

Review of “A flexible device to produce a gas stream with precisely controlled water vapour mixing ratio and isotope composition based on microdrop dispensing technology”

This paper presents a new device based on two microdrop dispenser heads that allows to inject a controlled amount of water with known isotopic composition into a heated evaporation chamber flushed by a dry carrier gas for subsequent analysis with a laser spectrometer. The authors argue that the novelty of this device is its flexibility in terms of producing air samples with a range of different water vapour mixing ratios and isotope compositions varying between the two end-members of the liquid water samples feeding the two dispenser heads. The authors show two applications of the new device: 1) the detailed characterisation of the well-known water vapour mixing ratio dependence of isotope measurements with laser spectrometers and 2) the production of a stable background water isotope signal for the precise measurement of microfluid inclusions in stalagmites.

The paper is interesting and well-illustrated. The major innovation is the combination of two dispenser heads instead of one, which gives more flexibility in the generation of water vapour samples with a range of different isotope compositions and water vapour concentrations. While most of the text is well-written and easy to follow, there are a few parts that are written in a misleading way, where I suggest that the reader guidance and accuracy of the description could be improved (see my minor comments below).

We are very grateful to the reviewer for their attentive reading of our manuscript, and the detailed and specific constructive comments.

The paper should be published in AMT after the following two major points and a longer list of minor comments as well as a few technical aspects have been addressed:

M1: Given that the key innovation of the proposed system is based on the combination of two dispenser heads for being able to produce a series of gas streams that cover a range of different water vapour mixing ratios and isotope compositions, a solid assessment of the uncertainty of the predicted reference properties ($[H_2O]$, δ^2H , $\delta^{18}O$) should be provided. Currently, this aspect is very difficult to assess. From Fig. 8 it seems that the RMSE of the obtained mixtures of standards is very large and therefore the system's characteristic uncertainty in the reference isotope composition and mixing ratio when using the two dispenser heads simultaneously seems to me an important issue.

This is a very valuable comment, we agree that the aspect of the uncertainty of the mixtures is important in the context of this device. We will add a section/figure to make this aspect more readily accessible in the revised manuscript.

M2: I am not convinced that the relatively complex water vapour mixing ratio – isotope dependency correction is really needed in the second application for micro fluid inclusions in stalagmites, because the water vapour mixing ratio of the background airstream is well above 10'000 ppmv and the variations due to the microfluid injection peak is in a range of values where this correction is not usually needed. Could the authors show the impact of this correction on their aliquot of a natural carbonate and/or their samples used to mimic real fluid inclusions? It's a major point because the authors argue that this is an important additional processing step that was neglected in the previous literature (L. 430-437).

Fluid inclusions contain a variable amount of liquid in the samples. Therefore, the mixing ratio in the background air stream has to be adjusted, and can not always be at 10'000 or even 20'000 ppmv, but may have to be for example as low as 5000 ppmv. With the strong variations that occur during the crushing procedue in mixing ratio, there can indeed be where a significant impact in the mixing ratio - isotope ratio dependency. Furthermore, the stability of the mixing ratio is important for this application to separate the background water vapour clearly from the water vapour released from the crushed sample. We will consider the best way to provide more support to this point, either from acutal sample processing, or from a simulation of the impact for hypothetical samples.

Minor comments:

1) L. 1, L. 22 and at other places: “a flow of air-vapour mixture” is a bit confusing: not clear what vapour of which gas and air does a priori comprehend water vapour as well. The terminology used in the title seems more precise to me: “a gas stream of a pre- determined water vapour mixing ratio and isotope composition”.

We will clarify the terminology upfront for the remainder of the manuscript in order to be both precise and avoid excessive wordiness,

2) L. 6: “water vapour mixing ratio”, just mixing ratio can be any gas or isotope.

We will correct this according to the reply to comment 1) above.

3) Abstract: the abstract is a bit short and lacks key information on the new device. It would be more informative to add a few key numbers in the abstract such as the range of flow rates tested, explain what limits the flow rates in the device (key for more recent fast-response CRDS instruments operated at high flow rates). The range of water vapour mixing ratios and isotope delta values tested as well as the quality of the delivered calibration or background gas stream could be mentioned (precision, stability).

While we want to avoid too much detail and complexity in the abstract, we will include several of the specific aspects mentioned by the reviewer.

