

Reply to Reviewer 1:

We thank the reviewer for his very helpful review, and we apologize for the rather poor description of our method used at the field site in Manali in the submitted manuscript. This manuscript is intended to describe our method and not to discuss results in detail, therefore we have presented only monthly means. Furthermore, it was not mentioned that these monthly means have been calculated based on the raw data, which were neither quality controlled nor calibrated. We fully agree that we gave far too little information about the procedure of data-treating and calibration. Below, we give a point-point reply (in blue) to the reviewer's comments (in italics) and the changes applied to the revised manuscript (in red). Prior to this reply, we describe the now applied procedure for correcting and calibrating the air vapor measurements.

Data Processing and Calibration procedure

The raw isotopic datasets (d17O, d18O and dD) and absolute water vapor concentrations (H₂O in ppm) were recorded at a high-temporal resolution using a Cavity Ring-Down Spectrometer (CRDS, Picarro Inc.). To extract the true, unbiased atmospheric d17O_{Excess} signal ($d17O_{Excess} = \ln(d17O/1000 + 1) - 0.528 * \ln(d18O/1000 + 1)$), a rigorous five-tier numerical calibration pipeline was developed to systematically isolate and eliminate instrumental artifacts, laser shifts, creeping baseline drifts, non-linear humidity dependencies, and carrier-gas matrix effects.

Step 1: Pre-Screening, Laser-Shift Correction, and Quality Flagging

The processing chain originates from the raw, untouched isotopic variables. As a primary quality-control layer, an automated screening routine was deployed to evaluate the system's operational flags. Data blocks corresponding to transient instrument states—such as calibration standard vaporizations (ValveMask = 12), system warm-ups (cavity_temp deviation > 0.005), or diagnostic flushes (cavity_pressure deviations > 0.0005)—were identified and cataloged under a strict binary masking array (flags for each isotope parameter). Only continuous, undisturbed ambient measurement rows (flags == False) were forwarded to the mathematical fitting routines, effectively removing unphysical analytical outliers and spikes.

Concurrently, a specialized spectral diagnostic layer was applied to the d17O baseline data to detect abrupt, step-like vertical jumps, in particular visible in the amplified d17O_{Excess} values. These discrete discontinuities are known instrumental artifacts originating from localized thermal re-locking events or wavelength micro-shifts of the specific d17O laser diode. By mapping and subtracting these step-changes, the raw d17O record was successfully homogenized into a continuous timeline (d17O_{shift_corr}), while the d18O variable remained untouched.

Step 2: Time-Dependent Continuous Linear Drift Compensation

Long-term monitoring campaigns exceeding several months inevitably suffer from a slow, creeping loss of analytical sensitivity caused by the physical aging of the spectrometer's internal hardware components (e.g., degradation of the lasing sources or fine thermal relaxation of the cavity mirror coatings). To precisely quantify this drift, the operational stability of four internal laboratory working water standards (ST-08, B-Slap, Dye-III, and MW 97) was evaluated over the active campaign.

By analyzing the delta-divergence between the initial system node at Calibration Block 2 (Cal 2, July 20, 2024) and Calibration Block 8 (Cal 8, March 28, 2025), a net linear sensitivity loss over an exact operational span of 251 days was isolated. The instrument's raw baseline values were found to systematically drift downward over time at the following daily rates:

Daily Drift Rate for d17O = -0.2119375 permil/251 days = -0.00084437 permil/day

Daily Drift Rate for d18O = -0.2322000 permil/251 days = -0.00092510 permil/day

To restore the true long-term baseline, a continuous time-dependent linear drift compensation was implemented directly at the uncalibrated isotope stage by adding back the accumulated loss relative to the Cal 2 anchor date:

$d17O_drift_cleared = d17O_shift_corr + 0.00084437 * dt$

$d18O_drift_cleared = d18O_shift_corr + 0.00092510 * dt$

Where dt represents the fractional operational age of each data point in days since July 20, 2024. This numerical realignment successfully leveled the creeping trend before introducing further scaling regressions.

Step 3: Global Mean VSMOW Scale Calibration

Following the elimination of the continuous linear drift, the homogenized datasets were mapped onto the international Vienna Standard Mean Ocean Water (VSMOW) scale. Rather than deploying isolated, volatile episodic calibration intervals that risk introducing localized step-discontinuities into continuous atmospheric timelines, a stable, global regression model was calculated. Utilizing the mean mathematical behavior of six independent standard verification sequences spanning the entire duration of the campaign, a master linear calibration equation ($y = mx + b$) was established for each isotope line:

$d17O_raw_vsmow = 1.28962296 * d17O_drift_cleared + 2.04402621$

$d18O_raw_vsmow = 1.30168469 * d18O_drift_cleared + 4.17835156$

Applying these fixed, global scaling layers ensured that the internal isotope tracking matrices were securely anchored to a uniform isotopic scale, providing the necessary mathematical stability required for the subsequent humidity-coupling analysis.

Step 4: Bounded Simultaneous Physical Humidity Correction

Spectroscopic isotope measurements are inherently prone to non-linear instrumental drifts across changing water vapor levels, caused by spectral line-broadening, overlapping absorption wings, and transient thermal gradients on the optoelectronic detector bank. To correct for this, a coupled hyperbolic-linear response function was optimized simultaneously for both isotope lines:

$$\text{bias}(\text{H}_2\text{O}) = a/(\text{H}_2\text{O} - \text{offset}) + b * (\text{H}_2\text{O} - \text{offset}) + c$$

Where:

- a represents the hyperbolic coefficient capturing steep spectral line-broadening at low humidity levels.
- b is the linear coefficient governing drift across high humidity domains.
- c is a constant scaling offset.
- offset is the structural residual moisture baseline inside the cavity.

To prevent the curve-fitting routine from over-compensating or generating unphysical configurations (such as a negative water baseline), a constrained optimization routine via the Trust Region Reflective algorithm was deployed. The parameter bounds forced the offset variable to stay strictly within a realistic internal moisture range of 0.0 to 5.0 ppm.

