyman and Jaffe, 2012 ; Brooks et al., 2014 ; Fu et al., 2016 ; Koenig et al., 2023). "

Line 228: "The isotopic composition of near-surface water vapor (d18Ov and dDv in ‰ versus SMOW)"

>> It has been modified

Line 242: "The calibration of the data is performed in different **several** steps following previous studies"

>> It has been modified

Lines 301-303: "...identical **to** the atmospheric setup of IPSL-CM6A (Boucher et al., 2020) used for phase 6 of the Coupled Model Intercomparison Project (CMIP6, (Eyring et al., 2016)). "

>> It has been modified

Line 373: "... very close to the one observed in Angot et al. (2014)."

>> It has been modified

Line 374: "During the period (2020-2021) of water vapor isotope measurements in AMS..."

Do you mean: During the period 2020-2021 of water vapor isotope measurements in AMS..?

>> Yes, this has been changed

Lines 390-391: "The annual cycles are also not visible..." Do you mean: "An annual cycle is not visible..." ?

>> Yes, this has been changed