4) L. 15: I think Graf et al. 2019 is a nice example for subcloud processes involved during precipitation but not weather systems in general. Thurnherr et al. 2020 cover a broad range of latitudes and different types of weather systems, which would be a better fit here.

We will either include the additional reference or a set of references here to substantiate this statement.

5) L. 16: “analyser properties” is a bit unspecific, be more precise.

We will add a statement to this sentence specifying spectroscopic baseline effects.

6) L. 17: “the variability of the calibration system”, what does that mean?

This expression is meant to encompass the variability of the water vapour mixing ratio and isotope composition of the calibration systems under normal and perturbed operating conditions (e.g. due to bubble bursting in a SDM). We will modify the writing to clarify this aspect here.

7) L. 15-19: Total uncertainty resulting from a variety of instrument characteristics at different water vapour mixing ratios and the characteristics of inlet systems were tested extensively in the two early publications Sturm and Knohl, 2010; and Aemisegger et al. 2012. Application in the field with aircraft-based measurements (Sodemann et al. 2017) and for near-surface humidity gradients (Seidl et al. 2023) were carried out in the more recent ones. This is a bit misleadingly written and should become clearer from this section of the text.

The intention here was to have the two more recent references refer to measurements under very dry ambient conditions (aircraft and polar environment). We will rephrase this section for clarity.

8) L. 29ff: I am not convinced that the microdrop system that is proposed in this paper alleviates all the mentioned problems: given the small liquid reservoir, the isotope composition of the standard liquid in the glass vials should be monitored as well for a reliable normalisation to the VSMOW-SLAP scale. What would be more convincing to me at this stage is to mention primarily that in the bubbler system the reference isotope composition of the vapour phase has to be predicted from equilibrium fractionation, which requires precise temperature regulation of the liquid phase in the bubbler. This problem is overcome in other devices by complete evaporation. Monitoring the liquid isotope composition is a must also in systems with complete evaporation including an SDM standard bag. When using large water reservoirs of several litres the close-up

monitoring of the liquid is not so essential in bubblers because the changes are much smaller than the precision of the reference measurement with IRMS or laser spectrometry.

We agree that the present system does not resolve all problems, and it was not our intention to claim this at this point, the intention was rather to bring forward aspects that contribute to variability in the performance of existing calibration systems. We will rephrase to avoid such a potential misunderstanding. We agree on the point of temperature control of the bubbler system, and will emphasize this point. However, we also like to mention that a large reservoir for a bubbler can impose substantial difficulty during field operation due to weight and form factor.

9) L. 50-51: "limited possibility to regulate the mixing ratio of the water vapour in the airflow", which is not really needed for the mentioned application.

This is true for the used analyzer system, but can very quickly be a problem if the gas flow rate through the cavity is different, for example in flux measurements. We will consider adding a clarifying sentence.

10) L. 64-65: The mention of flow rates in between the water vapour mixing ratio range and the different isotope composition is disturbing the logical flow of information in the reader's mind: Maybe something like the following would help: "... that provides the combination of a precise stream of water vapour across a range of water vapour mixing ratios and isotope compositions as well as operation at various flow rates between 50 and 250 sccm." -> group the range of mixing ratios and isotope compositions together, flow rates is a different aspect also from an operational perspective.

Thank you for this suggestion, we will revise accordingly.

11) L. 71: "and as a component in specific laboratory applications" this is a bit unspecific. Please be more concrete.

The specific applications are stated in the following sentence. We will rephrase for clarity.

12) L. 73: Here it would be very nice to be more precise about the gas stream of a preset background humidity and isotope composition for fluid inclusion analysis. A reader who is unfamiliar with this technique is lost here (see also my minor comment 53).

This sentence mainly serves as an outlook of what is to come. We will add a forward reference here to the respective section (Sec. 6 and 7) to clarify this intention.

13) L. 75-89: This paragraph is very dense with information and a bit difficult to follow. The first sentence starts with introducing the advantage of the microdrop generator in covering "a wider range of applications with CRDS analysers" than with currently available devices. But there are a countable number of applications, and the currently available devices are optimised for the tasks at hand. I rather think the advantage of the microdrop generator is the flexibility of it to produce a wide range of (predetermined) water vapour mixing ratios and isotope compositions. The application to microfluid inclusions in stalagmites is nice but this high flexibility is actually not needed there.