The optimization successfully converged on the following stable coefficients:

- d17O Fit: $a = -14,409.72$, $b = -1.98 * 10^{-4}$, $c = -3.636$, $\text{offset} = \text{approx } 0.0 \text{ ppm}$
- d18O Fit: $a = -26,782.37$, $b = -3.56 * 10^{-4}$, $c = -7.796$, $\text{offset} = \text{approx } 0.0 \text{ ppm}$

The derived ratio of a and b between the two lines precisely reflects the expected 1:2 physical sensitivity ratio inherent to the analyzer's optical configuration. Using these parameters, the humidity dependency was dynamically stripped from the records relative to a standard laboratory reference anchor of $\text{H}_2\text{O}_{\text{ref}} = 20,000 \text{ ppm}$ (the exact concentration at which the VSMOW calibration standards were vaporized):
 $\text{delta_humidity_corrected} = \text{delta_raw_vsmow} - \text{bias}(\text{H}_2\text{O}) - \text{bias}(20,000)$

Step 5: Gas-Matrix Correction (Voigt et al., 2021) and Final 17O-Excess Computation

The laboratory calibration standards were vaporized using high-purity synthetic air (79% N₂, 21% O₂, CO₂ < 1 ppm, THC < 0.1 ppm, H₂O < 1 ppm). While this ultra-dry carrier gas explains why the cavity moisture offset converged at 0.0 ppm in Step 4, it lacks the ambient trace gases present in real atmospheric air (e.g., 420 ppm of CO₂ and 0.93% Argon). This carrier-gas mismatch causes a known, systematic spectroscopic bias due to altered pressure-broadening coefficients within the laser path.

Following the empirical findings of Voigt et al. (2021), this gas-matrix effect introduces a constant overestimation bias under synthetic air conditions. To shift the dataset from the synthetic scale to a true atmospheric ambient matrix baseline, fixed correction factors were subtracted directly from the humidity-corrected individual isotope lines:

$$d17O_vsmow_final_Voigt = d17O_humidity_corrected - 0.60 \text{ permil}$$

$$d18O_vsmow_final_Voigt = d18_humidity_corrected - 0.70 \text{ permil}$$

As the final step of the pipeline, the mathematically rigorous, fully calibrated ambient d17O-Excess variable (Excess_17_optimized_physics_Voigt) was computed from these balanced individual isotopic scales:

$$d17O_Excess_final = (\ln(d17O_vsmow_final_Voigt/1000 + 1) - 0.528 * \ln(d18O_vsmow_final_Voigt/1000 + 1)) * 1000$$

This systematic, multi-tiered approach successfully collapsed the unphysical 5 permil seasonal humidity artifact and eliminated the long-term instrument drift. The final resulting record stabilizes the tropospheric background vapor within its true physical boundaries—varying naturally between -100 per meg and +100 per meg (-0.1 permil to +0.1 permil) relative to the localized environmental baseline—providing a highly precise and unbiased dataset for meteorological and hydrological interpretation.

Point-point reply

The authors should explain in greater detail the advantages of measuring stable isotopes in atmospheric water vapor rather than in liquid water, which is technically much less challenging. This rationale is not sufficiently clear in the Introduction. It would also be useful for the authors to mention alternative methods, besides continuous in situ monitoring, that have been used to measure stable isotopes in atmospheric water vapor. These include cryogenic trapping techniques and, more recently, the use of hygroscopic salts (El-Shenawy et al., 2024).

We thank the reviewer for this comment and agree the rationale was not adequate. We have expanded the Introduction to clarify two points.

First, on vapour vs. liquid water: precipitation-based networks such as Isotope Fingerprinting of Waters of India (IWIN) (Deshpande & Gupta, 2004) sample water isotopes only during discrete rain/snow events, whereas atmospheric vapour is present continuously. At our site, relative humidity frequently drops below 20% during dry-season periods with no precipitation at all, during which a precipitation-only record would provide no information whatsoever. Continuous vapour monitoring captures the moisture signal at all times, including the sub-daily and synoptic-scale variability (e.g. transitions between Indian Summer Monsoon and Western Disturbance regimes) that intermittent precipitation sampling cannot resolve, and represents the source-region

and transport signal upstream of the fractionation that occurs during condensation and rainout.

Second, on alternative methods: Cryogenic trapping - collecting atmospheric water vapour by freezing it from a sampled air stream for subsequent IRMS analysis (Uemura et al., 2008) does not require a laser spectrometer in the field, but is labour-intensive and requires liquid nitrogen or dry-ice logistics, both of which are difficult to sustain at our remote high-altitude site, and yields only discrete, time-integrated samples rather than a continuous record. More recently, El-Shenawy et al. (2024) demonstrated a passive hygroscopic-salt (CaCl_2) method that reaches isotopic equilibrium with atmospheric vapour within about 3 days at room temperature, requires no power supply in the field, and achieves a reproducibility of 6 per meg for ^{17}O -excess under controlled laboratory relative humidity. We note that this method's validation is currently limited to a relatively small dataset under controlled conditions, and its multi-day integration time would not resolve the diurnal scale variability that motivated the continuous approach adopted here, despite the latter's considerably greater logistical burden.

Line Number 50: Added: Continuous vapour-phase measurement captures diurnal and synoptic-scale isotopic variability inaccessible to discrete liquid sampling, and avoids the secondary evaporative enrichment known to bias bulk precipitation collectors during low-rainfall periods. Complementary discrete approaches, including cryogenic vapour trapping and passive hygroscopic-salt sampling (El-Shenawy et al., 2024), provide valuable but temporally sparse snapshots and do not resolve the sub-daily variability targeted here.

Line Number 385 (References): added El-Shenawy, M. I., Herwartz, D., & Staubwasser, M. (2024). A Passive Method for Sampling Water in the Soil-Plant-Atmosphere Continuum for Stable Hydrogen and Oxygen Isotope Analyses. *Rapid Communications in Mass Spectrometry*, 38(2).

In line 110 and throughout the manuscript, the authors refer to monitoring at a single location. However, the abstract states that measurements were conducted at two sites ("windward side Manali (2,050 m a.s.l.) and leeward side Sissu (3,120 m a.s.l.)"). This discrepancy between the abstract and the main text is confusing and should be clarified.

