We thank both reviewers for carefully reading our manuscript and for the detailed feedback aimed at helping us to further improve the manuscript.

Anonymous Referee 2

The discussion as a result of the two reviews reconfirmed my impression that there is very little to comment on the technical work of this manuscript. This part is very detail oriented and shows that the authors built and properly validated a very high quality hygrometer.

We really appreciate the comments in the first review which made our paper clearer and easier to understand for the community. Currently, the remaining questions seem to focus on the two areas a) is SEALDH-II a stratospheric instrument b) how much technical details should be in an instrument validation/comparison paper.
We hope that we can clarify these remaining issues with our response, so that the second reviewer agrees with the "publish as is" recommendation of the first reviewer.

Below we address the raised concerns in a point by point fashion. Changes are highlighted in the attached revised version.

However, my concerns regarding the novelty and importance have been reconfirmed. I particular, the connection with the earlier Hygrometer for Airborne Investigations (HAI) has not been adequately described. Furthermore, there apparently was an earlier version, SEALDH-I, which had the same features as SEALDH-II. In their replies the authors stated that "Maybe we didn't elaborate clearly enough on the connection between the instruments". That is somewhat of an understatement, since neither of the other two instruments were even mentioned. This was quite surprising, since both instruments come from the authors themselves. It is rare to see that authors ignore their own work. The authors also confirm my suspicion that these instruments are borne out of the same concept, I quote, "to ensure a permanent, highly defined, multi parameter instrument control for each individual data point". Therefore, the authors' claim that SEALDH-II is a novel concept is simply not correct and should not be described as such. This concept has already been implemented in HAI as atmospheric instrument and SEALDH-I as proof of concept. I fully appreciate the differences between HAI and SEALDH-II and the differing motivations for both instruments. HAI is more appropriate for atmospheric observations, while SEALDH-II was built with metrological traceability in mind. Nevertheless, these instruments are based on the same concept, measure the same atmospheric parameter using the same measurement principle, claiming to be calibration-free. Therefore, statement such as, I quote, "implementing a new holistic concept to achieve higher accuracy levels", made in the abstract, are simply not correct. The authors may claim novelty for HAI or possibly SEALDH-I, not for SEALDH-II. Monitoring many more instrument parameters than in HAI may improve the confidence in the observations and may allow a better characterization of the uncertainty budget. Their instrument is certainly of very high quality, but the authors cannot claim a 'new holistic' concept, since that has already been done. Instead the authors should include an extensive discussion of the relation between these instruments in the manuscript, not just the replies to the authors.
A key part of this manuscript is the metrological validation of SEALDH-II. Given that HAI is much

more suited to atmospheric measurements, the authors should elaborate, why they chose to build an entirely new instrument, instead of validating the first instrument they built for atmospheric measurements. Some of the discussion in their replies should be included into the manuscript as well as their plans for the validation of HAI.

Thank you very much for that comment. There seem to be a sticky idea, which the reviewer enjoys discussing. However, the questions are far away from the focus of this paper:

> The title of the publication is:
> "SEALDH-II – a calibration-free transfer standard for airborne water vapor measurements: **Pressure dependent absolute validation from 5 – 1200 ppmv at a metrological humidity generator**"
>
> The pure focus on this paper (which is also stressed out in the publication) is metrological validation of an airborne hygrometer: More specifically about the **first metrological pressure dependent, absolute validation in the range 5 – 1200 ppmV**.
> The reviewer is much too focused on the hardware novelties. Even though we indeed wanted to mention our new holistic data concept, we did not describe it in detail or put technical details in the focus of the paper (as it has already been described elsewhere). The same also holds for the hardware of SEALDH-II (also has been described previously in more detail), which is also not in the focus of this paper. However, some coarse description of the hardware and data management is needed in order to link the contribution of these developments to the performance of the instrument. Thus, the **novelty of this manuscript comes from the metrological validation of our sensor.** Such a validation has not been done before - neither with this metrological rigor nor in this concentration range. The reviewer also didn't name us papers where something quite similar was described. Hence, the purpose of this publication is by no means the announcement of a "new" instrument or some "new" hardware, but the description and discussion of the validation experiment. This is of high relevance if instruments like SEALDH-II are to be used as mobile metrological transfer standards for atmospheric humidity measurements in the future. The need for such transfer standards can be clearly motivated from the results of the AquaVIT comparison.
>
> The problem with the reviewer comments are therefore that they request to convert the paper more towards more hardware description and going very deeply into technical details between the different instrument generations. We are certainly very happy to discuss such details with the reviewer - might it be in an entirely technical discussion paper or in an offline conversation. But, for the presented manuscript, we have to object to the reviewers request as these details have been mentioned previously and so that there is no need for duplicating them in this manuscript. So this paper is particularly **NOT focusing** on the following topics:
>
> a) A detailed instrument description of SEALDH-II. We have published a technical instrument description already. We believe that leaving out all these fine technical details on the hardware and the holistic data management significantly improves the clarity of the manuscript. When we pointed out some of SEALDH-II features, then we felt that these are vital for the understanding of the presented data. Further, our presented measurements are meant as a validation (for SEALDH-II to be a transfer standard) rather than an instrument calibration.
> We addressed as many of reviewer questions in the first rebuttal as possible, because we understand the reviewers curiosity, but most of the questions where not related to the validation core

of this paper at all. Thus, at some level we had to decide to stay focused in the interest of the readers and not include all technical details in this manuscript. In particular as they have already been mentioned elsewhere. Therefore, we wrote "Maybe we didn't elaborate clearly enough on the connection between the instruments" to answer the reviewer's question.

b) Since we focus on a "metrological, pressure dependent, absolute validation" of SEALDH-II, any detailed discussions of HAI topics are completely out of the context. HAI is, as we described extensively in the last rebuttal, a multi-phase hygrometer. The challenges for the HAI instrument measuring at two different wavelengths, and with an open-path cell are entirely different. Yes, the HAI and SEALDH developments have overlapping areas, but this is not a redundancy but absolutely beneficial, as the "metrological, pressure dependent, absolute validation" exercise done with SEALDH-II can be transferred quite well to the extractive channels of HAI! Maybe the reviewer is not fully aware of the very significant complications of an individual metrological validation of the <u>open-path</u> channels of HAI, otherwise he would acknowledge that such an effort would be worth a full paper by its own. Luckily, the HAI concept allows – by design - during cloud free conditions an inflight cross-validation of the open path channels by referencing them to the extractive channels.

c) In the focus of a paper is a "metrological, pressure dependent, absolute validation"; we do not agree that one has to explain all other developments which one has done over the last years. HAI is for that publication not relevant and therefore not mentioned. (Usually, reviewers criticize authors about "over-citing" themselves). The comment that both of the instruments measure with the same technique (I guess that is related to TDLAS spectroscopy) is true but not in any way a down site, as we have explained in the previous point. There are, beside the many referred academia devices, also commercial sensors such as the ones from Picarro, SpectralSensors, Siemens, ABB etc. which use this basic technology.

d) The reviewer claims that there was a "SEALDH-I"; this is actually true, but we never published an instrumental paper about this development step between SEALDH-0 (a non-flight qualified laboratory test setup) and a SEALDH-II (a fully featured airborne transfer standard). The only "publication" ever explaining SEALDH-I was short German handout [1] at a German meeting the "GMA/ITG-Fachtagung Sensoren und Messsysteme 2012" for industrial partners, which via the language barrier certainly doesn't penetrate the international community at all. Due to the concise structure of this handout, we highly doubt that anyone would gain any significant knowledge out of it, so we don't see the need to reference such almost grey literature (and again if we would, most reviewers would complain about referencing of non-english grey literature); the argument stays the same for the few press release about the SEALDH developments and flights.
Since we do not have any SEALDH-I instrumental paper, we cannot refer to a publication.

To summarize:
The reviewer commented: ". Instead the authors should include an extensive discussion of the relation between these instruments in the manuscript, not just the replies to the authors."
This is NOT the focus of that paper at all. The title and the abstract clearly state:
"SEALDH-II – a calibration-free transfer standard for airborne water vapor measurements:
<u>Pressure dependent absolute validation</u> <u>from 5 – 1200 ppmv</u> at a metrological humidity generator"

> As a compromise to this reviewer, we can offer to modify the paper title to read as follows: "Pressure dependent, absolute validation of SEALDH-II – a calibration-free transfer standard for airborne water vapor measurements - from 5 – 1200 ppmv using a metrological humidity generator " or even ""Pressure dependent, absolute validation of a calibration-free, airborne laser hygrometer transfer standard (SEALDH-II) from 5 – 1200 ppmv using a metrological humidity generator"

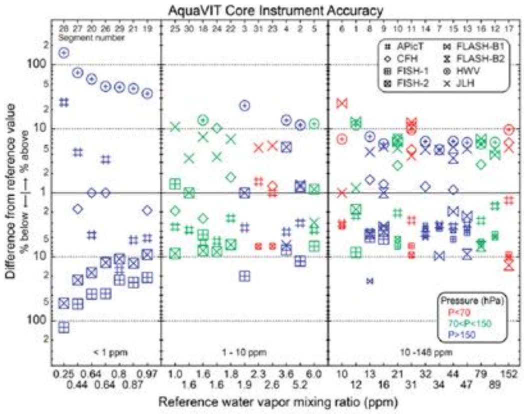
The second major issue I have with the comments is their relation to stratospheric measurements. The title includes "airborne water vapor measurements" and "5 – 1200 ppmv". Throughout the paper, the authors refer to a measurement range of 65 hPa – 950 hPa and that "the instrument is built for a concentration range from 3 - 40000 ppmv". The lower end of this pressure and mixing ratio range clearly includes stratospheric conditions. The authors motivate their work through significant differences between measurements, most of which are UTLS and stratospheric measurements. They discuss the AquiVIT campaigns, which were conducted largely for UTLS instruments to understand the difficulties of measuring stratospheric water vapor.

This question is pretty similar to the question which the reviewer asked the last time.
I will try to answer these questions first in the briefest possible way; this repeats the paper and the rebuttal:
a) "the instrument is built for a concentration range from 3 - 40000 ppmv"
=> Yes: The lower range limit defined by offset uncertainty, the higher by condensation risks when dew point equals instrument temperature.
See line 164 – 169 in the most recent version of the script (first revised version)

b) "The lower end of this pressure and mixing ratio range clearly includes stratospheric conditions" Yes, with the uncertainty stated in the paper (e.g. line 164 and 171); so at 3 ppmv the instrument has an uncertainty of 100% (!). Hence, we would not argue that SEALDH-II is a specialized UTLS instrument. If it had been the purpose, SEALDH-II's optical path length would have been certainly significantly longer.

c) "They discuss the AquiVIT campaigns, which were conducted largely for UTLS instruments to understand the difficulties of measuring stratospheric water vapor."
As written in the rebuttal extensively (and in the paper see from line 80), we referred to AquaVIT for two reasons: a) it is the one and only **representative airborne hygrometer comparison** ever done; if the reviewer knows about another one which is as representative as AquaVIT, **we would love to learn about it and reference it adequately**. b) AquaVIT covered a concentration range up to 150 ppmv. Especially the higher levels of the AQUAVIT overlap with the useful (=non offset-uncertainty driven) concentration range of SEALDH-II and allow an effective relation to SEALDH-II's validation results. Nevertheless, this

does not mean that SEALDH-II is a specialized UTLS instrument.



