

We thank the reviewers for very useful comments. We provide below the answers to the different comments and will revise the manuscript accordingly. Note that part of the answers as well as the need for a more detailed explanation of the theory associated with the vaporization of the drop is provided in a companion paper which is now available online (Kerstel, 2020). We will also add one co-author, Morgane Farradèche, who realized numerous of the additional tests requested by the reviewers.

Reviewer 1

Technical comments: 1- This study represents an extension of the work shown in Landsberg (2014). However, in his study a strong influence of the lead screw rotation was identified as one of the main source of noise in the water vapor signal at low humidity. Is the choice of the pump a critical point for the development of the instrument? There are other critical points for designing/replicating this device that must be taken into account (e.g. how important is the choice mass flow and pressure controller? The authors should discuss this point e.g. in section 2.2.

Indeed the choice of pump is important. First of all because it needs to provide an exceedingly small water flux, and secondly because it needs to do so with high stability. It is therefore noted that the oscillations that are in-phase with the lead-screw rotation were seen by Landsberg (2014) in an early prototype and were eliminated by better engineering, and in particular by the careful elimination of air-bubbles in the liquid water supply lines and syringe. This aspect is also addressed in our companion paper (Kerstel, 2020).

The precision of the pressure controller is not critical. Its purpose is to provide a steady inlet pressure to the optical spectrometer, also during syringe switching. Ideally the spectrometer is not sensitive to the inlet pressure.

The precision of the flow controller on the other hand does directly influence the precision of the volume mixing ratio (humidity level) produced by the instrument. Its precision ranges from about 0.5% at the highest flow setting, to almost 50% at the lowest air flow setting. In practice the device is used with an intermediate air flow of at which the precision of the controller is specified to be 1%. We observe, however, that in practice its short-term precision is much better. In any event, a precision of the order of 1% is normally comparable to the precision of the measurement of the humidity level with the optical spectrometer.

We will modify the text accordingly to provide quantitative information on these issues and cite the companion paper.

2- Since large stability of water vapor flux is expected by such device, one would expect an analysis of the stability of water vapor signal, at least in terms of the mixing ratio. An analysis of stability could be the identification/absence of trend during different humidity steps or the analysis of mixing ratio standard deviation as a function of different instrumental configuration (e.g. dry air flux, syringe speed etc). A stability analysis would provide additional evidence of the robustness/reliability of the instrument.

This issue was addressed through two different tests. First, we now present Allan variance obtained over 4 hours at different humidity levels by varying the syringe speed (Figure R1).

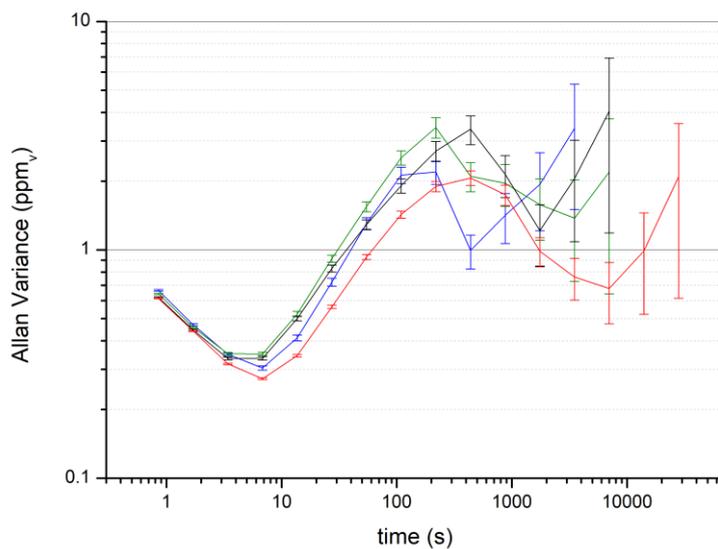


Figure R1: Allan variance over 4 hours for different humidity levels (blue 1080 ppmv; green 770 ppmv; black 400 ppmv; red 320 ppmv).

Second, we also test the stability of the system by varying the flow of dry air from 200 to 400 sccm (Table R1). The reported standard deviation (Table 1) is over a period of 10 minutes (last 10 minutes of the plateau) but the humidity and $\delta^{18}\text{O}$ values were stable over one hour (the experiment was stopped after one hour of stable humidity). We notice that the generated $\delta^{18}\text{O}$ values does not depend on the dry air flow nor on the infusion rate.

Air flow (sccm)	Infusion rate ($\mu\text{L}/\text{min}$)	Humidity (ppmv)	stdev humidity over 10 minutes (ppmv)	$\delta^{18}\text{O}$ (‰)	stdev $\delta^{18}\text{O}$ over 10 minutes (‰)
200	0.07	808	1	-7.88	0.89
300	0.11	851	2	-7.73	0.85
400	0.14	818	2	-7.95	0.90
200	0.03	374	1	-8.45	1.92
300	0.05	411	2	-9.16	1.64
400	0.07	415	3	-9.05	1.59

Table R1 : Evolution and stability of humidity and $\delta^{18}\text{O}$ (same water used for the different tests) for different syringe infusion rates and dry air flows.