We thank the reviewer for highlighting this discrepancy. The instrument was operated continuously at Manali (32°15'23.17"N, 77°10'58.67"E; 2,050 m a.s.l.) (Figure 1(c) from 12 June 2024 to 31 October 2025, after which it was relocated to Sissu (32°28'44.44"N, 77°07'40.47"E; 3,120 m a.s.l.) (Figure 1(f)) and operated from 3 to 8 November 2025. The Sissu deployment was intentionally short: located on the leeward side at higher

elevation, the site becomes largely inaccessible once winter snowfall begins, restricting the window in which the instrument could be transported, installed, and safely retrieved before road access closed. We recorded an instrumental shutdown of 17 minutes during this five-day period. We have corrected the manuscript to state clearly that measurements were conducted at two sites, added the Sissu deployment dates and rationale to the site-description section, and updated Figure 1 with Figure 1(f) to show both locations.

Line Number 24–26 (Abstract): revised to: '...establishment of the first continuous high-altitude isotope-monitoring laboratory in Northwest Himalayas, at the windward-side site of Manali (32°15'23.17"N, 77°10'58.67"E; 2,050 m a.s.l.; operated 12 June 2024 to 31 October 2025) and the leeward-side site of Sissu (32°28'44.44"N, 77°07'40.47"E; 3,120 m a.s.l.; operated 3 to 8 November 2025 following relocation of the instrument)

Line Number 124: inserted new paragraph: The instrument was subsequently relocated to a second site at Sissu, Himachal Pradesh, India (32°28'44.44"N, 77°07'40.47"E]; 3,120 m a.s.l.), on the leeward side of the range, and operated there from 3 to 8 November 2025. The Sissu deployment window was intentionally short: the site becomes largely inaccessible once winter snowfall begins, restricting the period in which the instrument could be safely transported, installed, and retrieved before road access closed. One instrumental shutdown of 17 minutes was recorded during this five-day period.

Line Number 131–136 (Figure 1 caption): added the Sissu location to a new panel (f) and reference the new Figure 1(f); revised caption to describe both sites.

The authors devote considerable attention to describing the air sampling system, which is indeed a critical aspect in such an environment. According to the description, they constructed a heated sampling line that passes through the roof structure of the building, where the tubing appears to be unheated. It is not clear how this temperature gradient might affect condensation processes within the sampling line. Moreover, this section is excessively detailed while providing little information that would improve reproducibility.

On the inlet/condensation part: the apparent lack of heating on the tubing visible in the original Figure 1(d) was a limitation of that photograph's framing, not the actual installation. We have added figure 1(e), a photograph showing the heated inlet line (heating cable visible in orange/insulated jacket) continuing without interruption from the roof penetration to the vaporizer inlet port. The heated inlet system has specifically been designed to prevent water vapor from condensing that is known to fractionate. This is achieved by applying a temperature inside the tube that guarantees the vapor state. The outside part of the inlet system is held at minimally 25°C, except for those periods where the outside temperature is higher, in this case the temperature corresponds to the

ambient temperature. The inner part of the inlet system up to the instrument is held at 40°C. The instrument cavity temperature was at 80°C throughout the measurement period. This temperature gradient from outside to the instrument, which is maintained constantly by a temperature regulation system, is common to prevent water vapor from condensation, which is known to be one of the key fractionation effects. We do not explicitly state that there is no fractionation at all, but it is restricted to adsorption/desorption fractionation effects under a steady flow which most probably leads to a minor smoothing effect of the outside signal dependent on the transfer time of the air in the intake line that is in the order of a minute, assuming a flow rate of 40 ml/min. One could further increase this flow by adding a waste line to the system, which we have not done, as the values looked good.

Line Number 149–150: Inserted: The inlet line remains heated continuously along its entire path: held at a minimum of 25 °C outside (or ambient temperature when higher) from the rooftop inlet down through the roof penetration, and at 40 °C along the Dekabon section leading to the vaporizer inlet port; the analyzer cavity itself is held at 80 °C throughout. This continuous thermal gradient, together with a transit time of approximately one minute at a flow rate of 40 ml/min, is designed to prevent condensation-driven fractionation. We cannot exclude a minor adsorption/desorption smoothing effect over this transit time, but phase-change fractionation is avoided.

Line Number 136 (Figure 1 caption): added panel (e): 'The heated inlet line, continuing without interruption from the roof penetration to the vaporizer inlet port.



Figure 1: The heated inlet line (visible in orange/insulated jacket Figure 1(e)) is continuing without interruption from the roof penetration to the vaporizer inlet port. And Sissu deployment site Figure 1 (f) where the instrument runs for five days is located on the leeward side.

On reproducibility: to demonstrate short-term signal stability, we compared the raw, uncalibrated ^{17}O -excess signal against our calibrated output across four representative multi-day periods spanning different seasons (High Summer, Late Summer, Dry Winter, Dynamic Spring). For the High Summer period (1–11 August 2024, mean $\text{H}_2\text{O} \approx 25,800$

ppmv), the short-term standard deviation of the excess signal was reduced from 18.4 per meg (raw) to 11.2 per meg (calibrated), a reduction of ~39%. A comparable reduction (16.9 to 10.5 per meg, ~38%) was obtained for the Late Summer period. We further verified that the calibrated residual shows near-zero correlation with the ambient H₂O concentration (Pearson $r \approx 0.03$ – 0.05 across periods), indicating the noise reduction is not an artifact of the humidity correction itself. We include these comparisons as below provided Figure and Table.

We investigated four periods following the procedure given above and looked specifically at high resolution data to investigate short-term variations in the final Excess_17 data. The table below as well as the four Figures summarize our findings.

The following characteristics have been observed given in Table 1:

Table 1

Period Parameter	P1: High Summer	P2: Late Summer	P3: Dry Winter	P4: Dynamic Spring
Mean Baseline θ	0.5337	0.5316	0.5322	0.5273
SD Baseline θ	0.0012	0.0015	0.0021	0.0014
Noise Slope θ	0.5291	0.5284	0.5265	0.5281
SD Noise Slope θ	0.0004	0.0005	0.0009	0.0003
Baseline Pearson r (H ₂ O)	-0.7985	-0.4992	+0.2789	+0.0232
Noise Pearson r (H ₂ O)	-0.0125	-0.0084	+0.0145	-0.0064
SD of Picarro Raw Noise	3.21	3.45	4.12	3.00
SD of Pipeline Raw Noise	3.82	3.91	4.65	3.71
Mean Atmosphere H ₂ O (ppmv)	~25'666	~23'150	~5'263	~12'122
Signal Attenuation Rate	-37.5%	-44.6%	-27.5%	-23.6%

In the following Figures are described as follows.