For a more detailed discussion, we refer the reviewer back to our first rebuttal where we answered in greater detail the same question.

One of the drivers for the AquaVIT campaigns was the disagreement between some aircraft and balloon borne observations. Water vapor observations of less than 5 ppmv were the most important range of this disagreement. Given the uncertainty of SEALDHII, this instrument would not contribute to this concentration range. Despite the un-questioned quality of the observations presented here, this raises the question of the importance of their results, especially given the frequent reference to AquaVIT. The authors should clearly point out, whether they have achieved a metrological validation of a field deployed instrument that can measure stratospheric water vapor (100 hPa, 5 ppmv) with an uncertainty of less than 5-10%.
==> Thank you for that comment; nevertheless we are also a bit puzzled by it.
SEALDH is NOT an instrument designed especially for UT/LS application, nor is it one of the common "standard" even commercially available laser hygrometers like the ones from WVSS, Picarro, LosGatos, etc. Instead, SEALDH is designed as a metrological transfer standard which is still suitable for field applications and ensures full traceability to the primary water scale. Thus, the application range is wider than just UTLS and the requirements are also much more rigid and demanding. It is clearly stated in the abstract that we claim a "primary validation" that covers the relevant mid to upper tropospheric and lower stratospheric (MT/UT/LS) concentration and pressure range. This was done via "15 different H2O concentration levels between 5 and 1200 ppmv". At each concentration level, we studied the pressure dependence at 6 different gas pressures between 65 and 950 hPa." The introduction states (as well as the referred paper in section 2 [4]) that the instrument is built for a concentration range from 3 - 40000 ppmv; 0 -1000 hPa with a calculated uncertainty of 4.3% ± 3 ppmv. The lower detection limit is defined at that point where measurement value equals uncertainty (as explained in the paper: "The calculated mixture fraction offset uncertainty of ±3 ppmv defines the lower detection limit."). The measurements in figure 8 show how conservative these estimations for the calculated uncertainties were since the "real" deviations are only in the 0 - 20% range (with the lowest level at 35%). SEALDHs science and metrology targets are thus much wider and of different perspective than those of the AQUAVIT 1 or 2.
The reasons why we explained the AquaVIT study in such a detail are the following: Firstly, it is the most extensive comparison exercise which was carried out on such a level (representative for the community, externally reviewed, blind submission, clear statements, exemplary work, etc.) that allows a reliable relative performance statement about the state of the art of airborne hygrometry. Secondly, as you can see in the graph above, AquaVIT covered the range up to 150 ppmv. Most instruments deployed during AquaVIT had a smaller concentration range compared to SEALDH-II because they were specifically developed for stratospheric measurements. With reference to the reviewer's question, we have to comment that one should not compare apples with oranges. SEALDH-II is intended as a transfer standard for the troposphere AND lower stratosphere and thus designed as a wide range hygrometer. This asks for pragmatic compromises which have consequences. If we had in mind to make a comparison only for the stratospheric range, we could easily replace the 1 m long extractive cell with a fiber-coupled multipath cell having 10x - 50x more path length. This would drastically improve SEALDH-II's sensitivity AND offset stability in the low concentration range. As the sensitivity and offset largely scales with the cell path length, this would lead for SEALDH-II to a calculated uncertainty (see [4]) of 4.3% ± 0.3ppmv (@ 10 m) and of 4.3% ± 0.06ppmv (@50 m) respectively, which seems quite well suited for a stratospheric application.
It also has to be kept in mind that we stated conservative calculated uncertainties; the "real" experi-mental deviations are usually smaller, as described and shown in this paper. In our experience – also as participant in AV 1 and 2 - we made the experience that metrological humidity traceability options in the atmospheric sciences are more or less ignored, despite the availability of a fully validated metrological reference scale and infrastructure. It seems highly likely for us that an improved traceability would lead to better absolute accuracy of airborne hygrometry. SEALDH was designed to enable this link between metrology and field

sciences. With the series of SEALDH papers and in particular with the one here under review, we intend to present the data that demonstrate this capability in a metrological sense and not just present another UTLS hygrometer.
So the argument of the reviewer that only improvements in stratospheric sensing would be required or relevant is a bit short sighted and ignores to a large extent our focus towards implementing general traceability for airborne hygrometry over the full range from LT to LS. Atmospheric science studies the entire atmospheric water cycle and not just its subsections in the stratosphere and as such, it would definitively be beneficiary to establish traceability with a wide range instrument such as SEALDH in order to improve the comparability between all atmospheric sub-compartments and not just between UTLS focused instruments.
We added a few lines to explain why we compare to AquaVIT.

The reviewer did not comment on this section in the first rebuttal or indicate why this discussion wasn't satisfactory. Maybe he overlooked it?

All of this gives the impression that their instrument covers UTLS and stratospheric measurements, which in fact they do not. The change in wording in the revised manuscript is insufficient and the authors should add a discussion about what their results mean for the low end of the mixing ratio and pressure range for that instrument. After all they still claim their instrument to be an instrument for airborne measurements.
We hope that we finally clarified that question with the text above.
Our current manuscript does -on purpose - NOT contain any specialized, dedicated UTLS / stratospheric claims; we described (e.g. 164) the range of SEALDH-II with (3 – 40000 ppmv). We truly hope that someone who knows the expression UTLS, also knows that 40000 ppmv or even the 1200 ppmv upper range covered in the present manuscript does NOT resemble a stratospheric water vapor mixing ratio. Thus again, we do NOT claim UTLS specialized performance or suitability for SEALDH-II

Maybe the reviewer got distracted by the sentence (line 55-59): "The ambient gas pressure (70 – 1000 hPa) and gas temperature (-80 – 40°C) ranges are large and both values change rapidly, the required H2O measurement range is set by the ambient atmosphere (3 – 40000 ppmv), mechanical stress and vibrations occur, and the sampled air contains additional substances from condensed water (ice, droplets), particles, or even aircraft fuel vapor (e.g. on ground)."
The typical humidity range of the atmosphere and the range of SEALDH-II is by accident the same (see above what drives SEALDH-II's limits).

Given the experience of the authors I fully believe that they could build an instrument optimized for UTLS work as they explained in their replies.
An instrument for UTLS would focus on the range between 0.1 – 50 ppmv (uncertainty 5% +/- 0.1 ppmv)). The setup for such an instrument would be pretty much different. Beside different laser wavelength/path length etc., the major challenges for such low water vapor mixing ratios are the sampling issues as well as the more rigorous parasitic water treatment. If the reviewer would like to cooperate in such a development, we would be more than happy to do so. SEALDH-II will never be able to achieve that low ppbv range, since it is as a wide range instrument 3 – 40000 ppmv (uncertainty 4.3% +/- 3ppmv), designed for a broad application range which certainly concentrates on the troposphere and not the stratosphere.

However, the manuscript is about the instrument they have, not about an instrument they could potentially build. Since the current instrument is not suitable for UTLS work, it would be much more appropriate if the authors motivated their work with the appropriate instruments, i.e. exclusively tropospheric instruments and better discuss, which part of the atmospheric water vapor range the current instrument can measure and which it cannot.

Yes, we cannot agree more! SEALDH-II's range is (3 – 40000 ppmv) it **not a UTLS** instrument. SEALDH-II can be used in all area where the measurements uncertainty is suitable (4.3% +/- 3ppmv). Therefore, if an uncertainty of e.g. 11% is acceptable, the instrument can be deployed at measurements of 30 ppmv.)

In the discussion it would be appropriate to then discuss that the features of this instrument could be implemented into a new instrument dedicated to upper tropospheric and stratospheric water vapor measurements.
The title of the publication is:
"SEALDH-II – a calibration-free transfer standard for airborne water vapor measurements:
Pressure dependent absolute validation from 5 – 1200 ppmv at a metrological humidity generator"
We don't believe the clarity of the paper increases if we extend it to a white paper of general instrument development discussions. That is not the focus of the paper at all and would risk that other reviewers than complain on a too broad and unspecific "focus".

The authors have now also included a reference to SEALDH-I and its validation with the German national standard. Therefore, the validation exercise of SEALDH-II is not the first effort of that type.
As mentioned above, there is no instrument publication of SEALDH-I. The comparison of SEALDH-0 (laboratory setup) with the primary standard [2], which **is** referred in the paper, was a) **NOT pressure dependent**, b) and at quite **high concentration levels** where the spectroscopic challenges are entirely different (keyword: saturation, high absorbance effects, linearity issues, condensation problems). The scope of this work is entirely different. There are clear explanations, why there has not been a "pressure dependent metrological validation", since the SI humidity is usually transferred at 1013 hPa. And there is also a clear reason why there was no metrological low humidity validation so far: Metrological low-humidity generators are usually operated with pure Nitrogen. This however changes the spectroscopy (N2 vs air broadening), so that a direct comparison with a primary standard is not possible.
So again – please – if the reviewer knows about any other "Pressure dependent absolute validation from 5 – 1200 ppmv at a metrological humidity generator", we would be very happy to read and include that.

From our knowledge, this is the first "Pressure dependent absolute validation from 5 – 1200 ppmv at a metrological humidity generator". This describes the novelty of the paper. Further, as this range coincides with as significant fraction of the atmospheric humidity range, we find that our paper is of broad interest for readers of AMT and thus well suited for a publication in AMT.

This criticism is not against the value of their work, which I certainly recognize and appreciate. My criticism is that the authors do not put their work into the proper context. AquaVIT and the discussion of the accuracy of stratospheric and upper tropospheric measurements is not the proper context.

We fully appreciate the discussion, but would also be very delighted if our arguments are recognized and discussed. For those readers which might also struggle to put our paper in the "right "context (which e.g. does neither include the second reviewer nor any of the numerous readers of the discussion paper, as there are no online comments), it is certainly very helpful that all reviewer discussions are also published with the manuscript. So the information to “clarify” this is certainly “nearby” and easily available. The idea to “treat” SEALDH-II as a specialized UTLS instrument was never our intention. This idea was entirely brought up by one of the two reviewers, we strongly objected to this idea from the first moment.

Individual, detailed comments:

Please note that the line numbers refer to the final manuscript version without changes markup.