These results have been implemented in the revised manuscript.

Other minor comments

L136 Period.

L147-148 What is the reason for using fractionation factors of Cappa et al. (2003)?

In this paper we are interested in the steady-state behavior of the device. In this case, the isotopic composition of the incoming liquid water is by mass balance considerations equal to the isotopic composition of the evaporated water flux. The fractionation factor between the liquid and vapor phase is thus of no importance for the work presented here. The previously mentioned companion paper, however, deals with the dynamic behavior of the humidity generator and discusses the fractionation factors in detail. There we use estimations of the fractionation factors that are based on the studies by Cappa et al. (2003) and Luz et al. (2009). The work concludes that the effective fractionation factors are largely determined by their equilibrium counter parts, already determined with high precision by Horita and Wesolowski (1994), combined with the ratio of diffusivities and a parameter that describes the flow regime (from laminar to slightly turbulent). The diffusivities were determined by Merlivat (1978), Cappa et al. (2003) and more recently by Luz et al. (2009). We used the values by Cappa et al. (2003), but the difference is minimal for our purpose. Cappa et al. (2003) predicts only slightly lower values of the effective fractionation factor in the laminar limit compared to other estimates.

As the fractionation factors are not necessary for this study, we will remove the reference to Cappa in this study. The dynamical aspects are dealt in the companion paper.

Figure 2. Check part labels (A/B) because different names are used in text, in table 1 (F1, G1 etc..) and in Figure 3.

>> The scheme has been fully changed, see answer to reviewer 2 and associated Figure R3.

L245 In relationship with my technical comment #2: what stable means? No trend observed in mixing ratio? Low RSD?

>> Indeed, stable means both no trend in the mixing ratio (< 20 ppmv/hour) and low RSD (< 10 ppmv over 10 minutes). This will be explained in the revised manuscript.

Figure 4. It is not clear how long it takes the signal to stabilize and whether this stabilization period is related to the humidity level. From Fig.4 it seems so, because injection steps are characterised by different lengths.

>> Actually, it can be variable between a few minutes only to 30 minutes in worst cases. In best case, we have stabilization in a few minutes but it may take more time for undetermined reason. This is the reason why, in routine mode (i.e. when the user is not checking visually the stability of the plateau), we run 50-minute long plateau. This will be explained in the revised text.

Section 3.2 Maybe, two words here (or a simple scatter plot, boxplot) about a possible relationship between precision and humidity level could help the reader understand the stability of the system.

>> There is actually no relationship between precision and humidity level.

Section 3.3, Figure 5 and Section 4: humidity correction is important. I would like to point out to the authors that a correction based on the difference between observed and reference values for a single standard might not be enough, as recently highlighted out in Weng et al. 2020. Under the perspective of the influence of both humidity and isotopic composition, the correction function should represent a plane and not a line (as one could expect from Figure 4). Moreover, from Figure 4 it is not clear if the scatterplots reports the difference between a single standard (obs-ref) or for different standards (with different isotopic composition) because the plural "standards" word was used. Maybe the authors could highlight that under low humidity and low isotopic

variability, the humidity response of the instrument can be determined by analysing one (or two) standard(s).

Indeed, we paid attention to this calibration issue and ran several standards. For standards with low $\delta^{18}\text{O}$ values (-30 to -55 ‰) used for calibrating our instrument in Antarctica, the relationship between $\delta^{18}\text{O}$ (δD) and humidity is the same which makes the calibration of the laser analyzer easier in this region. On the contrary, when we use a less depleted standard ($\delta^{18}\text{O} = -9$ ‰, $\delta\text{D} = -43.6$ ‰), the relationship between δD and humidity is not the same as displayed on Figure R2.

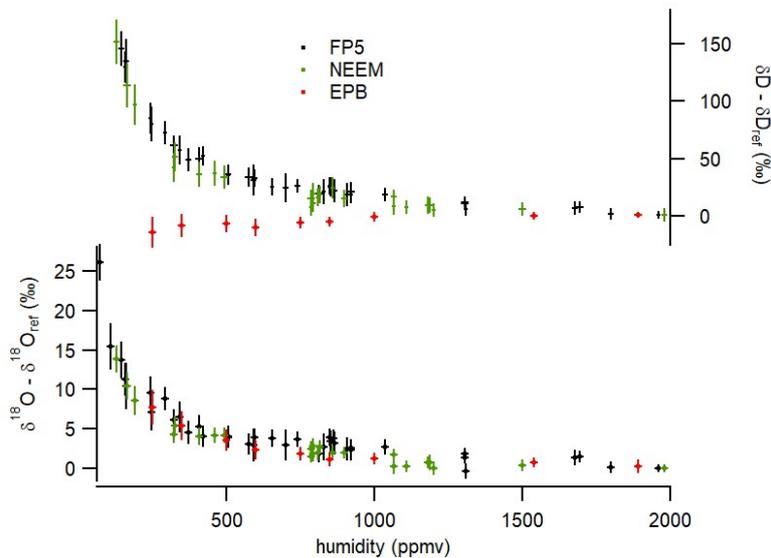


Figure R2: Relationship between humidity and $\delta^{18}\text{O}$ or δD (when corrected from the reference value, i.e. the value of the standard measured at 2000 ppmv). VSMOW-VSLAP calibrated values of the standard used for this plot: EPB ($\delta^{18}\text{O} = -6.24$ ‰; $\delta\text{D} = -43.6$ ‰), NEEM ($\delta^{18}\text{O} = -33.50$ ‰; $\delta\text{D} = -257.2$ ‰), FP5 ($\delta^{18}\text{O} = -48.33$ ‰; $\delta\text{D} = -383.5$ ‰).