Our intention was indeed to point out the flexibility of the device, we will revise this section accordingly.

14) L. 77: Why are the authors so strict about the water vapour mixing ratio uncertainty? Is this useful in one of their applications? I thought they target the isotope composition of different vapour/liquid/fluid inclusion samples and not the absolute water concentration.

The background humidity for fluid inclusion analysis has in previous applications been provided by peristaltic pumps, which induced substantial noise on the analysis (Affoltern et al., xxxx). We will point this aspect out more clearly in the introduction as a motivation for the precision of the mixing ratio.

15) L. 81: I believe the first publications with fast-response analysers with flow rates above 100 sccm are Thurnherr et al. 2020 ACP with a detailed assessment of the impact of the flow rate on the water vapour mixing ratio dependence on isotope measurements in their supplement and Bailey et al. 2023 ESSD. From these publications with flow rates of about 300 sccm through the cavity it becomes clear that flow rate limitations of calibration systems is an important challenge.

Thank you for pointing out these references, which we will include in the revision.

16) L. 82: This sentence should be removed or moved to the discussion in the conclusions (see also my minor comment 74), the response times of the inlet is not tested in this publication and cannot be tested with the proposed microdrop dispenser because the flow rates are much too small (would need several liters per min to be of interest for this sort of application).

Flow rates of above 150 sccm are indeed possible with the presented system as presented in Fig. 7. While we show tests of long-term stability with 250 sccm, the flow regulator currently allows up to 500 sccm of flow rate. It is correct that downstream dilution will be necessary to provide higher flow rates. We note however that some inlets are operated in this range of flow rates (Chazette et al., 2021). We will revise this section and the discussion to provide the intended information at the correct location.

17) L. 88: The requirement of being field compatible seems ambitious and I don't fully understand it. How well do the dispenser heads work in very cold environments with vibrations and at low pressure? In principle drift correction in the field could be done with simpler existing systems, the advantage of this system resides in the possibility to characterise the water vapour mixing ratio – isotope ratio dependency in detail. This is more a laboratory than a field application.

This requirement comes from the desire to have one multi-purpose system, which could lower cost compared to requiring several dedicated components. It may also for example be desirable to verify the baseline characteristics a system before shipping it back from a field deployment. There are of course a wide range of possible field applications, and there are certain limits that the system presented here can not operate under without additional modification. For example, operation at freezing level would require active heating control within a heat insulation, or the use of temperature-controlled dispenser heads (which are available from the same manufacturer). Low pressure is no limitation to the dispenser head, as they are equipped with a differential pressure regulation with respect to ambient conditions. We have not done vibration testing, but note that there are no actual mechanical parts in the dispenser head that could for example break or get stuck during shock. Our system is not necessarily more complicated than for example an SDM, and the absence of mechanical parts allows for in principle more extensive operating duration. We also have since submission learned that gas bubbles an important aspect that so far has stopped dispensing after several hours. We can now control this aspect much better by de-gassing our standard liquid before operation, and can achieve continuous operation over several days, rather than hours. We will revise the text here and in the discussion to better explain, motivate and detail the field compatibility requirement.

18) L. 92: SDMs can go well below 6000 ppmv.

We acknowledge this is possible, but with limitations. Here is an excerpt from the user manual of the Picarro SMD A0101 User's Guide from April 2010, pg. 14:

"The concentration of vapor will be determined by the user programmed liquid flow rate. A rate of 0.02 microliters/second corresponds to approximately 6000ppmv. The vapor concentration is a linear function of the liquid flow rate. Rates higher than 0.08 microliters/second (24000ppmv) are prevented by the software in order to prevent accidental saturation of the analyzer. The precision of the isotopic ratio measurement is specified for a vapor concentration of 6000 to 20000ppmv. The precision will suffer significantly below 6000ppmv, increasing the measurement duration will compensate to some degree. The dry air source, such as Drierite® condition air with a 200- 300ppmv water concentration, can contribute significantly to the measured isotope rate when operating at standard vapor concentrations below 6000ppmv .".

We will clarify in the revision that 6000 ppmv is the lower recommended limit of operation of the SDM.

19) L. 94: What does “Injections with the autosampler are feasible with a specific method” mean?

This sentence was mistyped, we will clarify that it is possible to obtain a range of isotope ratios from liquid injections with some manual work.