Lines 71ff: Statements such as "... those often show results which are not sufficient for validation of atmospheric models in terms of the required absolute accuracy, precision, temporal resolution, long-term stability, comparability, etc." are in that generality overstated. However, they were certainly true in the past for some sets of stratospheric measurements.
=> We revised that

Lines 80-81: Another major issue limiting the comparison of hygrometers in flight is the issue of how ambient air is moved to the instrument. Issues such as contamination and sampling line problems are additional issues, which have plagues in situ comparisons in the past. This needs to be discussed as this is also a limiting factor for SEALDH-II in atmospheric measurements. The authors nicely point out that contamination and line issues are handled carefully in laboratory calibrations by allowing instruments and air samples to equilibrate over long times. This is not possible in atmospheric measurements, the consequences of which need to be discussed.
While we agree that is an issue for all (!) airborne measurements, this is a general operational problem when deploying any instrument into an environment where the measurement signal is temporarily fluctuating. However, we again have to stress that this paper focuses on "Pressure dependent **absolute validation** from 5 – 1200 ppmv at a metrological humidity generator" and not on any in-flight problems or field topics. Such problems are typically discussed in papers on filed campaigns. Therefore, we clearly focus here on an absolute validation (see line 81 and below). As a side note: The published peer reviewed information about the AquaVIT comparison also exclusively discussed static (!) i.e. "non-dynamic" concentration levels; a separation of the different influences is vital to identify the real cause of the deviations.
A dynamic characterization is not in the focus of this publication. Furthermore, there is - to our knowledge - no metrologically validated possibility to generate and provide a humidity generator capable to realize a metrologically defined "step function" to validate the temporal behavior of a humidity sensor

Generally, this is NOT specifically a limiting factor of SEALDH-II rather than a limiting factor for every extractive instrument. What's more, the "limitations" highly depend on the gas sampling system itself (e.g. type of inlet, pipe length, pipe coding, pipe temperature (heated?), flow, humidity level, etc.). Therefore, an accurate statement would only make sense if a full system was analyzed, which in the vast majority of papers on airborne hygrometers is NOT described in detail again due to the lack of suitable dynamic "step" reference generators.

=> We added a few words for clarification

Line 149: "…The most holistic approach …" This wording should be changed. Better wording is "…most extensive approach…"
=> Changed

Lines 152 ff.: Delete lines 152 – 154 and better write: "SEALDH-II is described in detail in Buchholz et al., (2016)"

=> Revised

Line 174: Here the authors should explicitly state that the uncertainty of the lower end of the mixing ratio measurement range at 3ppm is 100% due to the offset component. Here the authors need to be clear that this instrument is not suitable for lower stratospheric measurements and only somewhat useful in the upper troposphere compared to other instruments, which have a better claimed (although not validated) uncertainty.

We wrote:
"SEALDH-II's calculated linear part of the measurement uncertainty is 4.3%, with an additional offset uncertainty of ±3 ppmv" (…) SEALDH-II's measurement range covers 3 – 40000 ppmv. The calculated mixture fraction offset uncertainty of ±3 ppmv defines the lower detection limit."

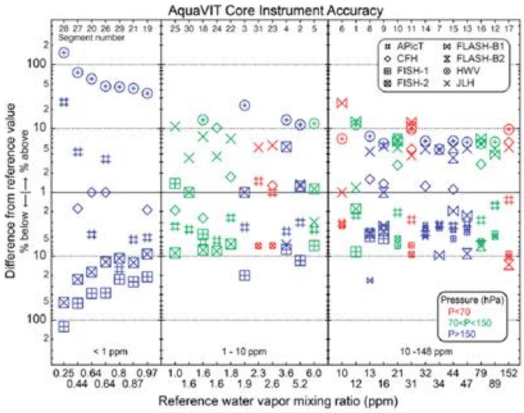
=> a) So we did exactly that - or did we misunderstood the reviewers comment?
=> b) We never (even if stated in the review many times) made the argument that SEALDH-II is in any kind a stratospheric instrument. On the contrary, we clearly stated e.g. in line 271 *"over a range which is particularly interesting for instruments on airborne platforms operating from troposphere to lower stratosphere where SEALDH-II's uncertainty (4.3% ± 3 ppmv) is suitable"*.
The word stratosphere is in the following contexts in the paper: a) explanation of the range of the atmosphere b) classification of typical operation ranges.
We have never and will never state that SEALDH-II is a stratospheric instrument.

Line 183: At least except for the two other instruments, the authors already built. This sentence should be deleted to avoid confusion.
Maybe there is a confusion with the line numbers: If it belongs to *"SEALDH-II's data treatment works differently from nearly all other published TDLAS spectrometers."* then this statement includes already the fact that only "nearly all" instruments do it differently. HAI and SEALDH-II are due to the first review so often cited in this paper that they just cannot be missed anymore - even if a reader reads just 20% of the publication.

Lines 280f.: "…over a range which is particularly interesting for instruments on airborne platforms operating from troposphere to lower stratosphere where SEALDH-II's uncertainty (4.3% ± 3 ppmv) is suitable". Here the authors imply explicitly that the uncertainty of SEALDH-II is suitable for lower stratospheric measurements, where in fact the numbers in that same sentence clearly say the opposite. An uncertainty of 3 ppmv is not suitable for lower stratospheric measurements.
We are a bit confused how this sentence can possibly be misunderstood. A reader who is not in the mindset of "forcing" SEALDH-II to be a stratospheric instrument will read the sentence as follows: a) Instrument has a range from 3 – 40000 ppmv with an uncertainty of (4.3% ± 3 ppmv). b) If he wants to measure 3 ppmv with that instrument, then he would need to use the provide uncertainty information, which yields a calculated uncertainty of 100% uncertainty at 3 ppm. This is exactly what we wrote and what the reviewer nevertheless requests?? By the way, SEALDH-II's "measured" value could still be much less noisy and imply a higher precision. But the uncertainty refers to the absolute accuracy and thus is defined by our statements as we targeted an "absolute validation". c) If he wants to measure 10 ppmv the uncertainty will be around 30%, at 100 ppmv around 5% and so on.
The part of the sentence "where SEALDH-II's uncertainty (4.3% ± 3 ppmv) is suitable" stated exactly

that.
Without starting another lengthy discussion, the reviewer might also think about the difference between a "calculated uncertainty" and the "measured uncertainty/error/deviation". The data in figure 8 show that the "measured deviation" at 5ppmv and 100 hPa is just around 5-10% (and not 60% like the calculated uncertainty provides). Therefore, most instruments (which do not have the possibility to calculate an uncertainty due to the physical detection principle and flowing evaluation) would need to state this value (maybe with a scaling factor) as an uncertainty. Therefore - **we fully agree that SEALDH-II is NOT a stratospheric instrument** - it can be used in lower stratospheric conditions, however. But we cannot guarantee the absolute accuracy under this conditions from a metrological point of view, but most likely the deviations would be in the same range like during the validation (and not 100% at 3 ppmv). An uncertainty is NOT a typical deviation – this might another reason for the confusion.

Lines 311-320: As said above, the core instruments of AquaVIT were built for upper tropospheric and stratospheric measurements in mind. In this sentence the uncertainties at the comfortable range for SEALDH-II are again compared to the challenging regime explored during AquiVIT, where SEALDH-II has itself a very large uncertainty. This is not appropriate and these sentences should be deleted.
Here again the final graph of AquaVIT:



The range which is covered here is up to 150 ppmv. We presented in our manuscript measurements which are well overlapping with this concentration range, namely at: 100 ppmv, 75 ppmv, 50 ppmv, 35 ppmv, 20 ppmv, 15 ppmv, 10 ppmv, 5 ppmv; and at each concentration we measured 6 different pressure. ; therefore 48 different validation steps. The ranges of the instrument used during AquaVIT are partly up to several 1000 ppmv. Every hygrometer has a certain measurement range, so does SEALDH-II.
Therefore, we cannot understand this comment why this comparison is not valid from the reviewer's point of view and why we should exclude the reference to Aquavit. In particular, it should be noted here that we actually participated in AQUAVIT with the APICT instrument (mainly developed by our TDLAS group at Heidelberg University) and that all the data from APICT were evaluated by our group. Furthermore, I was the co-initiator of AQUAVIT, together with colleagues from KIT and FZJ. Thus, we have been very closely involved in AQUAVIT.

Lines 375-378: This has been pointed out before and should be deleted here.
It is not uncommon to skim over publications and this very important information should therefore be repeated here because it is vital for the assessment.

Lines 389ff.: In the evaluation it is not acceptable to ignore the 65 and 125 hPa levels. The title of the paper clearly refers to airborne measurements; therefore, the validation effort needs to consider the atmospheric distribution of water vapor and not discuss how the validation result might look like without these levels.

"If one ignores the 65 hPa and 125 hPa measurements, which are clearly affected by higher order line shape effects (see above), the other measurements fit fairly well in a ±1 ppmv envelope function (grey)."

a) The sentence started with "if", which means that the second part of the sentence is only true if the first part is true as well.
b) We describe that this effect is visible; we do not infer any(!) performance data which are related to any "full performance statements". Higher order lines shape effects (which are here nicely visible in figure 8) are in spectroscopy a commonly seen artefact if the Voigt line shape profile is used for spectral analysis.

Line 403: Delete "To prevent further interpretations"

=> done

Lines 405 ff: better "This is an essential difference between calibration free instruments and …"

=> revised

Line 407: better "SEALDH-II tries to guarantee …"

The authors can never guarantee that ALL effects have been considered. They can only guarantee that all effects they know about have been considered.

=> revised

Line 428 f: Delete "novel" and "holistic"

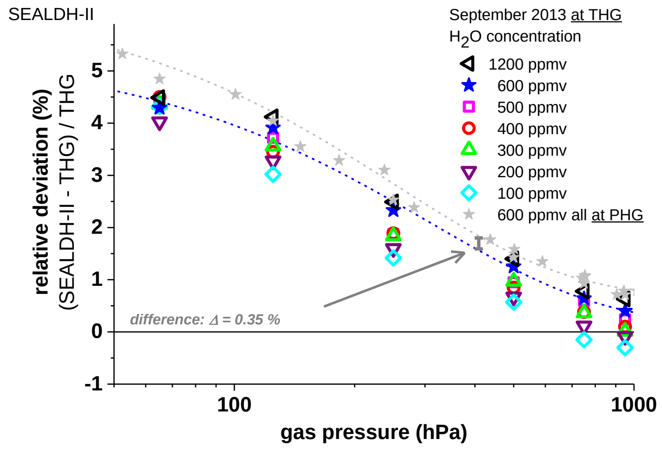
=> I hope we clarified that in the text above that a) SEALDH-II's concept is novel (even if the publication is not about the instrument's setup) and b) the control of the "core spectrometer" is intense - compared to standard TDLAS spectrometer. Therefore, I guess the comment is obsolete.