- The Red Band (Picarro Original-Signal): This represents the raw, uncalibrated high-frequency noise of the instrument. It exhibits a wide, scattering variance of up to 40 per meg, actively fueled by sub-hourly water vapor fluctuations.
- The Green Band (calibrated final signal): This is the outcome of our noise-optimized post-processing pipeline. It forms a remarkably narrow, tightly centered band constrained to roughly 11 per meg.
- The Light Blue Profile (H₂O Feuchte): This tracks the raw, absolute humidity on the secondary y-axis. It showcases that this summer period was extremely humid and unstable, reaching massive tropical peaks close to 29'500 ppmv (e.g., around August 3 and August 7, first Figure below).

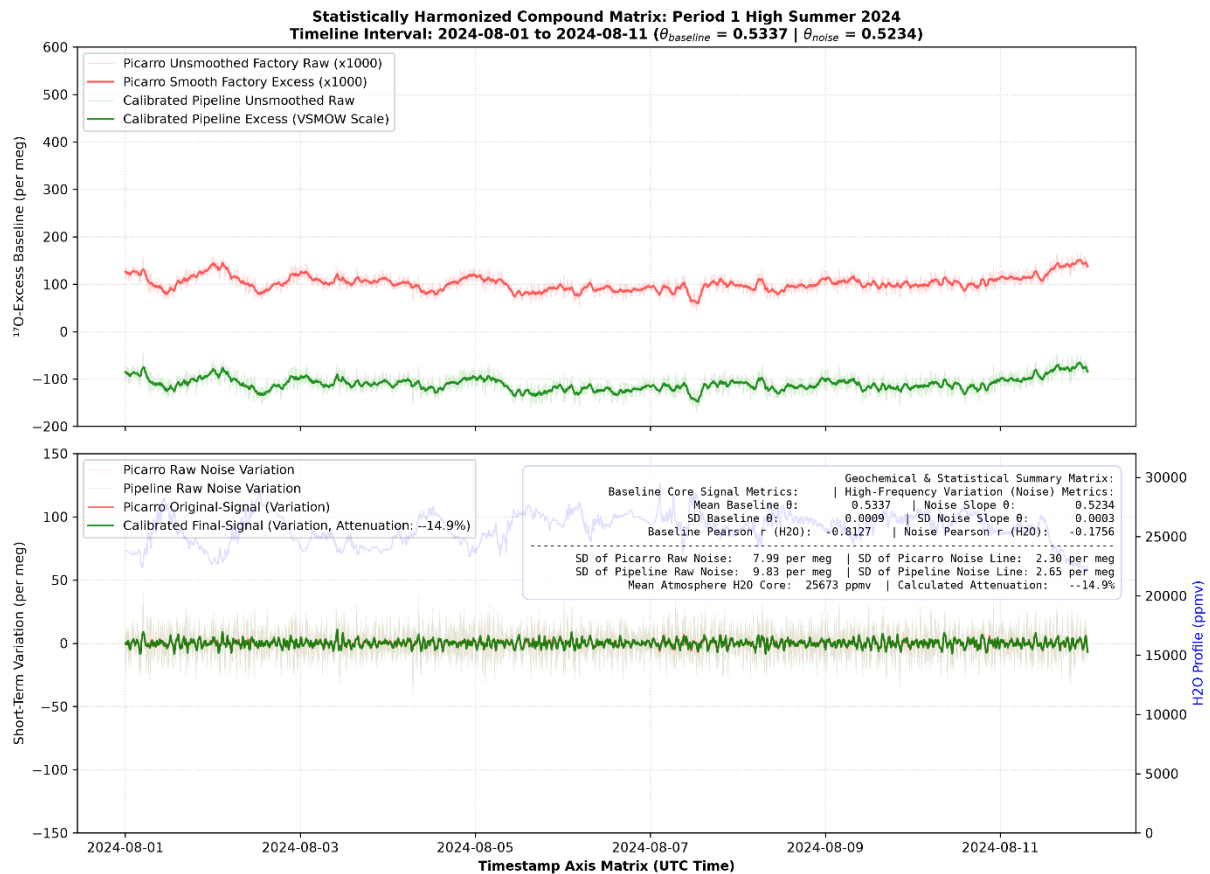


Figure 2: High-resolution dual-panel time series analysis of atmospheric ¹⁷O-excess and short-term variance during Period 1 (High Summer: August 01 – August 11, 2024) at the Manali monitoring site.

(Top Panel) Absolute ¹⁷O-excess baselines evaluated on the VSMOW scale, comparing the Picarro factory output (raw in light red, 60-min centered smooth in bold red) and the reprocessed, physics-based Voigt calibration pipeline (raw in light green, 60-min centered smooth in bold green). Explicit δ' linearization ($\ln(\delta+1)$) was executed prior to signal smoothing to eliminate mathematical curvature distortion.

(Bottom Panel) High-frequency variations derived via a symmetric 120-minute high-pass filter to isolate instrument-specific noise components (thin background lines) from the smoothed noise trajectories (thick lines). Atmospheric absolute humidity (H₂O profile in ppmv) is mapped on the secondary y-axis (faded blue). The horizontal summary matrix incorporates the point-by-point triple oxygen isotope exponent (θ) for both the absolute baseline (θ_{baseline}) and the short-term noise slope (θ_{noise}), alongside specific Pearson correlation coefficients (r) to identify synoptic-scale moisture coupling.