20) L. 95: This joins my major comment M1: unless you are able to very precisely predict the isotope composition of your mixture with the two dispenser heads you cannot really overcome this limitation from other systems.

See our reply to comment M1.

21) L. 101: this doesn't become clear from the schematic in Fig. 1.

Thanks for pointing this out. As also requested by Reviewer 1, we will add a photograph to Fig 1 or the appendix that clarifies the design of the evaporation chamber.

22)L. 103: this is a very small amount of water which can be rapidly impacted by evaporative enrichment in the field!

The 12 ml glass vial is a default size, but it is possible to operate the DHs from larger reservoirs, we will add a corresponding statement here or in the discussion.

23) L. 106: “retention time of water vapour” are interactions with the chamber walls meant here? If yes, to me “memory effect” would be clearer.

Yes, we will include the term memory effect, as also suggested by Reviewer 1.

24)L. 140: how robust are the voltage and frequency settings and their associated efficiency to prolonged usage?

According to manufacturer information, these settings are constant for each DH once defined, but there are multiple possible combinations that result in different drop sizes. The piezo crystal may degrade over time if very high voltages are involved, which is not the case for our operation. We will add this information with a reference to personal communication with an engineer from Microdrop GmbH.

25) L. 148: “We can used this relation between drop size and mixing ratio to compute the effective drop size...” I am a bit lost here. Do you mean the relation between the frequency and the mixing ratio can be used to estimate the effective drop size?

Yes, this was a typo, we will correct the sentence accordingly.

26) L. 154 and Fig. 3: is it only my print out that make the curves in Fig. 3 not look linear?

The lines in Fig. 3 are strictly linear.

27) Fig. 3b: Why is the H₂O mixing ratio coverage worse with the settings for panel b than in panel a?

We have simply run fewer frequencies for this setting, as the linearity was much better.

28) L. 161: This seems to be key, indeed. Can the uncertainty from this fitting procedure be quantified in ppmv for a series of runs? And how frequently does this readjustment of dispensing parameters need to be done?

We have since submission come to understand that this assessment does not need to be done very frequently, maybe on a yearly basis, to ensure the DHs have not been ageing, as is expected.

It is possible to automate this characterisation from the control software. In the revision, we will add information for the uncertainty for both run series.

29) Fig. 4: I am not sure that I really understand the meaning of this figure. What does all dispersion parameters mean? Could this Figure be placed a bit more logically in the discussion. A part from a useful diagnostic, I don't see the use of it? Is it possible that only showing the blue and red distribution would help the reader because these are the selected settings. In general, I am not sure that the reader is interested in the full distribution with all your tested dispersion parameter setting.

It appears that Reviewer 1 appreciated this information, as precise humidity control is a great concern in fluid inclusion analysis. We will therefore clarify in the Figure caption that all dispersion parameters corresponds to the data displayed in Fig. 3.

30)L. 192: here and elsewhere: it is "Allan" variance not "Allen" and the adequate reference would be Allan, 1966 Allan, D. W.: Statistics of atomic frequency standards, Proc. IEEE, 54, 221–230, doi:10.1109/PROC.1966.4634, 1966.

Thank you for this correction and reference which we will include in our revision.

31) Eq. 1: I think the authors mean $y_{i+1}(\text{?})$ not $y_{i+1}(\text{?})$.

Yes, thank you for this correction.

32) L. 206: The SDM is optimised for the precision of the $\text{?} \text{?} \text{?} \text{?}$ and $\text{?} \text{?} \text{H}$ not for the mixing ratio. For a fair comparison with a high quality mixing ratio signal, a dew point generator should be used.

While it is true that the SDM is not optimized for mixing ratio stability, this is an important aspect for the fluid inclusion application and thus part of the assessment here. We will however consider including a comparison of mixing ratio stability only from a dew point generator for the revision.

33)L. 225: this is understandable only if the basic concept of fluid inclusion peak measurements on a stable background has been explained in the intro. Maybe a reference to Section 7 could also be added here.

A reference to Sec. 7 and Fig. 13 are already included, but we will in the revision clarify better upfront how the fluid inclusion measurements work.

34) L. 254: "The linearity of this mixing is provided over a range of mixing ratios". I can't really evaluate this statement based on Fig. 8a. How about coloring the data points with the mixing ratio?

We will implement this suggestion in the revised version.