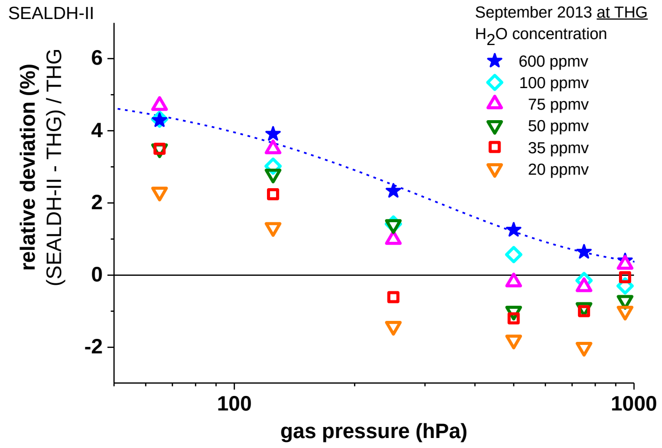
Lines 451 ff: I would hope that the authors will as well validate HAI, which has been built by them and which is much more suitable for atmospheric measurements. Given the use of that instrument, validating it would be more than useful.

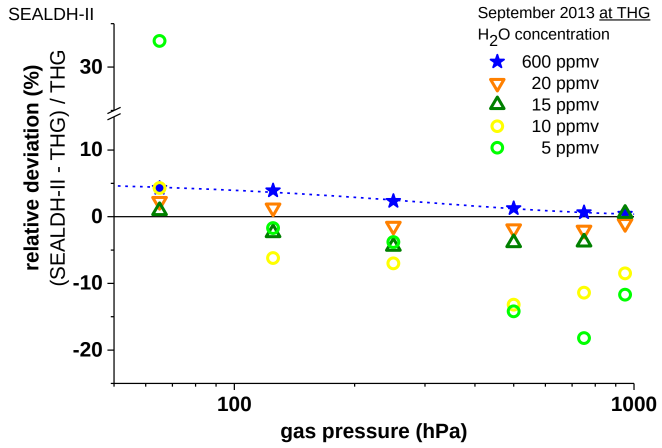
=> Our main goal is to valid a concept which is deployed in different instruments. The instrument by itself is then just a representation of the concept. Therefore, several validation results are directly transferable to HAI, in particularly to the HAI's extractive signal channels.

Figure 6-8: I maintain my earlier comment that the abscissa should be log(P), since the authors write about an instrument for airborne use, not laboratory use. For many atmospheric measurements, in particular aircraft based measurements, it is much easier to relate to altitude which varies nearly linear with log(P) than with pressure itself. This emphasizes the entire tropospheric distribution better than linear pressure.

We would agree if SEALDH-II was presented here as a stratospheric instrument, which is the omnipresent idea of the reviewer. We think that the proposed graphical representation would really make the "main results graphs" much more difficult to understand. We will put the graphs with a log (p) scale here; so everyone can find the graphs in this representation in the discussion part.







[1] B. Buchholz, N. Böse, S. Wagner, and V. Ebert, “Entwicklung eines rückführbaren, selbstkalibrierenden, absoluten TDLAS-Hygrometers in kompakter 19" Bauweise,” in *AMA-Science, 16. GMA/ITG-Fachtagung Sensoren und Messsysteme 2012*, (2012), pp. 315–323, doi:10.5162/sensoren2012/3.2.3.

[2] B. Buchholz, N. Böse, and V. Ebert, “Absolute validation of a diode laser hygrometer via

intercomparison with the German national primary water vapor standard," *Applied Physics B*, vol. 116, no. 4, pp. 883–899, (2014), doi:10.1007/s00340-014-5775-4.

Absolute, pressure dependent validation of a calibration-free, airborne laser hygrometer transfer standard (SEALDH-II) from 5 – 1200 ppmv using a metrological humidity generator

~~SEALDH-II – a calibration-free transfer standard for airborne water vapor measurements: Pressure dependent absolute validation from 5 – 1200 ppmv at a metrological humidity generator~~

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Abstract

Highly accurate water vapor measurements are indispensable for understanding a variety of scientific questions as well as industrial processes. While in metrology water vapor concentrations can be defined, generated and measured with relative uncertainties in the single percentage range, field deployable airborne instruments deviate even under quasi-static laboratory conditions up to 10-20%. The novel SEALDH-II hygrometer, a calibration-free, tuneable diode laser spectrometer, bridges this gap by implementing a new holistic concept to achieve higher accuracy levels in the field. Here we present the absolute validation of SEALDH-II at a traceable humidity generator during 23 days of permanent operation at 15 different H_2O concentration levels between 5 and 1200 ppmv. At each concentration level, we studied the pressure dependence at 6 different gas pressures between 65 and 950 hPa. Further, we describe the setup for this metrological validation, the challenges to overcome when assessing water vapor measurements on a high accuracy level, as well as the comparison results. With this validation, SEALDH-II is the first airborne, metrologically validated humidity transfer standard which links several scientific airborne and laboratory measurement campaigns to the international metrological water vapor scale.

1. Introduction

Water vapor affects, like no other substance, nearly all atmospheric processes (Ludlam, 1980; Möller et al., 2011; Ravishankara, 2012). Water vapor represents not only a large direct feedback to global warming when forming clouds, but also plays a major role in atmospheric chemistry (Held and Soden, 2000; Houghton, 2009; Kiehl and Trenberth, 1997). Changes in the water distribution, as vapor or in condensed phases (e.g. in clouds), have a large impact on the radiation balance of the atmosphere. This justifies that water vapor is often mentioned as the most important greenhouse gas and one of the most important parameters in climate research (Ludlam, 1980; Maycock et al., 2011). Water vapor measurements are often needed for

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other in-situ atmospheric analyzers to correct for their water vapor cross-interference. The high (spatial and temporal) variability of atmospheric water vapor, its large dynamic range (typically 3 – 40 000 ppmv), and its broad spectroscopic fingerprint typically require complex multi-dimensional calibrations, in particular for spectroscopic sensors. These calibrations often embrace the water vapor content of the gas flow to be analyzed as one of the key calibration parameters even if the instrument (e.g. for CO_2), is not intended to measure water vapor at all.

In particular for field weather stations, water vapor analyzers often are seen as part of the standard instrumentation in atmospheric research. This seems reasonable due to several reasons: slow H_2O concentration change over hours, the typical mid-range humidity levels (approx. above 5000 ppmv), no significant gas pressure or temperature change, target accuracy often only in the on the order of 5-15% relative deviation, and the absence of “non-typical atmospheric components” such as soot or hydrophobic substances. Water vapor measurements under these conditions can be performed by a variety of different devices (Wiederhold, 1997): Capacitive polymer sensors e.g. (Salasmaa and Kostamo, 1986) are frequently deployed in low cost (field) applications. Small-scale produced, commercially available spectral absorption devices e.g. (Petersen et al., 2010) are often used in research campaigns. Dew-point mirror hygrometers (DPM) are known for their high accuracy. However, this is only true if they are regularly calibrated at high accuracy (transfer-) standards in specialized hygrometry laboratories such as in metrology institutes (Heinonen et al., 2012).

As soon as hygrometers have to be deployed in harsh environments (e.g. on airborne platforms), this situation changes entirely: The ambient gas pressure (10 – 1000 hPa) and gas temperature (-90 – 40°C) ranges are large and both values change rapidly, the required H_2O measurement range is set by the ambient atmosphere (typically 3 – 40000 ppmv), mechanical stress and vibrations occur, and the sampled air contains additional substances from condensed water (ice, droplets), particles, or even aircraft fuel vapor (e.g. on ground). These and other impacts complicate reliable, accurate, long-term stable H_2O measurements and briefly outline why water vapor measurements remain difficult in-situ measurements in the field, even if they are nearly always needed in atmospheric science. Usually, the availability and coverage of observations limit model validation studies in the first place but also the lack of sufficient accuracy may have limited important scientific interpretations (Krämer et al., 2009; Peter et al., 2006; Scherer et al., 2008; Sherwood et al., 2014).

Over the last decades, numerous hygrometers were developed and deployed on aircraft (Buck, 1985; Busen and Buck, 1995; Cerni, 1994; Desjardins et al., 1989; Diskin et al., 2002; Durry et al., 2008; Ebert et al., 2000; Gurlit et al., 2005; Hansford et al., 2006; Helten et al., 1998; Hunsmann et al., 2008; Karpechko et al., 2014; Kley and Stone, 1978; May, 1998; Meyer et al., 2015; Ohtaki and Matsui, 1982; Roths and Busen, 1996; Salasmaa and Kostamo, 1986; Schiff et al., 1994; Silver and Hovde, 1994a, 1994b; Thornberry et al., 2014; Webster et al., 2004; Zöger et al., 1999a, 1999b) (non-exhaustive list). While for some atmospheric questions the quality level of the data often is sufficient (e.g. typically climatologies), there are also a variety of questions, especially validation of atmospheric models, where the required absolute accuracy, precision, temporal resolution, long-term stability, comparability, etc. needs to be higher. These problems can be

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grouped into two major categories: accuracy linked problems and time response linked problems. The latter is particularly important for investigations in heterogeneous regions in the lower troposphere as well as for investigations in clouds. In these regions, even two on average agreeing instruments with different response times yield local, large, relative deviations on the order of up to 30% (Smit et al., 2014). Important to keep in mind is that the total time response of a system is a superposition of the instrument's time response plus the sampling system's time response which typically depends on many parameters (e.g. type of inlet, pipe length, pipe coding, pipe temperature (heated?), flow, humidity level, etc.).

In contrast to time response studies, accuracy linked problems in flight are difficult to isolate since they are always covered by the spatial variability (which leads to temporal variability for moving aircraft) of atmospheric H_2O distribution. Comparing hygrometer in flight, such as, for example in (Rollins et al., 2014), does not facilitate a clear accuracy assessment.