Reviewer 2:

Remarks

2.1 Evaporation Model

The theoretical foundation of the vapor generator presented here is described in section 2.1 of the manuscript. It is based on the assumption that a constant flow of water ending on a syringe needle tip combined with a finely controlled flux of dry air will produce an isotopically stable stream of vapor via a zero fractionation process. A prerequisite for this, is that the size of the droplet remains stable throughout the experiment. It is still a question to me how a droplet that sustains its size by constantly losing mass on its surface (regained by the incoming liquid water flow) via evaporation yields a vapor stream that has the same isotopic composition as the liquid. This is a typical Rayleigh evaporation likely with a strong kinetic component due to the very low humidity of the carrier gas and the quick -but incomplete- removal of water molecules from the droplet. Even though the treatment of the model has been presented in previous works, this is so central to this system that at least an appendix with more information is needed.

We thank the reviewer for raising the question as it is indeed at first sight not immediately obvious that we can be certain that no fractionation factor is involved in the determination of the isotopic composition of the output vapor. We have therefore decided to submit a companion paper that describes and validates a model of the dynamic behavior of the humidity generator (Kerstel, AMTD, 2020). This manuscript is still in the reviewing process. In short, the mass balance model convincingly demonstrates that the isotopic composition of the generated vapor is identical to that of the incoming liquid, in steady-state. It also shows that in steady-state the droplet volume is constant and that the enrichment occurring at the surface of the droplet due to evaporation of its surface water is compensated by an inward diffusion of heavy isotopologues. This diffusion process, however, is frozen in place by the continued renewal of water arriving in the droplet through the syringe needle. This is important, as otherwise the bulk syringe water would become enriched. Measurements performed on the syringe water before and after several hours of operation show that this is indeed not the case, as predicted by our model.

We will add a brief discussion to this issue in the text and refer to the companion paper for more details and insight. This also addresses the reviewer's request for an appendix, which we have thus decided to replace by the companion paper.

2.1.1 Some points to consider

- How is the size of the droplet controlled when there is no camera or some other monitoring tool inside the evaporation chamber?

The humidity level generated by the device is controlled in feed forward by the ratio of water to air flow. The droplet size simply follows. There is thus no need to actually measure the droplet size. This said, an early prototype contained a camera that allowed us to produce the photograph in Figure 1 in the initial manuscript.

- The manuscript mentions in Line147 that the fractionation factor by Cappa et al (2003) are used. But why if there is no fractionation?

We thank the reviewer for pointing out that the mention of Cappa, or other fractionation factors, is superfluous here.

We will eliminate the mention of Cappa et al. in the revised version of the manuscript, and instead refer to the companion paper for a discussion of the dynamic behavior of the device.

- The manuscript claims that under steady-state the isotopic composition of the generated vapour does not depend on the injected flux of water nor the specific humidity. Since this is a technical paper I would appreciate a simple experiment where the flux of the carrier gas is ramped up and down allowed to reach steady state. I have a very hard time seeing how this experiment will produce a vapor stream of the same isotopic composition equal to the $_{18}\text{O}$ of the liquid water. Similarly if the specific humidity of the carrier gas is changed one ends up with a simple mixing experiment (Mook , 2000) where the resulting vapour isotopic composition naturally will be different.

See previous answers for this point. We also followed the suggestion of the reviewer and run experiment with different dry air flows. Because the air is dry, it is not expected to modify the isotopic composition of the produced water vapor if the humidity is the same. This is exactly what is observed in Table R1.

- Throughout the whole manuscript, I have not seen a note on the temperature of the evaporation. This is a critical parameter affecting the efficiency of the evaporation, the saturation vapour pressure of the mixture and the fractionation factors in the (Cappa et al , 2003) parameterization. Since there is no active control and regulation of the temperature, all these parameters will vary.

We thank the reviewer for bringing up this issue, as the temperature is indeed a factor to consider for several reasons. Temperature variations affect the stability of the device by their effect on the stability of the flow regulator, which in turn affects the stability of the humidity level. Our flow regulator having a temperature sensitivity of $0.05\% / ^\circ\text{C}$, this factor is deemed not critical. Temperature also intervenes through its effect on fractionation and the evaporation rate, as mentioned by the reviewer. Therefore, it can force a departure from steady-state operation, which in turn would drive isotope variations. For these reasons, the device is carefully insulated, leading to a temperature of the evaporation chamber of about 20°C varying within 1°C (over 24h).