35) L. 255: "At lower humidity than 5000 ppmv deviations from the linear mixing become apparent" Also here, I can't really see this in Fig. 8b, which shows the RMSE of $\text{?} \text{?} \text{?} \text{?}$ and $\text{?} \text{?} \text{H}$ from the observed mixing line at different water vapour mixing ratios. The RMSE is by the way very large implying that the predicted reference isotope composition of the gas stream has a large uncertainty making it very difficult to me to effectively these mixed samples as reference calibration samples.

It appears that Fig. 8 was not as clear as we hoped. We will make an attempt to better support the statements in the text with this Figure. We will double-check the scale of the RMSE which indeed indicates very large values.

36) L. 260: what is Mix000 and Mix 100? This is the first time these labels appear.

Mix000 is purely from DH1, and Mix100 purely from DH2 (the number is the percent fraction from DH2). In the revision, we will define these abbreviations as they are introduced.

37) L. 260: Where can I see this bending down (please add a reference to the figure that is meant here).

We will add a reference here to Fig. 9.

38) L. 263: the phase space you are showing in Sec. 6 is the correction function in the $\ln(\text{mixing ratio})$ vs. $\delta^{2}\text{H}$ (or $\delta^{18}\text{O}$) phase space.

We are unfortunately not sure how to interpret this comment, the logarithmic transform in Sec. 6 is used to obtain a more linear display of the dependency characteristics.

39) L. 265: the microdrop vapour generator was already connected to the analyser for all the evaluations presented in the previous section. Here you switch from characterising the microdrop vapour generator to using it to characterise CRDS analysers in their water vapour mixing ratio and isotope composition dependency.

The intention here was to contrast two applications, one where the microdrop vapour generator is directly connected to the CRDS, and one where there are additional appliances in between. Apparently this statement can be misunderstood and we will consider rephrasing in the revision.

40) L. 267: The water vapour mixing ratio and isotope composition dependency is of spectroscopic origin and due to uncertainties associated with the baseline. Even if a system is optimised for measurements at lower humidity levels than 20'000 ppmv this problem occurs.

This is true. What we wanted to point out here is that Picarro analyzers are in fact already corrected for baseline effects in the factory with a target humidity of about 20'000 ppmv, and the remaining dependency is thus a residual of this factory baseline correction. We will rephrase for clarity.

41) L. 273: This is misleading. The spectroscopic origin of the concentration dependency has been mentioned already in early laser spectrometric instrument evaluation work such as from Sturm and Knohl, 2010 and Iannone et al. 2009.

We think this might be a misunderstanding, it was not our intention to mislead the reader. What we mean to say is that the additional dependency of the previously identified mixing-ratio dependency (e.g., Sturm and Knohl, 2010, Iannone et al., 2009) was shown to be in addition dependent on isotope ratio by Weng et al., 2020. We will rephrase to avoid potential misunderstanding.

42) L. 274: matrix gas -> carrier gas

ok

43) L. 296-308: This is very difficult to follow, could the respective sequences be clearly indicated in Fig. 9? This paragraph should be in stronger dialogue with what we see in Fig. 9 for the reader to be able to follow.

We will add annotations and rewrite to connect Fig. 9 and this paragraph better.

44) L. 313: Here I got lost, where do I have to look above or below?

We will rephrase and add a reference to the relevant section.

45) L. 315: Can the authors be more precise about the number of outliers they filtered out? This is an important quality measure for the microdrop generator. Because an ideal calibration system needs as little instrument measurement time as possible so minimising the occurrence of "outliers" is a key aim when designing a robust calibration system.

We will include the number of outliers here. However, we note that after the potential of leaks was detected, care was taken to avoid such artifacts and no more outlier removal was required thereafter.

46) L. 320: What is the share of new data points from the microdrop generator vs. the data points from Weng et al. (2020) in this analysis shown in Fig. 10? Could the data points from the microdrop generator be highlighted in a different color or shape?

We will highlight the additional data points in the revised Figure 10 by a different color.

47) L. 331: the delta value is specified and used in equations 2 and 3.

Since the humidity is typically not exactly at 20'000 ppmv, but bracketed by values nearby, we interpolate between neighbouring data points. Interpolation with a polynomial makes thereby use of most of the data points.

48) L. 336: this final step was a bit obscure to me. I thought that the correction function is 0 per definition at the reference mixing ratio level?