Therefore in 2007, an international intercomparison exercise named "AquaVIT" (Fahey et al., 2014) was carried out to compare airborne hygrometers under quasi-static, laboratory-like conditions for upper tropospheric and stratospheric humidity levels. AquaVIT (Fahey et al., 2014) encompassed 22 instruments from 17 international research groups. The instruments were categorized in well-validated, often deployed "core" instruments (APicT, FISH, FLASH, HWV, JLH, CFH) and "younger" non-core instruments. AquaVIT revealed in the important 1 to 150 ppmv H_2O range, that -even under quasi-static conditions- the deviation between the core instrument's readings and their averaged group mean was on the order of ±10 %. This result fits to the typical interpretation problems of flight data where instruments often deviate from each other by up to 10%, which is not covered by the respective uncertainties of the individual instruments. AquaVIT was a unique first step to document and improve the accuracy of airborne measurements in order to make them more comparable. However, no instrument could claim after AquaVIT that its accuracy is higher than any other AquaVIT instrument, since no "gold standard" was part of the campaign, i.e., a metrological transfer standard (JCGM 2008, 2008; Joint Committee for Guides in Metrology (JCGM), 2009) traced back to the SI units. There is no physical argument for the average being better than the measured value of a single instrument. Instead, many arguments speak for systematic deviations of airborne hygrometers: Most hygrometers have to be calibrated. Even for a perfect instrument, the accuracy issue is represented by the calibration source and its gas handling system, which in this case leads to two major concerns: First, one has to guarantee that the calibration source is accurate and stable under field conditions, i.e., when using it before or after a flight on the ground. This can be challenging especially for the transportation of the source with all its sensitive electronics/mechanics and for the deviating ambient operation temperature from the ambient validation temperature (hangar vs. laboratory). Even more prone to deviations are calibration sources installed inside the aircraft due to changing ambient conditions such as cabin temperature, cabin pressure, orientation angle of instrument (important, if liquids are used for heating or cooling). Secondly, the gas stream with a highly defined amount of water vapor has to be conveyed into the instrument. Especially for water vapor, which is a strongly polar molecule, this gas transport can become a critical step. Changing from high to low concentrations or even just changing the gas pressure or pipe temperature can lead to signal creep due to slow adsorption and desorption processes, which can take long to equilibrate. In metrology, this issue is solved by a long validation/calibration time (hours up to weeks, depending on the H_2O concentration level), a generator without any connectors/fittings (everything is welded) and piping made out of electro-polished, stainless steel to ensure that the equilibrium is established before the actual calibration process is started. However, this calibration approach is difficult to deploy and maintain for aircraft/field operations due to the strong atmospheric variations in gas pressure and H_2O concentrations, which usually leads to a multi-dimensional calibration pattern (H_2O concentration, gas pressure, sometimes also gas temperature) in a short amount of calibration time (hours). Highly sensitive, frequently flown hygrometers like (Zöger et al., 1999a) are by their physical principle, not as long-term stable as it would be necessary to take advantage of a long calibration session. Besides the time issue to reach a H_2O equilibrium between source and instrument, most calibration principles for water vapor are influenced by further issues. A prominent example is the saturation of air in dilution/saturation based water vapor generators: gas temperature and pressure defines the saturation level (described e.g. by Sonntag's Equation (Rollins et al., 2014)), however, it is well-known that e.g. 100.0% saturation is not easily achievable. This might be one of the impact factors for a systematic offset during calibrations in the field. The metrology community solves this for high humidity levels with large, multi-step saturation chambers which decrease the temperature step-wise to force the water vapor to condense in every following step. These few examples of typical field-related problems show, that there is a reasonable doubt that deviations in field situations are norm-distributed. Hence, the mean during AquaVIT might be biased, i.e. not the correct H_2O value.

The instruments by themselves might actually be more accurate than AquaVIT showed, but deficiencies of the different calibration procedures (with their different calibration sources etc.) might mask this. To summarize, AquaVIT documented a span of up to 20% relative deviation between the world's best airborne hygrometers – but AquaVIT could not assess absolute deviations nor explain them, since a link to a metrological H_2O primary standard (i.e., the definition of the international water vapor scale) was missing.

Therefore, we present in this paper the first comparison of an airborne hygrometer (SEALDH-II) with a metrological standard for the atmospheric relevant gas pressure (65 – 950 hPa) and H_2O concentration range (5 – 1200 ppmv). We will discuss the validation setup, procedure, and results. Based on this validation, SEALDH-II is by definition the first airborne transfer standard for water vapor which links laboratory and field campaigns directly to metrological standards.

2. SEALDH-II

2.1. System description

This paper focuses on the metrological accuracy validation of the **S**elective **E**xtractive **A**irborne **L**aser **D**iode **H**ygrometer (SEALDH-II). SEALDH-II is the airborne successor of the proof-of-concept spectrometer (SEALDH-I) study published in (Buchholz et al., 2014), which showed the possibility and the achievable accuracy level for calibration-free dTDLAS hygrometry. The publication (Buchholz et al., 2014) demonstrates this for the 600 ppmv to 20000 ppmv range at standard ambient pressure). The instruments SEALDH-I, SEALDH-II and also HAI (Buchholz et al., 2017) are all three built with the design philosophy that every single reported value of the instrument should have a "related boundary/operation condition snap shot" allowing to exclude the possibility of any instrumental malfunction during the measurement. SEALDH-II is from this perspective the most extensive approach (capturing much more boundary condition data (Buchholz et al., 2016)), while HAI can serve as a multi-channel, multi-phase hygrometer for a broader variety of scientific questions.

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SEALDH-II integrates numerous different principles, concepts, modules, and novel parts, which contribute to or enable the results shown in this paper. SEALDH-II is described in detail in (Buchholz et al., 2016). The following brief description covers the most important technical aspects of the instrument from a user's point of view:

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Gelöscht: for more details the reader is referred to

SEALDH-II is a compact (19" rack 4 U (=17.8 cm)) closed-path, absolute, directly Tunable Diode Laser Absorption Spectroscopy (dTDLAS) hygrometer operating at 1.37 µm. With its compact dimensions and the moderate weight (24 kg), it is well suited for space- and weight-limited airborne applications. The internal optical measurement cell is a miniaturized White-type cell with an optical path length of 1.5 m (Kühnreich et al., 2016; White, 1976). It is connected to the airplane's gas inlet via an internal gas handling system comprising a temperature exchanger, multiple temperature sensors, a flow regulator, and two gas pressure sensors.

Approximately 80 different instrument parameters are controlled, measured, or corrected by SEALDH-II at any time to provide a holistic view on the spectrometer status. This extensive set of monitoring data ensures reliable and well-characterized measurement data at any time. The knowledge about the instruments status strongly facilitates metrological uncertainties calculations. SEALDH-II's calculated linear part of the measurement uncertainty is 4.3%, with an additional offset uncertainty of ±3 ppmv (further details in (Buchholz et al., 2016)). The precision of SEADLH-II was determined via the Allan-variance approach and yielded 0.19 ppmv (0.17 ppmv·m·Hz-½) at 7 Hz repetition rate and an ideal precision of 0.056 ppmv (0.125 ppmv·m·Hz-½) at 0.4 Hz. In general, SEALDH-II's time response is limited by the gas flow through the White-type multi-pass measurement cell with a volume of 300 ml. With the assumption of a bulk flow of 7 SLM at 200 hPa through the cell, the gas exchange time is 0.5 seconds.

SEALDH-II's measurement range covers 3 – 40000 ppmv. The calculated mixture fraction offset uncertainty of ±3 ppmv defines the lower detection limit. This offset uncertainty by itself is entirely driven by the capability of detecting and minimizing parasitic water vapor absorption. The concept, working principle, and its limits are described in (Buchholz and Ebert, 2014). The upper limit of 40000 ppmv is defined by the lowest internal instrument temperature, which has to always be higher than the dew point temperature to avoid any internal condensation. From a spectroscopic perspective, SEALDH-II could handle concentrations up to approx. 100000 ppmv before spectroscopic problems like saturation limit the accuracy and increase the relative uncertainty beyond 4.3%.

2.1. Calibration-free evaluation approach

SEALDH-II's data treatment works differently from nearly all other published TDLAS spectrometers. Typically, instruments are setup in a way that they measure the absorbance or a derivative measurand of absorbance, and link it to the H_2O concentration. This correlation together with a few assumptions about long-term stability, cross interference, gas temperature dependence, gas pressure dependence is enough to calibrate a system (Muecke et al., 1994). Contrarily, a calibration-free approach requires a fully featured physical model describing the absorption process entirely. The following description is a brief overview; for more details see e.g. (Buchholz et al., 2014, 2016; Ebert and Wolfrum, 1994; Schulz et al., 2007).

In a very simplified way, our physical absorption model uses the *extended* Lambert-Beer equation (Equation 1) which describes the relationship between the initial light intensity $I_0(\lambda)$ before the absorption path (typically being in the few mW-range) and the transmitted light intensity $I(\lambda)$.

Equation 1: $I(\lambda) = E(t) + I_0(\lambda) \cdot Tr(t) \cdot exp[-S(T) \cdot g(\lambda - \lambda_0) \cdot N \cdot L]$

The parameter S(T) describes the line strength of the selected molecular transition. In SEADLH-II's case, the spectroscopic multi-line fit takes into account 19 transition lines in the vicinity of the target line at 1370 nm (energy levels: 110 – 211, rotation-vibrational combination band). The other parameters are the line shape function $g(\lambda - \lambda_0)$, the absorber number density N, the optical path length L and corrections for light-type background radiation E(t) and broadband transmission losses Tr(t).

Equation 1 can be enhanced with the ideal gas law to calculate the H_20 volume mixing ratio c:

Equation 2: $$c = -\frac{k_B \cdot T}{S(T) \cdot L \cdot p} \int ln\left(\frac{I(\nu) - E(t)}{I_0(\nu) \cdot Tr(t)}\right) \frac{d\nu}{dt} dt$$

The additional parameters in Equation 2 are: constant entities like the Boltzmann constant k_B; the optical path length L; molecular constants like the line strength S(T) of the selected molecular transition; the dynamic laser tuning coefficient $\frac{d\nu}{dt}$, which is a constant laser property; continuously measured entities such as gas pressure (p), gas temperature (T) and photo detector signal of the transmitted light intensity I(ν) as well as the initial light intensity $I_0(\nu)$, which is retrieve during the evaluation process from the transmitted light intensity I(ν).

Equation 2 facilitates an evaluation of the measured spectra without any instrument calibration at any kind of water vapor reference (Buchholz et al., 2014; Ebert and Wolfrum, 1994; Schulz et al., 2007) purely based on first principles. Our concept of a fully calibration-free data evaluation approach (this excludes also any referencing of the instrument to a water standard in order to correct for instrument drift, offsets, temperature dependence, pressure dependence, etc.) is crucial for the assessment of the results described in this publication. It should be noted that the term "calibration-free" is frequently used in different communities with dissimilar meanings. We understand this term according to the following quote (JCGM 2008, 2008): "calibration (…) in a first step, establishes a relation between the measured values of a quantity with measurement uncertainties provided by a measurement standard (…), in a second step, [calibration] uses this information to establish a relation for obtaining a measurement result from an indication (of the device to be calibrated)". Calibration-free in this sense means, that SEALDH-II does not use any information from "calibration-, comparison-, test-, adjustment-" runs with respect to a higher accuracy "water vapor standard" to correct or improve any response function of the instrument. SEALDH-II uses as described in (Buchholz et al., 2016) only spectroscopic parameters and the 80 supplementary parameters as measurement input to calculate the final H_2O concentration. The fundamental difference between a calibration approach and this stringent concept is that only effects which are part of our physical model are taken into account for the final H_2O concentration calculation. All other effects like gas pressure or temperature dependencies, which cannot be corrected with a well-defined physical explanation, remain in our final results even if this has the consequence of slightly uncorrected results deviations. This strict philosophy leads to measurements which are very reliable with respect to accuracy, precision and the instrument's over-all performance. The down-side is a relatively computer-intensive, sophisticated evaluation. As SEALDH-II stores all the raw spectra, one could – if needed for whatever reason – also calibrate the instrument by referencing it to a high accuracy water vapor standard and transfer the better accuracy e.g. of a metrological standard onto the instrument. Every calibration-free instrument can be calibrated since pre-requirements for a calibration are just a subset of the requirements for a calibration-free instrument. However, a calibration can only improve the accuracy for the relatively short time between two calibration-cycles by adding all uncertainty contributions linked to the calibration itself to the system. This is unpleasant or even intolerable for certain applications and backs our decision to develop a calibration-free instrument to enable a first principles, long-term stable, maintenance-free and autonomous hygrometer for field use e.g. at remote sites or aircraft deployments.