2.2 Presentation–Standard nomenclature

Any physicist or engineer with moderate training in laboratory instrumentation should be able to look into the flow diagram of a manuscript in AMT or any other equivalent journal and get a basic idea of the method described in it. It is a very important element of a publication of this type, therefore it is my view that the authors should use standard P&ID nomenclature. The current flow diagram is a collection of coloured boxes from which little can be understood. Part of the text uses the photograph supplied (Figure 3) to explain the system something that confuses the reader even more.

2.2.1 Points to consider

- There are standard P&I symbols for valves, pumps, gauges etc that should be used, while colour should be added only if it aids in better explaining the system and not cosmetically. Standard names for the components should be used. For example three pumps with three different descriptions (pressure pump, pump and picarro pump) are found in the block diagram when they (most likely) are of the very same type ie vaccum pump.

The scheme initially presented on Figure 2 has been drawn again as presented in Figure R3 below.

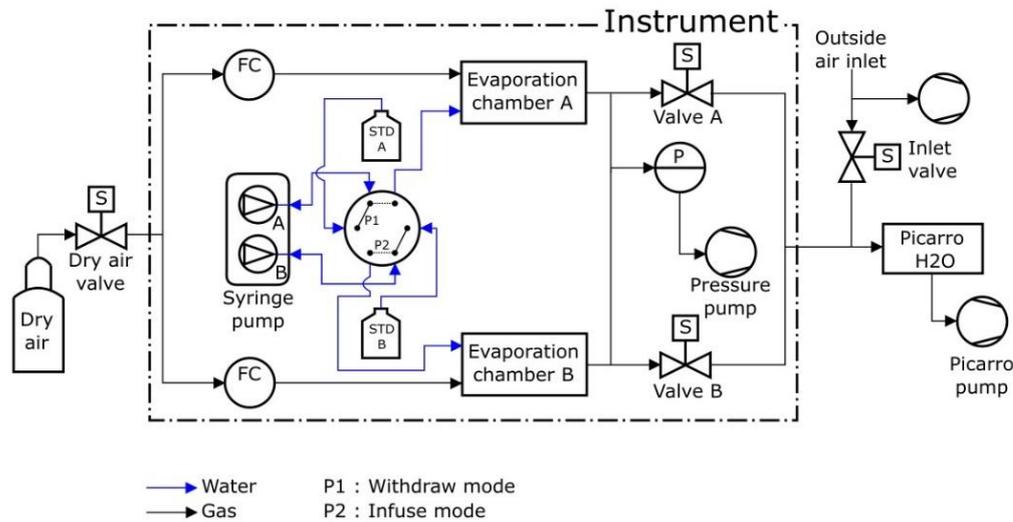


Figure R3: Scheme of the humidity generator to replace figure 2 of the initial manuscript.

• One of the main elements of the paper according to the authors is the “double 3-way valve” added to the system. The valve’s type is a 6-port switching valve and a standard symbol for it exists that should be used in this manuscript. It is central to describing the cycles of the system. Feel free to use the supplied (arguably not perfect) Fig. 1 that I am including and describes the valve operation for the syringe filling position.

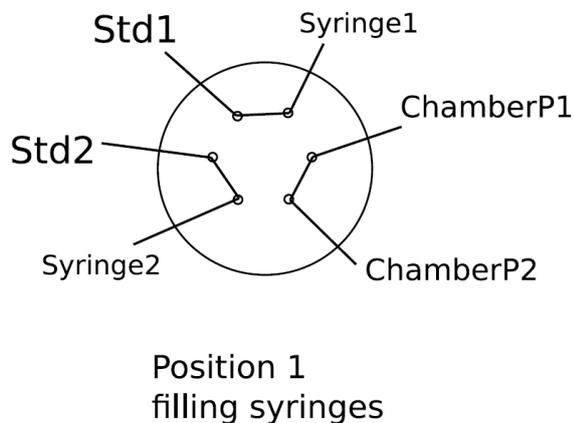


Figure 1

As suggested by the reviewer, we now use the 6-port valve symbol in figure R3.

• Flows and pressure set points for the regulators also belong to the P&I diagram and since the authors claim that they are important to the operation of the system should be given. A table with the ID numbers of the control units and their set points would be very much appreciated. Currently Table 1 blends names of components from Figure 2 and Figure 3 so the reader has to guess. This is not informative.

In order to better document the system, we propose to add the following tables R2 and R3.

Instruments	Notation on Figure R3	Set points	Accuracy
Vögtlin GSC-A9TS-DD22	FC	300 and 150 sccm	3.3 sccm
Harvard Apparatus Pump 11 Pico Plus Elite Dual	Syringe pump	0.01 to 0.3 $\mu\text{L}/\text{min}$ using 100 μL syringes	0.35% of the set speed
Hamilton syringes	A and B	100 μL	
Swagelok Ultra-Torr SS-4CD-TW-25	Evaporation chamber A and B	Internal volume of 25 cm^3	
Bronkhorst P-702CV-1K1A-AAD-22-V	P	650 to 950 mbar	3 mbar
KNF N86KNDC 24V	Pressure pump		

Table R2: Description and setting points of the instruments

Humidity (ppmv)	Infusion rate ($\mu\text{L}/\text{min}$)	Dry Air flow (sccm)
80	0.01	300
160	0.02	300
320	0.04	300
800	0.1	300
1200	0.15	300
1600	0.2	300

Table R3: Set points for water infusion rate and dry air flow at a temperature of 20°C

2.3 Experimental System—Explanation of operation

Section 2.2 of the manuscript contains information on the principle of operation. One of the puzzling pieces of information in this section is the carrier gas flow. Following the block diagram and the information in Table 1, we see that for the “Drying Mode”, the Flow Controllers A and B sustain each a 400 sccm₁ flow through the system. The block diagram does not indicate any open splits and for this particular mode the inlet valve is closed and the exhaust vacuum pump is disabled. This is an immense flow for the standard of a picarro spectrometer that normally can sustain its cavity pressure with sample flows in the order of 30-100 sccm₁.