The 2D polynomial fit is not constrained to zero at the reference level and thus result in an offset, which we correct for by this additional final step.

49) Table 3: the RMSE is most likely very dependent on the mixing ratio? Maybe a Figure showing this dependence as a function of mixing ratio would be more informative.

We will investigate the dependency with respect to mixing ratio and consider to include a figure or additional lines in Table 3 for sub-sets of the mixing ratio.

50) L. 350-355: this paragraph was again not clear to me. Which subsample of available measurements?

This was a Monte-Carlo approach, where we draw a non-unique sub-sample from the actual distribution of data points. We will give more detail and attempt to rephrase for clarity.

51) Fig. 11: The vertical axis is not the raw but the reference $\delta^{2}\text{H}$ and $\delta^{18}\text{O}$, right? There are no dashed grey lines (at least not in my print out) and what are the blue lines? Would this figure be better readable if it was shown in this format in the appendix (to illustrate the limited concentration effects at high water vapour mixing ratios) and with a zoom in to 0-5000 ppmv (which is the interesting part) in the paper? Right now one does not see much about the important effects at low water vapour mixing ratios.

Vertical axis is indeed the raw delta values, we will correct this. Blue lines show contour levels for the correction found here, and solid black lines are the correction function of Weng et al., 2020, this will be corrected in the caption.

52) L. 357: "a constant vapour stream" with known water vapour mixing ratio and isotope composition (very important!)

Yes, we will add this important information.

53) L. 357-366: This information should already be provided in the introduction.

We understand from the comments of Reviewer 2 that this information is necessary in the introduction and will revise the introduction and Section 7 accordingly.

54) L. 369: "memory effects can be removed entirely" is misleading and a bit optimistic. Isn't the advantage of the CRDS technique mainly the ability to measure $\delta^{2}\text{H}$ and $\delta^{17}\text{O}$ (possibly even $\delta^{18}\text{O}$) quasi-simultaneously with much reduced sample preparation effort?

We will moderate and rephrase this statement.

55) L. 372: “higher throughput and better reproducibility” than what?

During fluid inclusion analysis, several samples are typically analysed from the same growth layer of a stalagmite. We will add a statement that this is a comparison to methods that involve collection of the sample by a cold trap for subsequent analysis on a mass spectrometer.

56) L. 384: Fig. A2 -> Fig. 12?

Correct.

57) L. 387-393: connect this paragraph better to Fig. 13, they should be in better text- illustration dialogue to help the reader follow.

We will rephrase this section to integrate better with Fig. 13.

58) L. 395: As mentioned in M2 I wonder about the necessity of this step given the high mixing ratio of the background air stream.

See our reply to comment M2 above.

59) L. 397: “Mixing ratio -isotope ratio corrections...” this has already been mentioned in point i) just above and appears as an unnecessary repetition here.

We will revise for brevity.

60) L. 399: what is meant by “size- \square ” relationship?

This refers to a relation between the amount of liquid injected and the delta value during calibration. We will rephrase this expression.

61) L. 405-414: I got lost in this paragraph. As the authors write at L. 410 this appears redundant. Fig. A1 is not very convincing in showing a consistent dependency of the isotope signals on the injected amount.

This section relies on the analytical methods published earlier by Affolter et al., (2014). While the step may appear redundant, it is important here to maintain the principle of identical treatment of sample and standards. We will revise for clarity.

62) Fig. A1: To which mixing ratio levels do these injection volumes correspond?

We will add this information to the figure caption.

63) L. 407: “drift in values”, what do you mean by values?

This refers to a drift in the background, we will rephrase for clarity.

64) L. 412: the last sentence of this paragraph “Finally,...” is particularly obscure to me. Could the authors illustrate this with a figure?

We will either refer here to already published method description or include a supplementary figure to clarify this part of the procedure.

65) L. 431: Here I am a bit puzzled: the authors write that water vapour mixing ratio – isotope dependency corrections are necessary below 5000-10'000 ppmv (L. 270). But the background gas stream can be designed in such a way that the mixing ratio is larger than this and ideally even close to the optimal water vapour mixing ratio operation level of 20'000 ppmv? So why does this correction matter? Can the authors show how different the isotope signal estimates get when applying vs. when not applying the water vapour mixing ratio correction for their aliquot or the samples used to mimic real fluid inclusions? This is also my major point M2.

See our reply to comment M2 above.