3. SEALDH-II validation facility

3.1. Setup

Figure 1 right shows the validation setup. As a well-defined and highly stable H_2O vapor source, we use a commercial Thunder scientific model (TSM) 3900, similar to (Thunder-Scientific, 2016). This source saturates pre-dried air at an elevated gas pressure in an internally ice covered chamber. The gas pressure in the chamber and the chamber's wall temperature are precisely controlled and highly stable and thus define the absolute water vapor concentration via the Sonntag equation (Sonntag, 1990). After passing through the saturator, the gas expands to a pressure suitable for the subsequent hygrometer. The pressure difference between the saturation chamber pressure and the subsequent step give this principle its name "two pressure generator". The stable H_2O concentration range of the TSM is 1 – 1300 ppmv for these specific deployment conditions. This generator provides a stable flow of approximately 4 – 5 SLM. Roughly 0.5 SLM are distributed to a frost/dew point hygrometer, D/FPH, (MBW 373) (MBW Calibration Ltd., 2010). SEALDH-II is fed with approx. 3.5 SLM, while 0.5 SLM are fed to an outlet. This setup ensures that the dew

Gelöscht: Figure 1

point mirror hygrometer (DPH)[1] operates close to the ambient pressure, where its metrological primary calibration is valid, and that the gas flow is sufficiently high in any part of the system to avoid recirculation of air. The vacuum pump is used to vary the gas pressure in SEALDH-II's cell with a minimized feedback on the flow through the D/FPH and the TSM. This significantly reduces the time for achieving a stable equilibrium after any gas pressure change in SEALDH-II's chamber. SEALDH-II's internal electronic flow regulator limits the mass flow at higher gas pressures and gradually opens towards lower pressures (vacuum pumps usually convey a constant volume flow i.e., the mass flow is pressure dependent). We termed this entire setup "traceable humidity generator", THG, and will name it as such throughout the text.

3.1. Accuracy of THG

The humidity of the gas flow is set by the TSM generator but the absolute H_2O values are traceably determined with the dew point mirror hygrometer (D/FPH). The D/FPH, with its primary calibration, thus guarantees the absolute accuracy in this setup. The D/FPH is not affected by the pressure changes in SEADLH-II's measurement cell and operates at standard ambient gas pressure and gas temperature where its calibration is most accurate. The D/FPH was calibrated (Figure 2) at the German national standard for mid-range humidity (green, 600 – 8000 ppmv) as well as at the German national standard for low-range humidity (blue, for lower values 0.1 – 500 ppmv). The two national standards work on different principles: The two pressure principle (Buchholz et al., 2014) currently supplies the lower uncertainties (green, "±"-values in Figure 2). Uncertainties are somewhat higher for the coulometric generator (Mackrodt, 2012) in the lower humidity range (blue). The "Δ"-values in Figure 2 show the deviations between the readings of the D/FPH and the "true" values of the national primary standards.

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4. SEALDH-II validation procedure

4.1. Mid-term multi-week permanent operation of SEALDH-II

One part of the validation was a permanent operation of SEALDH-II over a time scale much longer than the usual air or ground based scientific campaigns. In this paper, we present data from a permanent 23 day long (550 operation hours) operation in automatic mode. Despite a very rigorous and extensive monitoring of SEALDH-II's internal status, no malfunctions of SEALDH-II could be detected. One reason for this are the extensive internal control and error handling mechanisms introduced in SEALDH-II, which are mentioned above and described elsewhere (Buchholz et al., 2016). Figure 3 shows an overview of the entire validation. The multi-week validation exercise comprises 15 different H_2O concentration levels between 2 and 1200 ppmv. At each concentration level, the gas pressure was varied in six steps (from 65 to 950 hPa) over a range which is particularly interesting for instruments on airborne platforms operating from

Gelöscht: Figure 3

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[1] The used dew point mirror hygrometer can measure far below 0°C; therefore, it is a dew point mirror above > 0°C and a frost point mirror as soon as there is ice on the mirror surface. We will use both DPH and D/FPH abbreviations interchangeably.

troposphere to lower stratosphere where SEALDH-II's uncertainty (4.3% ± 3 ppmv) is suitable. Figure 3 (top) shows the comparison between SELADH-II (black line) and the THG setup (red). Figure 3 (bottom) shows the gas pressure (blue) and the gas temperature (green) in SEALDH-II measurement cell. The gas temperature increase in the second week was caused by a failure of the laboratory air conditioner that led to a higher room temperature and thus higher instrument temperature. Figure 4 shows the 200 hPa section of the validation in Figure 3. To avoid any dynamic effects from time lags, hysteresis of the gas setup, or the instruments themselves, every measurement at a given concentration/pressure combination lasted at least 60 min. The data from the THG (red) show that there is nearly no feedback of a gas pressure change in SEALDH-II's measurement cell towards the D/FPH, respectively the entire THG. The bottom subplot in Figure 4 shows the relative deviation between the THG and SEALDH-II. This deviation is correlated to the absolute gas pressure level and can be explained by deficiencies of the Voigt lines shape used to fit SEALDH-II's spectra (Buchholz et al., 2014, 2016). The Voigt profile, a convolution of Gaussian (for temperature broadening) and Lorentzian (pressure broadening) profiles used for SEALDH-II's evaluation, does not include effects such as Dicke Narrowing, which become significant at lower gas pressures. Neglecting these effects cause systematic, but long-term stable and fully predictable deviations from the reference value in the range from sub percent at atmospheric gas pressures to less than 5 % at the lowest gas pressures described here. We have chosen not to implement any higher order line shape (HOLS) models as the spectral reference data needed are not available at sufficient accuracy. Further, HOLS would force us to increase the number of free fitting parameters, which would destabilize our fitting procedure, and lead to reduced accuracy/reliability (i.e., higher uncertainty) as well as significantly increased computational efforts. This is especially important for flight operation where temporal H_2O fluctuations (spatial fluctuations result in temporal fluctuations for a moving device) occur with gradients up to 1000 ppmv/s.

Gelöscht: Figure 3

Gelöscht: Figure 3

Gelöscht: Figure 4

Gelöscht: Figure 4

These well understood, systematic pressure dependent deviations will be visible in each further result plot of this paper. The impact and methods of compensation are already discussed in (Buchholz et al., 2014). The interested reader is referred to this publication for a more detailed analysis and description.

SEALDH-II's primary target areas of operations are harsh field environments. Stability and predictability is to be balanced with potential, extra levels of accuracy which might not be required or reliably achievable for the intended application. Higher order line shape models are therefore deliberately traded for a stable, reliable, and unified fitting process under all atmospheric conditions. This approach leads to systematic, predictable deviations in the typical airborne accessible atmospheric gas pressure range (125 – 900 hPa) of less than 3%. One has to compare these results for assessment to the non-systematic deviations of 20% revealed during the mentioned AquaVIT comparison campaign (Fahey et al., 2014). Hence, for field/airborne purposes, the 3% seems to be fully acceptable – especially in highly H_2O structured environments.

This comparison with AquaVIT should just provide a frame to embed the 3%. The H_2O concentration range of Aquavit (0 – 150 ppmv) versus this validation range (5 -1200 ppmv) and the instruments configuration at AquaVIT (mainly (upper) stratospheric hygrometers) versus SEALDH-II as a wide range instrument (3 – 40000 ppmv) do not allow a direct comparison. Sadly, there is no other reliable (representative for the community, externally reviewed, blind submission, etc.) comparison exercise such as AquaVIT for higher concentration ranges.

4.1. Assessment of SEALDH-II's mid-term accuracy: Dynamic effects

Besides the pressure dependence discussed above, SEALDH-II's accuracy assessment is exacerbated by the differences in the temporal behavior between the THG's dew/frost point mirror hygrometer (D/FPH) and SEALDH-II: Figure 5 (left) shows an enlarged 45 min. long section of measured comparison data. SEALDH-II (black) shows a fairly large water vapor variation compared to the THG (red). The precision of SEALDH-II (see chapter 2) is 0.056 ppmv at 0.4 Hz (which was validated at a H_2O concentration of 600 ppmv (Buchholz et al., 2016)) yielding a signal to noise ratio of 10700. Therefore, SEALDH-II can very precisely detect variations in the H_2O concentration. Contrarily, the working principle of a D/FPH requires an equilibrated ice/dew layer on the mirror. Caused by the inertial thermal adjustment process, the response time of a dew/frost point mirror hygrometer has certain limitations due to this principle (the dew/frost point temperature measurement is eventually used to calculate the final H_2O concentration), whereas the optical measurement principle of SEALDH-II is only limited by the gas transport, i.e., the flow (exchange rate) through the measurement cell. The effect of those different response times is clearly visible from 06:00 to 06:08 o'clock in Figure 5. The gas pressure of SEALDH-II's measurement cell (blue), which is correlated to the gas pressure in the THG's ice chamber, shows an increase of 7 hPa – caused by the regulation cycle of the THG's generator (internal saturation chamber gas pressure change). The response in the THG frost point measurement (green, red) shows a significant time delay compared to SEALDH-II, which detects changes approx. 20 seconds faster. This signal delay is also clearly visible between 06:32 to 06:40 o'clock, where the water vapor variations detected by SEALDH-II are also visible in the smoothed signals of the THG. Figure 5 right shows such a variation in detail (5 min). The delay between the THG and SEALDH-II is here also approximately 20 seconds. If we assume that SEALDH-II measures (due to its high precision) the true water vapor fluctuations, the relative deviation can be interpreted as overshooting and undershooting of the D/FPH's controlling cycle, which is a commonly known response behavior of slow regulation feedback loops to fast input signal changes. The different time responses lead to "artificial" noise in the concentration differences between SEALDH-II and THG. Theoretically, one could characterize this behavior and then try to correct/shift the data to minimize this artificial noise. However, a D/FPH is fundamentally insufficient for a dynamic characterization of a fast response hygrometer such as SEALDH-II. Thus, the better strategy is to keep the entire system as stable as possible and calculate mean values by using the inherent assumption that under- and overshoots of the DPM affect the mean statically and equally. With this assumption, the artificial noise can be seen in the first order as Gaussian distributed noise within each pressure step (Figure 4) of at least 60 min. The error induced by this should be far smaller than the above discussed uncertainties of the THG (and SEALDH-II).