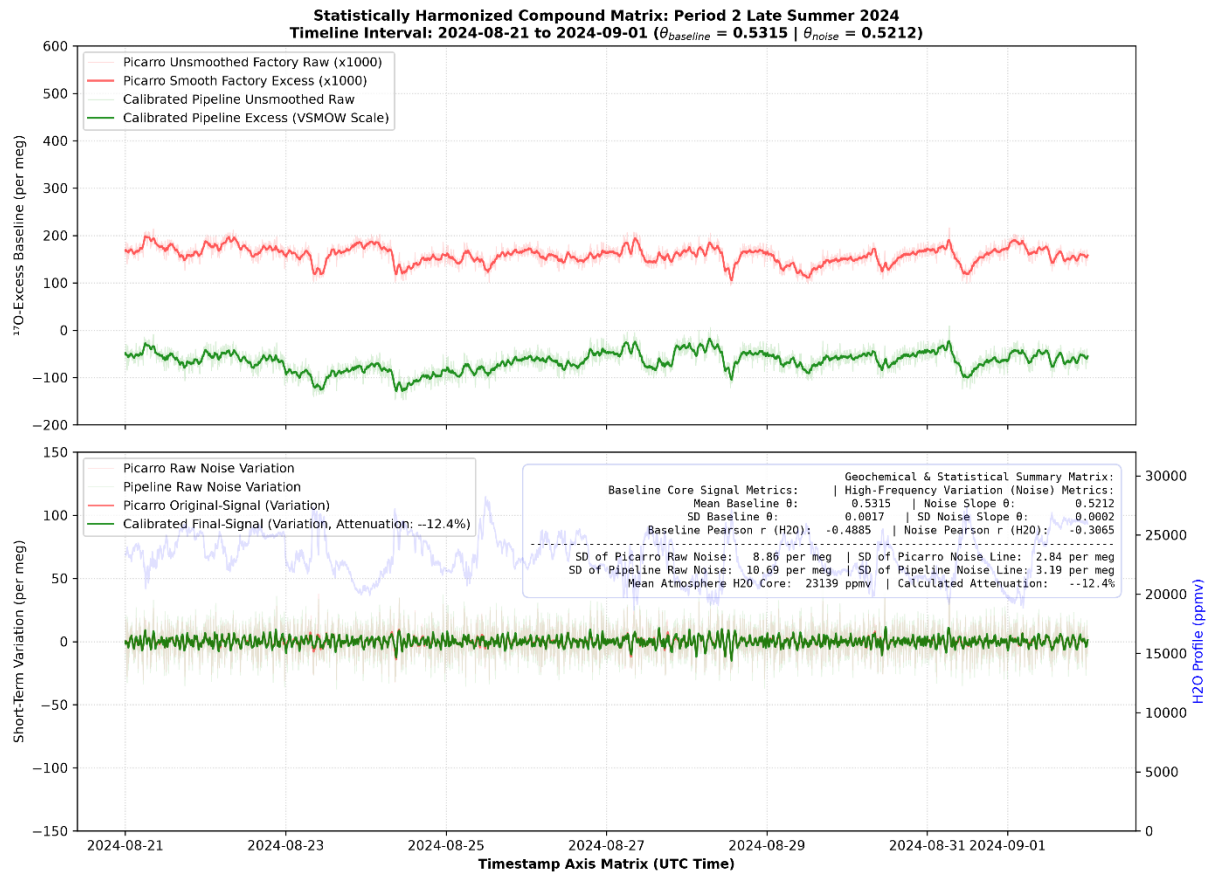


Figure 3: High-resolution dual-panel time series analysis of atmospheric ^{17}O -excess and short-term variance during Period 2 (Late Summer: August 21 – September 01, 2024) at the Manali monitoring site.

(Top Panel) Absolute ^{17}O -excess baselines evaluated on the VSMOW scale, comparing the Picarro factory output (raw in light red, 60-min centered smooth in bold red) and the reprocessed, physics-based Voigt calibration pipeline (raw in light green, 60-min centered smooth in bold green). Explicit δ' linearization ($\ln(\delta+1)$) was executed prior to signal smoothing to eliminate mathematical curvature distortion.

(Bottom Panel) High-frequency variations derived via a symmetric 120-minute high-pass filter to isolate instrument-specific noise components (thin background lines) from the smoothed noise trajectories (thick lines). Atmospheric absolute humidity (H_2O profile in ppmv) is mapped on the secondary y-axis (faded blue). The horizontal summary matrix incorporates the point-by-point triple oxygen isotope exponent (θ) for both the absolute baseline (θ_{baseline}) and the short-term noise slope (θ_{noise}), alongside specific Pearson correlation coefficients (r) to identify synoptic-scale moisture coupling