Similarly, during the injection of Standard A or Standard B, the flow is in the order of 450 sccm₁, a very high flow level. In this mode though the exhaust vacuum pump is enabled, however we see that the lines of Standard A and Standard B are connected. Thus in Standard A mode, valve B is closed and the generated vapour from standard B, hopefully is evacuated via the exhaust vacuum pump. Is the pressure of 905 mbar enough to make sure that all the 150 sccm₁ of standard B are off the system and not mixed with the vapour from Standard A? Why was this value chosen and what kind of tests assure that the two lines are never mixed even though physically connected. The safe way to proceed here would be to simply isolate the two channels, remove the pressure regulator and the vacuum pump and simply install two 3-way valves in the position of valves A and B (which I assume are On/Off???) on which one port goes to the instrument inlet and the other is simply open to the atmosphere in an open split configuration.

2.3.1 Points to consider

- A more thorough description of the flow path is needed and the issue of possible cross contamination between Standard A and B should be addressed.

The instrument has been designed with tubing long enough to prevent any mixing and the issue of mixing has been extensively tested during conception by comparing the isotopic composition of the produced water vapor when only one way was in operation (bottle A filled with standard and bottle B empty) and when two ways were in operations (bottles A and B filled with standards of different isotopic composition). the isotopic composition produced on way A was always the same.

To double check this effect when receiving this review, we performed an additional comparison:

- Case A: bottle B was filled with FP5 standard ($\delta^{18}\text{O} = -48.33 \text{ ‰}$) and bottle A filled with EPB standard ($\delta^{18}\text{O} = -6.24 \text{ ‰}$). Measured $\delta^{18}\text{O}$ on way B was $-48.96 \pm 1 \text{ ‰}$ (humidity $768 \pm 5 \text{ ppmv}$)
- Case B: bottle A and B were filled with the FP5 standard. Measured $\delta^{18}\text{O}$ on way B was $-49.05 \pm 1 \text{ ‰}$ (humidity $765 \pm 4 \text{ ppmv}$).

There is thus no difference between the two cases both for $\delta^{18}\text{O}$ and δD which confirms that there is no mixing in the system. We will explain these tests and the absence of mixing effect in the revised version of the manuscript.

- The flows are very high - How does the picarro cope with this condition?

There is a waste line on this instrument (corresponding to the outlet of the pressure pump) so that the Picarro laser analyzer is only pumping what it needs through valves A and B as it does during the analysis of atmospheric air through inlet valve. As a consequence, there is no problem for the system. This will be mentioned in the revised version of the manuscript.

- There is no information on the volume of the evaporation chambers and no mention regarding the temperature of the system.

Volume of the evaporation chamber is 25 cm^3 (see reference of the chamber on Table R2). Temperature is around 20°C .

- There are references to a two standard calibration protocol in this section. It is not exactly clear if this concerns some water concentration correction calibration or a linear slope SMOW-SLAP calibration—see specific comment on the notable absence of any reference to SMOW-SLAP below.

All the lab-standards used in this study are indeed calibrated versus SMOW using a 3-point calibration which will be mentioned in the revised version of the manuscript. Then, we used two ways for our generator in order to perform two-point calibrations of the laser analyzer when it is used to measure water vapor isotopic composition at low humidity. We hope that the answers provided for the other comments below now explain better this aspect and it will also be implemented in the revised manuscript. This is indeed very important to mention.

- I assume that the system is also used in order to perform a SMOW-SLAP calibration. How are evaporation/fractionation effects in the standards' vials handled? Based on Figure 3 we see no precautions concerning this.

Thank you for this comment. Indeed, this was not mentioned in the manuscript. This has been tested by sampling the water from the flasks every 2 weeks. We observed a maximum evolution of the isotopic composition of the standard in the bottle by 0.05 ‰ for $\delta^{18}\text{O}$ and 0.5 ‰ in δD within 2 months in case of intense use of the instrument. This will be mentioned in the new version of the manuscript.

2.4 Evaluation–Measurement Stability and precision

The manuscript lacks important information on the evaluation of the performance of the system. Section 3.1 assures the reader that no fractionation is observed during the generation of the vapour and its transfer in the flow lines while the agreement with the commercial standard delivery module is within 0.2 and 1‰ for $_{18}\text{O}$ and $_{\text{D}}$ respectively. It is almost impossible to judge these discrepancies based on figure 5 and in fact when looking closely it is rather difficult to see how the differences between the two systems are of this order.