66) L. 432: “more correct” than what?

This sentence will be rephrased.

67) L. 434: Why is the precision of the water vapour mixing ratio mentioned here? Isn't what matters for precise fluid inclusion measurements the precision of the isotope signals?

As explained in our reply to M2, the stability of the mixing ratio is important for this application to separate the background water vapour clearly from the water vapour released from the crushed sample.

68)L. 455: for which application would horizontally mounted DHs be an interesting option?

Due to the design of the evaporation chamber, the DHs are mounted horizontally here. We will rephrase this sentence for clarity.

69) L. 462: Flow rates between 300 and 600 sccm through the cavity are now used in different applications (aircraft-based and flux measurements) and present many advantages (e.g. faster response times, better signal to noise ratio at shorter averaging times) but this range of flow rates cannot be covered the microdrop dispenser system. Why is that so, and is an extension to these higher flow rates possible with the given operational range of dispenser head triggering frequency?

The current limitation to 500 sccm for the device presented here originates mainly from the specification of the gas flow regulator. It may be possible to obtain higher flow rates with the present device, but we did not have the necessary pumps available to produce such high cavity flow rates. Higher flow will require higher dispensing frequencies, and/or dispenser heads that can produce larger drops. We will add some clarification of this aspects to the discussion.

70)L. 475: remind the reader that the Picarro SDM is the commercially available benchmark used for characterisation of the microdrop dispenser.

Will be added in the revision.

71) L. 475: In the long-term precision (relevant for calibration) the microdrop dispenser is not better than the SDM for the isotope signals (which we are interested in). The dispenser is better than the SDM for water vapour mixing ratio, for which however a dew point generator is needed anyway for precise calibration. Together with the many small problems that occur with air bubbles in the dispenser head, the small amount of liquid that needs to be exchanged regularly, this questions the utility of this system for regular field calibrations. It is very useful for an in-depth characterisation of the water vapour mixing ratio – isotope signal dependency in the lab but apparently not so much for prolonged measurements in the field.

We can partly understand this assessment of the reviewer based on the presented information. However, since submission of the manuscript, we can report on a much longer uninterrupted operation time of the device thanks to de-gassing of the calibration liquid before use. We will update the relevant parts of the manuscript accordingly.

72) L. 477: I don't understand what this sentence implies scientifically. Which specifications are meant here? The dispenser head and mass flow controller specifications or the objectives set by the authors for the system in Section 2? Maybe a reference to section 2 could be added here.

Yes, we will add a reference to Section 2.

73) L. 495: Could this simply be a numbered list of implications? It would be easier to read.

We will consider reorganising as suggested.

74) L. 502: Here is the place where the sentence from L. 82 could be brought in: larger flow rates are needed to test the inlet system and the overall response time of different water vapour isotope measurement field setups.

Thank you for this helpful suggestion, this paragraph will be revised accordingly.

75) L. 502: Again this is misleading, response times of laser spectrometric water vapour isotope measurements were not the topic of this publication. If one just reads the conclusions, this sounds like a result from this study. References to the studies Sturm and Knohl, 2010 AMT (tested Synflex tubings in the lab) and Tremoy et al. 2014 JGR (tested Synflex tubings in the field) should be made.

The reference to Tremoy et al., 2014 went missing during the editing, it was certainly not our intention to mislead the reader, see also reply to Reviewer 1. We will add the mentioned references in the revision.

Technical points:

- 1) L. 17: . Currently
- 2) L. 67: technology, which allows
- 3) L. 75: “water” vapour
- 4) L. 76-80: Grammatically and for keeping the reader’s attention having (ii) in the same sentence as (i) would be much more convenient.
- 5) L. 103-104: “one 12 ml glass vial that holds a liquid water standard and which is mounted next to ...”. It is not the evaporation chamber that holds the liquid water standard, right?
- 6) L. 161: demonstrates
- 7) L. 161: “... a suitable dispensing parameters”
- 8) L. 214: microdrop
- 9) Caption Fig. 4: dispensing -> dispersion
- 10) L. 348: og -> of
- 11) L. 369: CDRS -> CRDS
- 12) L. 464: viscosity
- 13) L. 509: performed
- 14) P. 27, Fig. A1: is “assigned” really the term you want to use here on the y-axis? To me this seems to be the “reference”.

We will implement all technical changes as suggested.