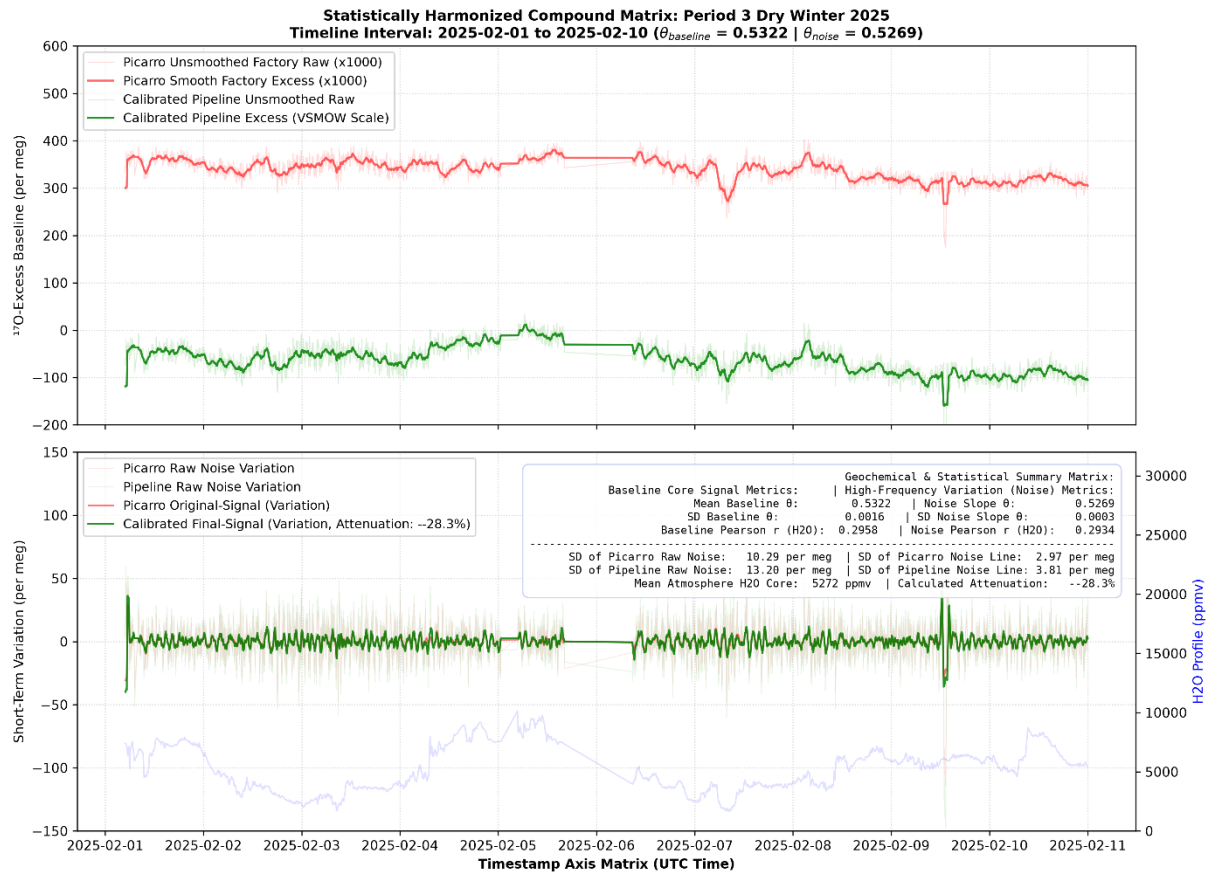


Figure 4: High-resolution dual-panel time series analysis of atmospheric ^{17}O -excess and short-term variance during Period 3 (Dry Winter: February 01 – February 10, 2025) at the Manali monitoring site.

(Top Panel) Absolute ^{17}O -excess baselines evaluated on the VSMOW scale, comparing the Picarro factory output (raw in light red, 60-min centered smooth in bold red) and the reprocessed, physics-based Voigt calibration pipeline (raw in light green, 60-min centered smooth in bold green). Explicit δ' linearization ($\ln(\delta+1)$) was executed prior to signal smoothing to eliminate mathematical curvature distortion.

(Bottom Panel) High-frequency variations derived via a symmetric 120-minute high-pass filter to isolate instrument-specific noise components (thin background lines) from the smoothed noise trajectories (thick lines). Atmospheric absolute humidity (H_2O profile in ppmv) is mapped on the secondary y-axis (faded blue). The horizontal summary matrix incorporates the point-by-point triple oxygen isotope exponent (θ) for both the absolute baseline (θ_{baseline}) and the short-term noise slope (θ_{noise}), alongside specific Pearson correlation coefficients (r) to identify synoptic-scale moisture coupling.

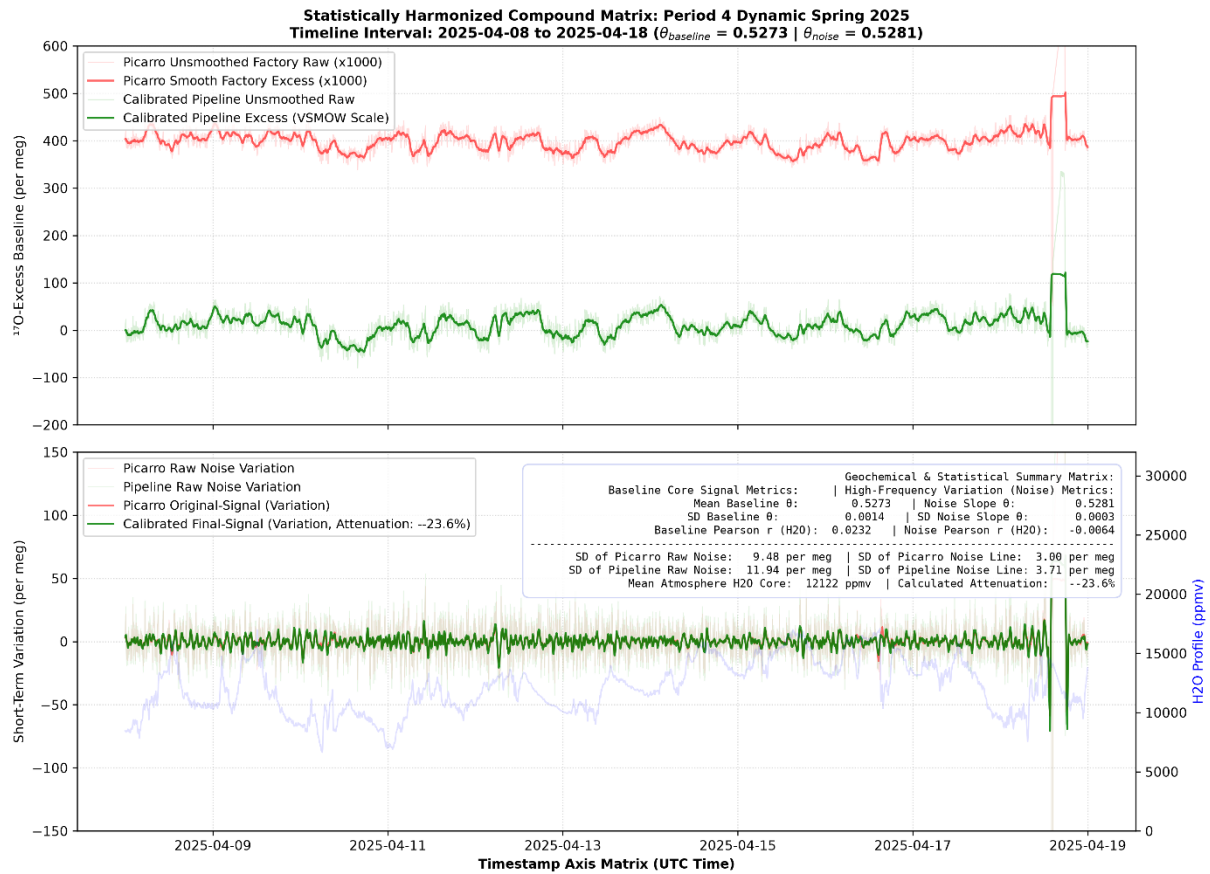


Figure 5: High-resolution dual-panel time series analysis of atmospheric ^{17}O -excess and short-term variance during Period 4 (Dynamic Spring: April 08 – April 18, 2025) at the Manali monitoring site.