With respect to figure 4 and the evaluation of the precision for $[\text{H}_2\text{O}]$ and $_{18}\text{O}/_{\text{D}}$ I strongly believe that the manuscript should include a proper Allan variance test (Werle et al, 2011; Steig et al, 2014). It is obvious that the system experiences drifts, whose origing is the vapour generator. Calculating the standard deviation on a 10 min window of a time series that obviously has a changing mean value looks and feels like possible cherry-picking.

2.4.1 Points to consider–Suggestions

- Show better evidence of zero fractionation. If you choose the SDM as a measure of comparison show a detailed comparison and plot the results clearly. The axes of fig 5 span 300 ‰ for $_{\text{D}}$. It is impossible to say anything.

In addition to the current text which already mention the detailed comparison on l. 250 to 253, a focus on the comparison between the SDM and the low-humidity level generator is displayed on Figure R4 below. This figure will be implemented in the revised manuscript.

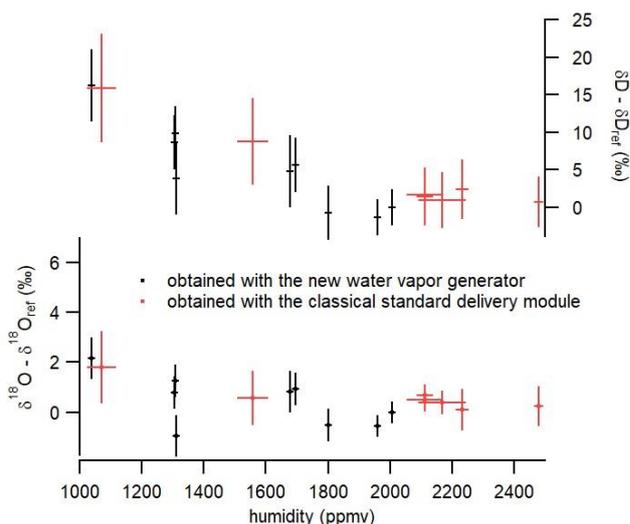


Figure R4: Comparison of the difference in isotopic composition (δD top, $\delta^{18}\text{O}$ bottom) between the measured and the reference (V-SMOW calibrated) values as obtained by the SDM (red) and the low-humidity level generator (black) coupled to the same Picarro L 2130-i with the same lab-standard (FP5).

- Calculate the Allan variance of the system for 4-5 $[\text{H}_2\text{O}]$ levels. Let one water run

for several hours and so you get a more honest idea about precision and drifts for all three parameters under consideration. It is a very simple experiment that can provide a valuable insight and by looking in the bibliography it is a rather standard evaluation tool for laser spectroscopy based measurements.

Thank you for the suggestion. Indeed, this is a good idea and we propose to include the figure R1 (see answer to reviewer 1) in the revised manuscript

2.5 Water concentration correction

The water concentration correction is described in section 3.3 with Figure 5 being the main source of information for this part of the analysis. I would consider this one of the most important sections in the paper and unfortunately it spans only one paragraph. It is repeated in the manuscript that the system presented here is superior to the commercial standard delivery module below the level of 500 ppm. How can we see this when the SDM measurements in this are of [H₂O] are not presented (measurements stop at [H₂O]₂₀₀₀ ppm)?

It is very difficult to get stable humidity plateau with a SDM below 500 ppmv. This is the reason why we could not display them. This will be mentioned in the new manuscript.

The dataset in Figure 5 presents the difference of the raw isotopic value and a value that the authors call “reference” and “real”. These words commonly refer to very specific things in isotope geochemistry and my guess is that the reference value is a SMOW-SLAP calibrated value (?). The term SMOW-SLAP calibration is not to be found at all in the manuscript. It is to some extent problematic that a water concentration correction measurement compares a raw value at a number of [H₂O] levels with a value post SMOW-SLAP calibration. Please see my comments on SMOW-SLAP in the next section.

As mentioned above, the SMOW-SLAP calibration will be addressed in the revised manuscript and all our lab-standards are calibrated vs V-SMOW (using also V-SLAP as a reference).

The type of correction is not described in the manuscript. This is what this work is all about. There can be various approaches on how it can be done (see for example (Schmidt et al , 2010; Gkinis et al , 2010; Aemisegger et al , 2012)) but the authors need to be open and specific about what they did exactly. Moreover (Schmidt et al , 2010) suggest that the response to changing [H₂O] levels varies with the isotopic composition. I do not know how important this effect is for this present work, but I believe that the authors should perform two separate experiments with two different waters (technically the system offers this possibility). It would also strengthen their claims for a very stable system if they show the full experiment with its raw data as a function of time and not only the averages.

There is also a claim in this section that the stability of the system allows via the daily measurement of standards waters to quantify instrumental drifts. There is no strict evaluation of the stability of the system so far and I think that a proper Allan variance test at various levels of [H₂O] is necessary. But even in the absence of an Allan variance test the data given in Table 2 do not necessarily indicate instrumental drifts. All the isotope data in the table are within 1 σ of the noise level for a 10-min period mentioned in the caption and there does not seem to be a one way trend. So how is it possible to say anything about instrumental drifts? Also the noise levels

mentioned in the caption of Table 2 are very different for roughly the same [H₂O] when one looks at the data of Figure 4 (4.5 versus 14).