Gelöscht: Figure 5

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Gelöscht: Figure 4

5. Results

The results of this validation exercise are categorized in three sections according to the following conditions in atmospheric regions: mid-tropospheric range: 1200 – 600 ppmv (Figure 6), upper tropospheric range: 600 – 20 ppmv (Figure 7), and lower stratospheric range: 20 – 5 ppmv (Figure 8). This categorization is also justified by the relative influence of SEALDH-II's calculated offset uncertainty of ±3 ppmv (Buchholz and Ebert, 2014): At 1200 ppmv, its relative contribution of 0.25% is negligible compared to the 4.3% linear part of the uncertainty of SEALDH-II. At 5 ppmv, the relative contribution of the offset uncertainty is 60% and thus dominates the linear part of the uncertainty. Before assessing the following data, it should be emphasized again that SEALDH-II's spectroscopic first-principles evaluation was designed to rely on accurate spectral data instead of a calibration. SEALDH-II was never calibrated or referenced to any kind of reference humidity generator or sensor.

Gelöscht: Figure 6

Gelöscht: Figure 7

Gelöscht: Figure 8

5.1. The 1200 – 600 ppmv range

Figure 6 shows the summary of the pressure dependent validations in the 1200 – 600 ppmv range. Each of the 48 data points represents the mean over one pressure measurement section of at least 60 min (see Figure 4). A cubic polynomial curve fitted to the 600 ppmv results (blue) serves as an internal quasi-reference to connect with the following graphs. The 600 ppmv data (grey) are generated via a supplementary comparison at a different generator: The German national primary mid-humidity generator (PHG). This primary generator data at 600 ppmv indicate a deviation between PHG and THG of about 0.35 %, which is compatible with the uncertainties of the THG (see chapter 3.1) and the PHG (0.4%) (Buchholz et al., 2014). The PHG comparison data also allow a consistency check between the absolute values of (see Figure 2) the PHG (calibration-free), the THG (DPM calibrated) and SEALDH-II (calibration-free).

Gelöscht: Figure 6

Gelöscht: Figure 4

Gelöscht: Figure 2

5.2. The 600 – 20 ppmv range

In this range, the linear part of the uncertainty (4.3%) and the offset uncertainty (±3 ppmv) have both a significant contribution. Figure 7 shows a clear trend: The lower the concentration, the higher the deviation. We believe this is being caused by SEALDH-II's offset variation and will be discussed in the 20 – 5 ppmv range.

Gelöscht: Figure 7

5.3. The 20 – 5 ppmv range

The results in this range (Figure 8) are dominated by the offset uncertainty. It is important to mention at this point, that the ±3 ppmv uncertainties are calculated based on assumptions, design innovations, and several independent, synchronous measurements which are automatically done while the instrument is in operation mode (see publication (Buchholz et al., 2016; Buchholz and Ebert, 2014)). Hence, the calculated uncertainties resemble an upper uncertainty threshold; the real deviation could be lower than 3 ppmv. A clear assessment is fairly difficult since at low concentrations (i.e., low optical densities) several other effects

Gelöscht: Figure 8

occur together such as, e.g., optical interference effects like fringes caused by the very long coherence length of the used laser. However, Figure 9 (left) allows a rough assessment of the offset instability. This plot shows all the data below 200 ppmv, grouped by the gas pressure in the measurement cell. If one ignores the 65 hPa and 125 hPa measurements, which are clearly affected by higher order line shape effects (see above), the other measurements fit fairly well in a ±1 ppmv envelope function (grey). In other words, SEALDH-II's combined offset "fluctuations" are below 1 ppmv H_2O. All validation measurements done with SEALDH-II during the last years consistently demonstrated a small offset variability so that the observed offset error is around 0.6 ppmv — i.e., only 20% of the calculated ± 3 ppmv.

Gelöscht: Figure 9

5.4. General evaluation

Figure 9 presents a summary of all 90 analyzed concentration/pressure-pairs during the 23 days of validation. The calculated uncertainties (linear 4.3% and offset ±3 ppmv) of SEALDH-II are plotted in purple. This uncertainty calculation doesn't include line shape deficiencies and is therefore only valid for a pressure range where the Voigt profile can be used to represent all major broadening effects of absorption lines (Dicke, 1953; Maddaloni et al., 2010). This is the case above 250 hPa. The results at 950, 750, 500, 250 hPa show that the maximum deviations, derived from these measurements, can be described by: linear +2.5%, offset -0.6 ppmv.

Gelöscht: Figure 9

It should be noted that this result doesn't change the statement about SEALDH-II's uncertainties, since these are calculated and not based on any validation/calibration process. This is a significantly different approach between calibration-free instruments such as SEALDH-II and other classical spectroscopic instruments which rely on sensor calibration. SEALDH-II provides correctness of measurement values within its uncertainties because any effect which causes deviations has to be included in the evaluation model – otherwise it is not possible to correct for it.

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As mentioned before, any calibration-free instrument can be calibrated too (see e.g. (Buchholz et al., 2013)). However by doing so, one must accept to a certain extent loss of control over the system, especially in environments which are different from the calibration environment. For example, if a calibration was used to remove an instrumental offset, one has to ensure that this offset is long-term stable, which is usually quite difficult, as - shown by the example of parasitic water offsets in fiber coupled diode laser hygrometers (Buchholz and Ebert, 2014). Another option is to choose the recalibration frequency high enough; i.e., minimizing the drift amplitude by minimizing the time between two calibrations. This, however, reduces the usable measurement time and leads to considerable investment of time and money into the calibration process. For the case of SEALDH-II, a calibration of the pressure dependence – of course tempting and easy to do – would directly "improve" SEALDH-II's laboratory overall performance level from ±4.3% ±3 ppmv to **±0.35% ±0.3 ppmv**. At first glance, this "accuracy" would then be an improvement by a factor of 55 compared to the mentioned results of AquaVIT (Fahey et al., 2014). However, it is extremely difficult – if not impossible – to guarantee this performance and the validity of the calibration under harsh field conditions; instead SEALDH-II would "suffer" from the same typical calibration associated problems in stability and in predictability. Eventually, the calibration-free evaluation would define the trusted values and the "improvement", achieved by the calibration, would have to be used very carefully and might disappear eventually.

6. Conclusion and Outlook

The SEALDH-II instrument, a novel, compact, airborne, calibration-free hygrometer which implements a holistic, first-principles directly tuneable diode laser absorption spectroscopy (dTDLAS) approach was stringently validated at a traceable water vapor generator at the German national metrology institute (PTB). The pressure dependent validation covered a H_2O range from 5 to 1200 ppmv and a pressure range from 65 hPa to 950 hPa. In total, 90 different H_2O concentration/pressure levels were studied within 23 days of permanent validation experiments. Compared to other comparisons of airborne hygrometers - such as those studied in the non-metrological AquaVIT campaign (Fahey et al., 2014), where a selection of the best "core" instruments still showed an accuracy scatter of at least ± 10% without an absolute reference value - our validation exercise used a traceable reference value derived from instruments directly linked to the international dew-point scale for water vapor. This allowed a direct assessment of SEALDH-II's absolute performance with a relative accuracy level in the sub percent range. Under these conditions, SEALDH-II showed an excellent absolute agreement within its uncertainties which are 4.3% of the measured value plus an offset of ±3 ppmv (valid at 1013 hPa). SEALDH-II showed at lower gas pressures - as expected - a stable, systematic, pressure dependent offset to the traceable reference, which is caused by the line shape deficiencies of the Voigt line shape: e.g. at 950 hPa, the systematic deviation of the calibration-free evaluated results could be described by (linear +0.9%, offset -0.5 ppmv), while at 250 hPa the systematic deviations could be described by (linear +2.5%, offset -0.6 ppmv). If we suppress this systematic pressure dependence, the purely statistical deviation is described by linear scatter of ±0.35% and an offset uncertainty of ±0.3 ppmv.

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Due to its extensive internal monitoring and correction infrastructure, SEALDH-II is very resilient against a broad range of external disturbances and has an output signal temperature coefficient of only 0.026%/K, which has already been validated earlier (Buchholz et al., 2016). Therefore, these results can be directly transferred into harsh field environments. With this metrological, mid and upper atmosphere focused validation presented here, we believe SEALDH-II to be the first directly deployable, metrologically validated, airborne transfer standard for atmospheric water vapor. Having already been deployed in several airborne and laboratory measurement campaigns, SEALDH-II thus directly links for the first time, scientific campaign results to the international metrological water vapor scale.

Data availability

The underlying data for the results shown in this paper are raw spectra (time vs. photo current), which are compressed to be compatible with the instruments data storage. In the compressed state the total amount is approximately 6GB of binary data. Uncompressed data size is approx. 60 GB. We are happy to share these data on request.

Author Contributions

Bernhard Buchholz and Volker Ebert conceived and designed the experiments. Bernhard Buchholz performed the experiments; Bernhard Buchholz and Volker Ebert analyzed the data and wrote the paper.

Conflicts of Interest

The authors declare no conflict of interest

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Figures:

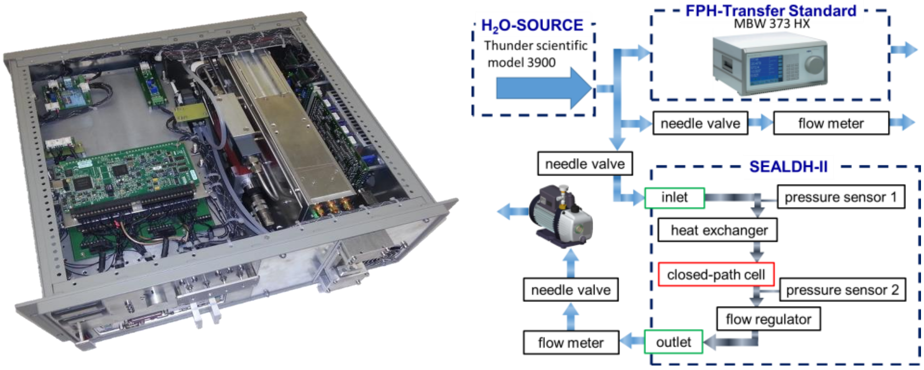

Figure 1: Left: Photo of SEALDH-II, the **S**elective **E**xtractive **A**irborne **L**aser **D**iode **H**ygrometer (dimension 19" 4 U). Right: Setup for the metrological absolute accuracy validation. The combination of a H_2O source together with a traceable dew point hygrometer, DPM, is used as a transfer standard – a traceable humidity generator (THG).