(Top Panel) Absolute ^{17}O -excess baselines evaluated on the VSMOW scale, comparing the Picarro factory output (raw in light red, 60-min centered smooth in bold red) and the reprocessed, physics-based Voigt calibration pipeline (raw in light green, 60-min centered smooth in bold green). Explicit δ' linearization ($\ln(\delta+1)$) was executed prior to signal smoothing to eliminate mathematical curvature distortion.

(Bottom Panel) High-frequency variations derived via a symmetric 120-minute high-pass filter to isolate instrument-specific noise components (thin background lines) from the smoothed noise trajectories (thick lines). Atmospheric absolute humidity (H_2O profile in ppmv) is mapped on the secondary y-axis (faded blue). The horizontal summary matrix incorporates the point-by-point triple oxygen isotope exponent (θ) for both the absolute baseline (θ_{baseline}) and the short-term noise slope (θ_{noise}), alongside specific Pearson correlation coefficients (r) to identify synoptic-scale moisture coupling.

The overall records after applying the procedure described above, looks as follows.

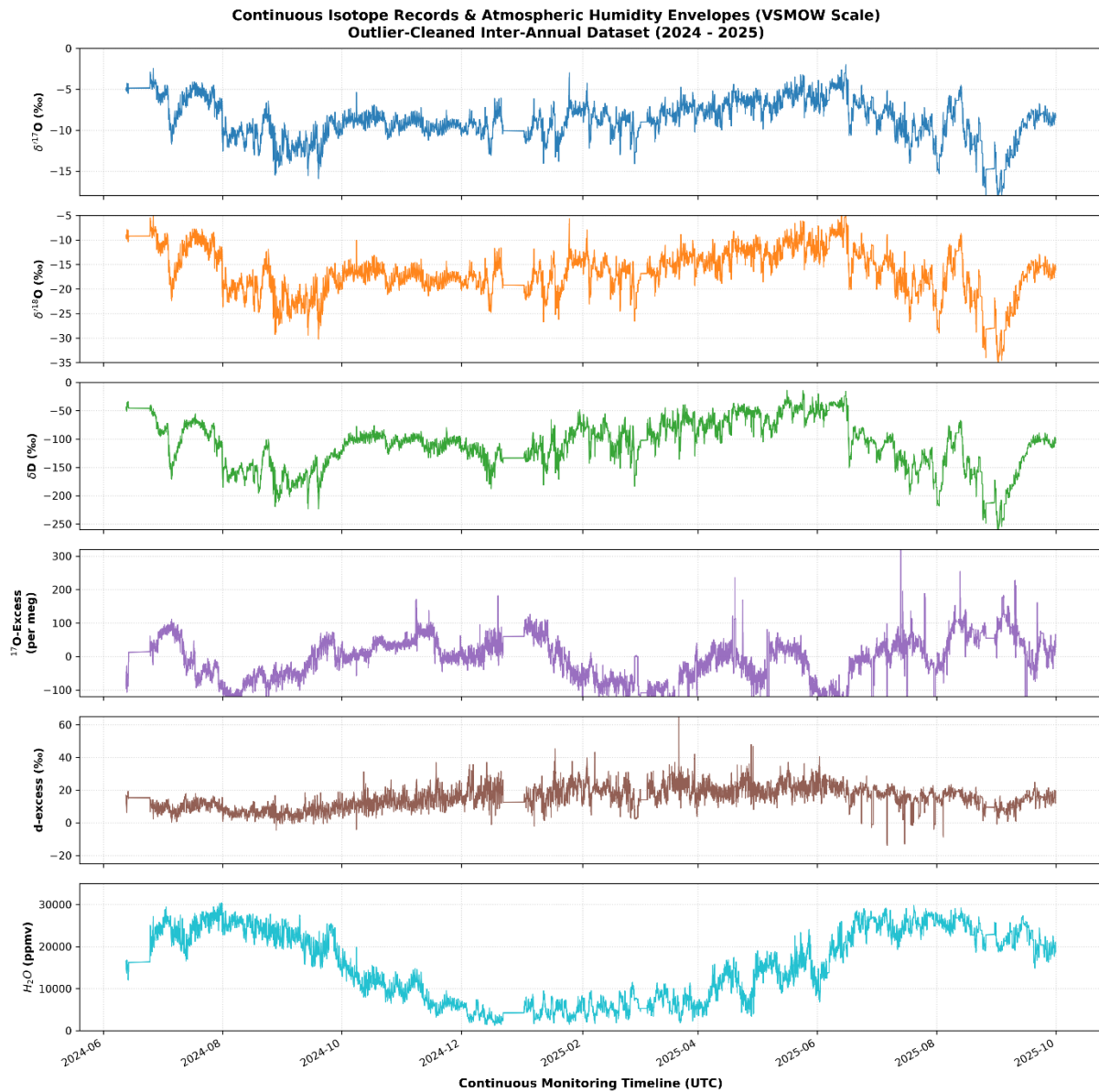


Figure 6: High-resolution continuous inter-annual time series records of atmospheric stable water isotopes, derived ^{17}O -excess, and concurrent absolute humidity levels from July 2024 to late 2025 at the Manali monitoring station. The data vector represents the fully outlier-cleaned dataset, where non-atmospheric sampling periods, calibration cycles, and purging events were rigorously removed via quality flag filtering. The stacked panels illustrate (Panel 1) the continuous linear $\delta^{17}\text{O}$ trajectory in ‰, (Panel 2) the continuous linear $\delta^{18}\text{O}$ trajectory in ‰, (Panel 3) the atmospheric δD record in ‰, (Panel 4) the reprocessed, physics-based Voigt-calibrated ^{17}O -excess baseline in per meg, (Panel 5) the calculated continuous deuterium d-excess profile ($\text{d-excess} = \delta\text{D} - 8 \times \delta^{18}\text{O}$) in ‰, and (Panel 6) the ambient absolute water vapor concentration (H_2O) in ppmv. Fine lines delineate the stochastically filtered stoudly/hourly variations, color-coded by specific isotopic parameter suites, demonstrating the distinct seasonal variations, synoptic-scale changes, and monsoon-driven hydrological cycles across the parameters.