This is a good remark indeed and we are sorry for the confusion. Actually, while the noise on the humidity level is mainly due to the low-humidity level generator, the noise on the isotopic ratios depends on the laser analyzer which explains some inconsistency in the results which were displayed (we presented results obtained with different instruments). The inconsistencies will be fixed in the revised version. Indeed, Table 2 shows that there is no measurable drift and this will be explained more clearly in the revised version.

2.5.1 Points to consider-Suggestions

- A clear description of the correction is missing. Some math is needed here.

We propose to provide the following explanation in the revised manuscript.

In order to correct the dependence of the isotopic composition ($\delta^{18}\text{O}$, δD) to humidity, we use the LHLG with two lab-standards with isotopic composition ideally bracketing the isotopic composition of the water vapor measured by the analyzer. In our case, we used our two lowest lab-standards and obtained for the two standards the same dependency of isotopic composition vs humidity (in the case of Antarctica, we use the NEEM and FP5 standards, Figure R2). We express this dependency as the relationship between the difference in δD or $\delta^{18}\text{O}$ between the measured value at the given humidity and the value of the same standard measured at a humidity of 2000 ppmv. The experimental data from figures 5 and R2 are fitted through a polynomial function with respect to humidity h (in ppmv):

$$\delta^{18}\text{O} - \delta^{18}\text{O}_{\text{ref}} = 0.0000000000000000397 \times h^6 - 0.00000000000003586315 \times h^5 + 0.00000000012843645994 \times h^4 - 0.00000023087753445094 \times h^3 + 0.00021857285350473100 \times h^2 - 0.10603325432255400000 \times h + 23.7 \quad (\text{eq. 1})$$

$$\delta\text{D} - \delta\text{D}_{\text{ref}} = 0.00000000000000006859 \times h^6 - 0.00000000000060047709 \times h^5 + 0.00000000207903313490 \times h^4 - 0.00000361319302207374 \times h^3 + 0.00330716141498371000 \times h^2 - 1.53651645114701000000 \times h + 313 \quad (\text{eq. 2})$$

A second important correction step is to take into account the slope of the VSMOW calibrated δD and $\delta^{18}\text{O}$ on the Picarro laser analyzer vs humidity-corrected measured $\delta^{18}\text{O}$ and δD . Depending on the instrument used, the slope lies between 0.96 and 1, cf Table R4 below for an example.

- An experiment at two different isotopic levels showing the raw data versus time will show if there is an isotope effect in the water concentration dependence.

Thanks for this suggestion. We will display the tests performed in figure R2 (see also corresponding answer to reviewer 1).

- A proper quantification of instrumental drifts (and this concerns the system as a whole and not only the spectrometer) can be done with a proper Allan variance test.

This is the aim of figure R1 which will be implemented in the revised version of the manuscript.

2.6 Lack of reference to SMOW-SLAP calibration

The main goal of building a water vapour generator as a peripheral for isotope measurements of water vapour is to be able to calibrate the dataset on the SMOW-SLAP

scale. This is the only way to communicate and compare the measurements with other existing data sets and produce some science out of them. It is also even more important if the deuterium excess parameter will be studied as it is very sensitive to this calibration procedure. Therefore it appears very awkward that a manuscript dealing with this topic does not include a single note, comment or reference to this very important step of the measurement process. The dataset presented later in section 4 of the manuscript are impossible to evaluate if they are not calibrated in the SMOW-SLAP scale.

One more purpose of performing such calibrations, is that they can reveal possible accuracy issues in the instrumentation system. Given two standard waters one should be able to produce a calibration line and thereafter measure a third water of known isotopic composition treating it as an unknown. If the resulting value lies beyond the 3 σ range then there is likely something wrong with the system. That could be any part from the water standard storage to the water vapour generation system or the spectrometer itself. Currently there is no way to say anything about the accuracy of the system. With this in mind, section 4 of the manuscript is of very little use as the dataset is reported on some local instrument scale.

A SMOW-SLAP calibration experiment at various [H₂O] levels using the SDM and the current system would provide a proper comparison between the two systems and therefore it would be a very important addition to the manuscript.

2.6.1 Points to consider

- A proper treatment of the SMOW-SLAP calibration step is notably missing.

We will explain in the revised manuscript and in the caption of figure 6 that all our lab-standards are regularly calibrated vs SMOW using IAEA VSMOW and VSLAP standards (every 3 years in minimum). In order to calibrate our lab-standards, we use determination from Picarro L 2130-i for δ D and for δ^{18} O. In addition, we use a δ^{18} O calibration using an IRMS through water – CO₂ equilibration.

- Performing 2-standard calibrations and measuring a third water standard treated as an unknown will be a valuable -almost essential- addition to the manuscript, offering important information on the accuracy of the system.