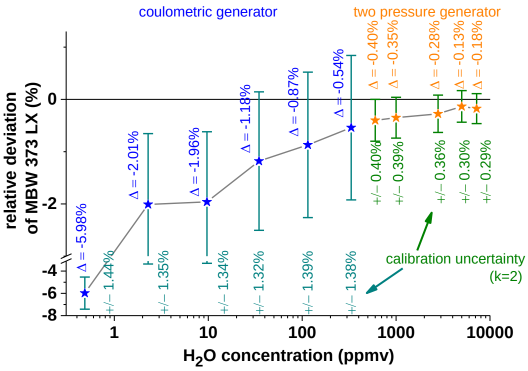

Figure 2: Calibration of the DPM (dew/frost point mirror hygrometer, MBW 373 LX, which is used as part of the THG) at the national primary water vapor standards of Germany. The standard for the higher H_2O concentration range (orange) is a "two pressure generator" (Buchholz et al., 2014); for the lower concentration range (blue) a "coulometric generator" (Mackrodt, 2012) is used as a reference. The deviations between reference and DPM are labelled with "Δ". The uncertainties of every individual calibration point are stated as green numbers below every single measurement point.

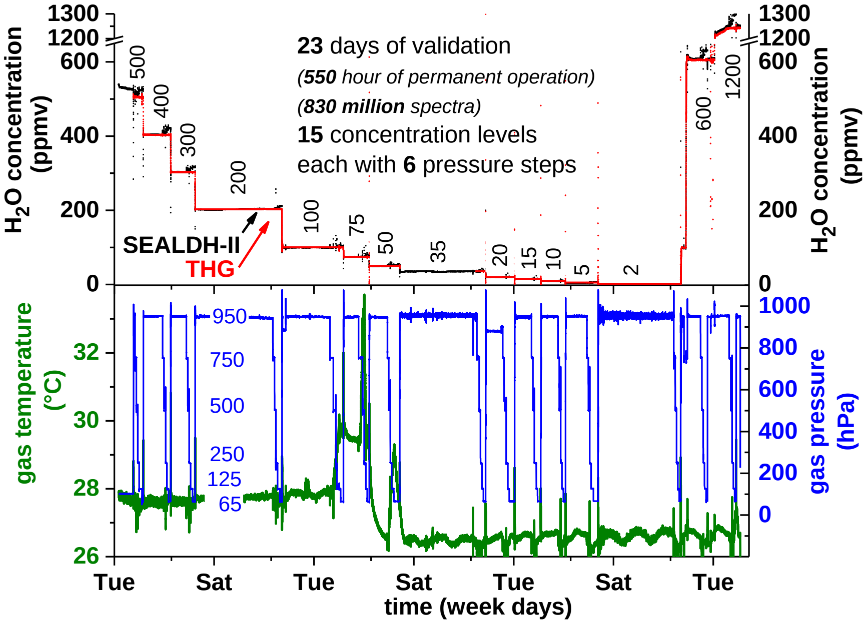

Figure 3: Overview showing all data recorded over 23 days of validation experiments. Measurements of the traceable humidity generator (THG) are shown in red, SEALDH-II data in black, gas pressure and gas temperature in SEALDH-II's measurement cell are shown in blue and green. Note: SEALDH-II operated the entire time without any malfunctions; the THG didn't save data in the 35 ppmv section; the temperature increase during the 75 ppmv section was caused by a defect of the air conditioning in the laboratory.

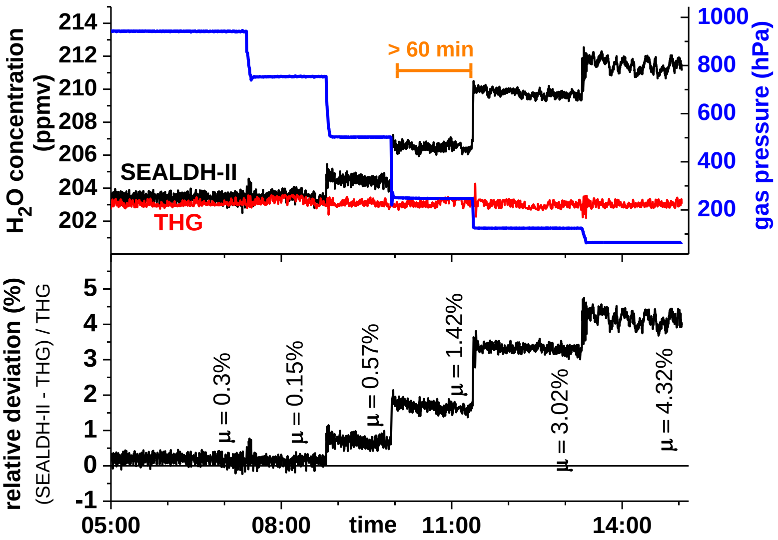

Figure 4: Detailed plot of the validation at 200 ppmv with six gas pressure steps from 50 to 950 hPa. Each individual pressure level was maintained for at least 60 minutes in order to avoid any dynamic or hysteresis effects and to facilitate clear accuracy assessments. The μ-values define the averaged relative deviation on every gas pressure level.

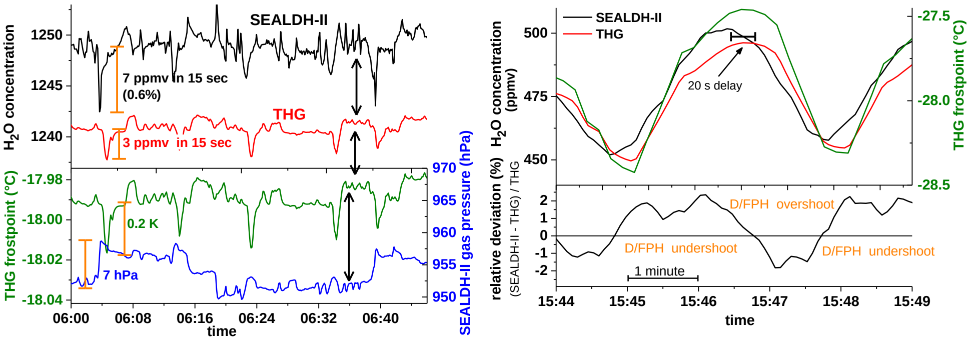

Figure 5: Short term H$_2$O fluctuations in the generated water vapor flow measured by SEALDH-II and the dew/frost point mirror hygrometer (D/FPH) of the traceable humidity generator (THG). The different dynamic characteristics of SEALDH-II (fast response time) and THG (quite slow response) lead in a direct comparison to artificial noise. Oscillating behaviors like in the right figure occur when the THG is not equilibrated. We did not use such data segments for the accuracy assessments.

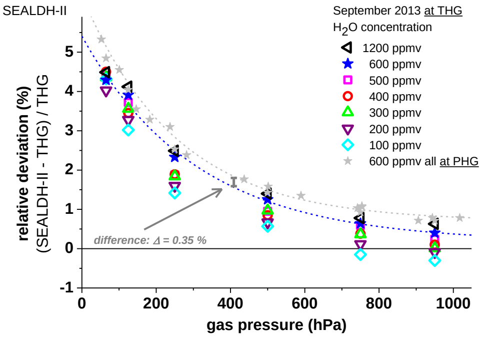

Figure 6: Gas pressure dependent comparison between SEALDH-II and THG over a H_2O concentration range from 600 to 1200 ppmv and a pressure range from 50 to 950 hPa. The 600 ppmv values (in grey) are measured directly at the national primary humidity generator (PHG) of Germany; all other H_2O concentration values are measured at and compared to the traceable humidity generator (THG). All SEALDH-II spectra were evaluated with a calibration-free first principles evaluation based on absolute spectral parameters. No initial or repetitive calibration of SEALDH-II with respect to any "water reference" source was used.

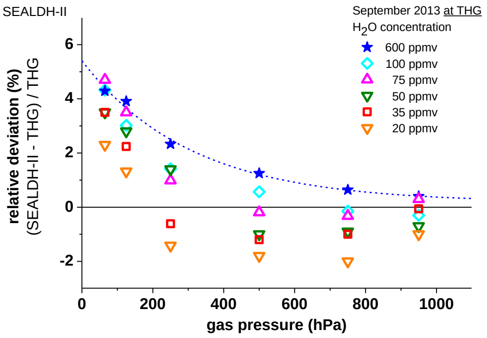

Figure 7: Comparison results as in Figure 6 but for the 200 – 600 ppmv range.

Gelöscht: Figure 6

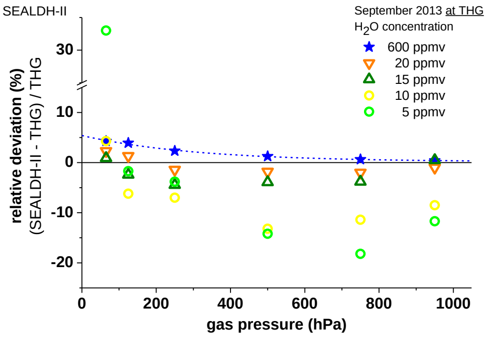

Figure 8: Comparison results as in Figure 6 and Figure 7 but for the 5 – 20 ppmv range. All spectra are determined with a calibration-free first principles evaluation concept. The major contribution to the higher fluctuations at lower concentrations is the accuracy of the offset determination (details see text).

Gelöscht: Figure 6

Gelöscht: Figure 7

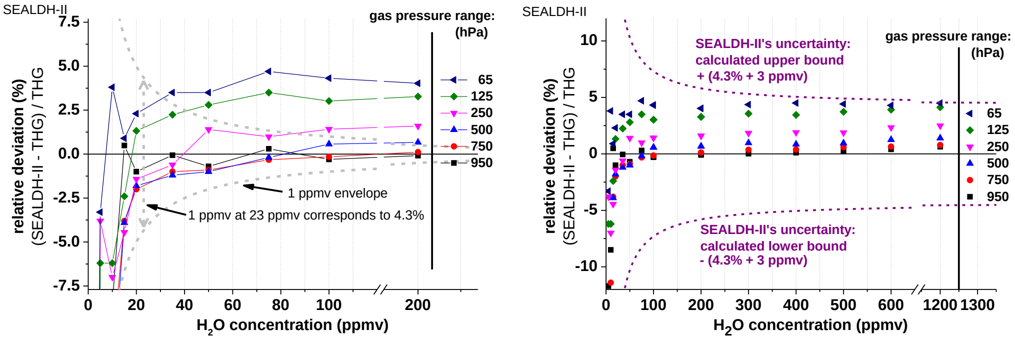

Figure 9: Direct comparison of SEALDH-II versus THG for H$_2$O concentrations between 5 and 200 ppmv and gas pressures from 65 to 950 hPa. Both figures show the relative deviations between SEALDH-II and THG grouped and color-coded by gas-pressure. Left plot: relative deviations of SEALDH-II versus THG below 200 ppmv; the grey line indicates the computed relative effect in SEALDH-II's performance caused by ±1 ppmv offset fluctuation. This line facilitates a visual comparison between an offset impact and the 4.3% linear part of the uncertainty of SEALDH-II. Right plot: relative deviations for all measured data in the same concentration range. Also shown is SEALDH-II's total uncertainty of 4.3% ±3 ppmv (calculated for 1013 hPa) as a dashed line.