This is an excellent suggestion and this test has been performed as follows:

Standard	Calibrated value (VSMOW – VSLAP)	Measured value at 800 ppmv	Measured value corrected from humidity dependence (Equation 1)
EPB	-6.24 ‰	-8.27 ‰	-10.78 ‰
NEEM	-33.5 ‰	-34.48 ‰	-36.99 ‰
FP5	-49.11 ‰	-49.02 ‰	-51.53 ‰

Table R4: Comparison of measured vs VSMOW-VSLAP calibrated δ^{18} O values for 3 standards measured with a Picarro analyzer after generation of water vapor using the low-level humidity generator.

We used the measured and true values of EPB and FP5 to estimate the δ^{18} O value of the NEEM standard from its measured value. Using the linear relationship obtained from VSMOW-VSLAP calibrated EPB and FP5 δ^{18} O vs measured EPB and FP5 δ^{18} O values ($\delta^{18}O_{cal} = 1.0329 * \delta^{18}O_{meas} + 4.8965$) leads to an estimated NEEM δ^{18} O of -33.31 ‰.

Another approach because EPB and FP5 $\delta^{18}\text{O}$ are very different, is to scale the isotopic composition using the isotopic ratios $(1+\delta)$ so that:

$$(1+\delta^{18}\text{O}_{\text{cal, NEEM}}) = (1+\delta^{18}\text{O}_{\text{meas, NEEM}})/(1+\delta^{18}\text{O}_{\text{meas, EPB}})*$$

$$[(1+\delta^{18}\text{O}_{\text{cal, FP5}})/(1+\delta^{18}\text{O}_{\text{cal, EPB}})]/[(1+\delta^{18}\text{O}_{\text{meas, FP5}})/(1+\delta^{18}\text{O}_{\text{meas, EPB}})]*(1+\delta^{18}\text{O}_{\text{cal, EPB}})$$

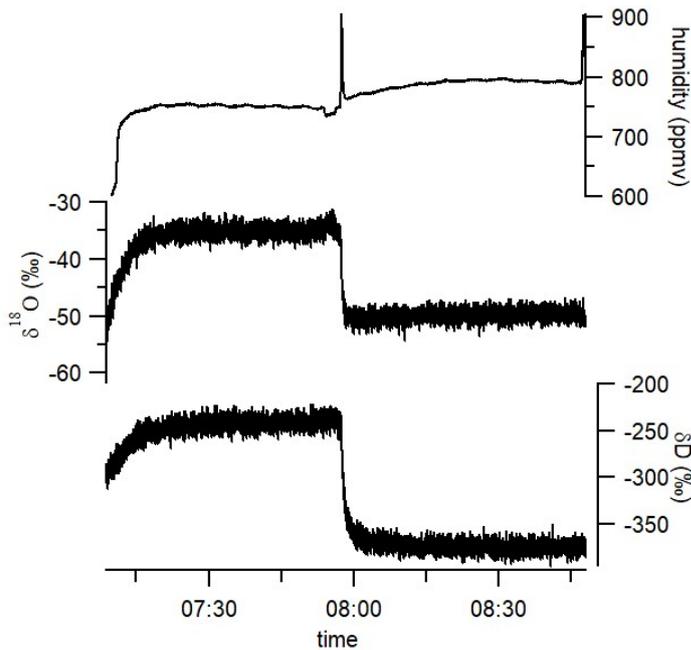
With this approach, the estimated NEEM $\delta^{18}\text{O}$ value is -33.74 ‰.

Given the uncertainty of about 0.8-1 ‰ when measuring $\delta^{18}\text{O}$ around 800 ppmv, we can conclude that the system is accurate.

- Since a lot has been written about the performance of the commercial SDM it will be proper to perform 2-standard calibrations for various $[\text{H}_2\text{O}]$ levels and compare the results.

See answer above as well as the Figure R4.

We also propose to display the comparison between SDM and the low-humidity level generator (Figure R5) which shows the high stability of humidity with the new instrument compared to the SDM.



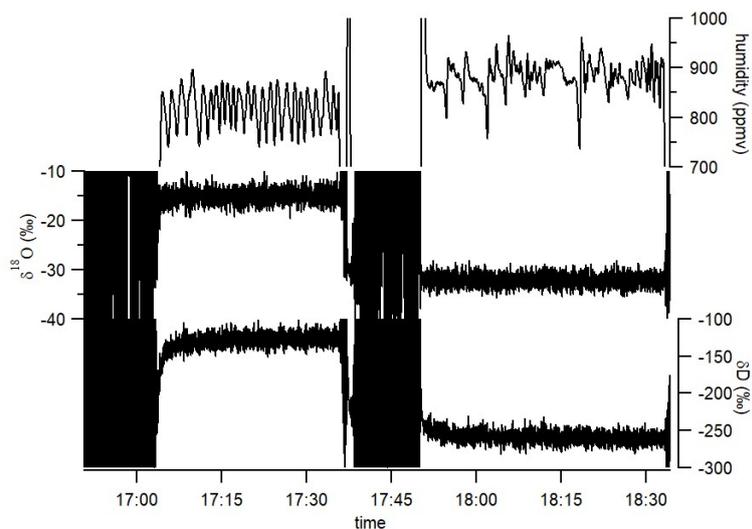


Figure R5: Comparison of humidity plateaus (800 ppmv) generated with the low-level humidity generator (top) and with the SDM (bottom).

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