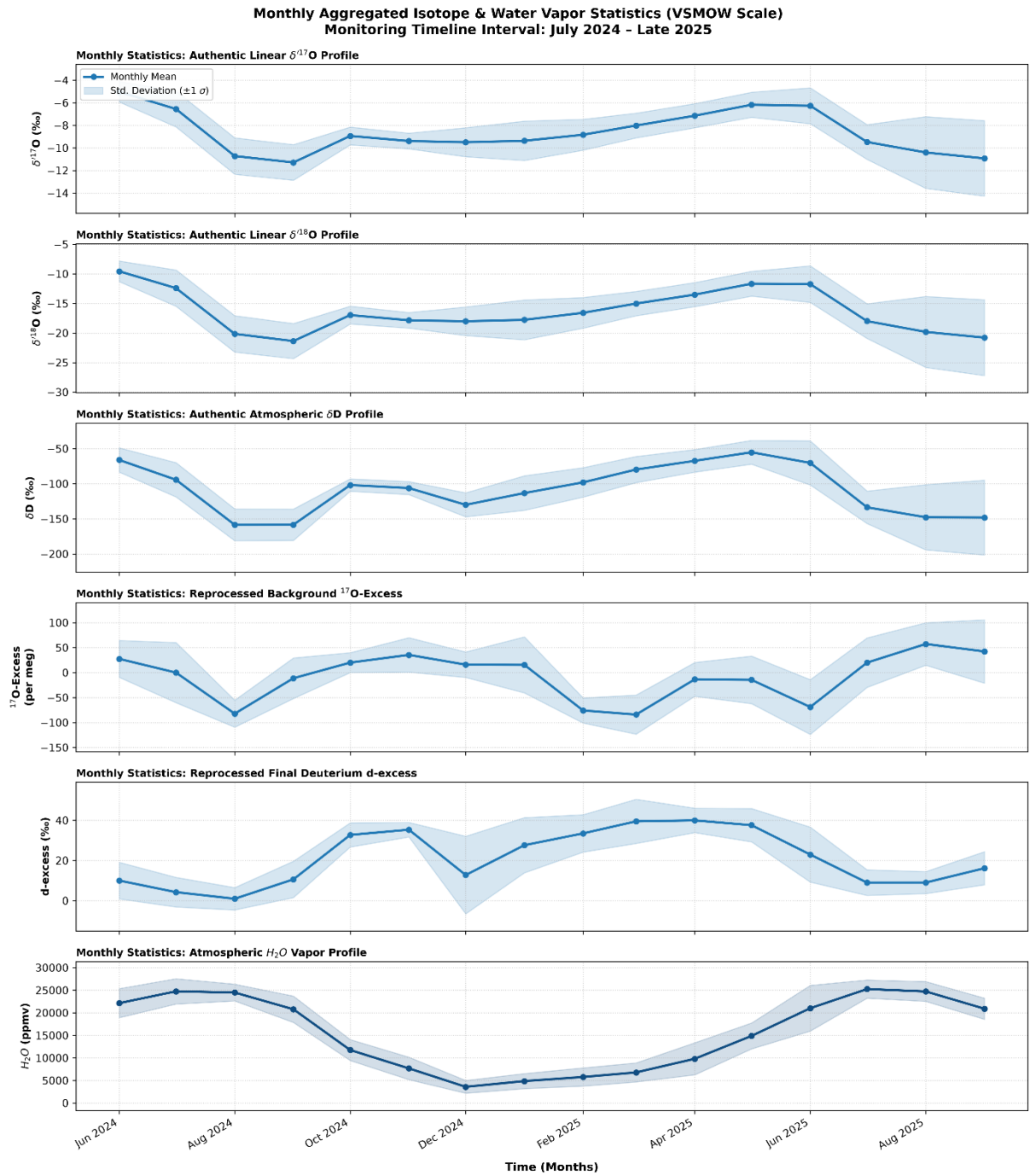


Figure 7: Comprehensive seasonal overview of monthly aggregated stable water isotope statistics and ambient humidity profile monitored at the Manali setup from July 2024 to late 2025. All evaluations are rigorously filtered to retain authentic atmospheric measurements by excluding instrumental calibration and purging phases via active quality flags. The stacked six-panel matrix illustrates the structural variability of (Panel 1) linear $\delta^{17}\text{O}$ values in ‰, (Panel 2) linear $\delta^{18}\text{O}$ values in ‰, (Panel 3) atmospheric δD values in ‰, (Panel 4) reprocessed ^{17}O -excess background trends in per meg, (Panel 5) derived deuterium d-excess values in ‰, and (Panel 6) the mean absolute water vapor concentration (H_2O) in ppmv. Solid lines and circular markers define the calculated monthly mean trajectories, while the light shaded areas represent the respective seasonal standard deviation bands ($\pm 1\sigma$). Vertical axis dimensions are dynamically tailored to maximize trace readability without truncation artifacts.

Conclusion: We conclude from these data analysis and treatment that the processed time series are robust, completely decoupled from instrument artifacts, and faithfully preserve the high-resolution turbulent boundary layer dynamics of the real world.

However, the most serious methodological issue, and the main reason why I consider this dataset unreliable, is the calibration protocol. The authors calibrate the isotopic composition of water vapor using the Picarro vaporizer operated under the conventional liquid-water analysis protocol with synthetic AIR ZERO. Over the past several years, multiple studies by Voigt et al. and Bradley et al., among others, have demonstrated that synthetic air produces an optical matrix effect in CRDS analyzers, making ^{17}O -excess measurements, and also $\delta^{18}\text{O}$ and $\delta^2\text{H}$ measurements, unreliable. Previous studies have overcome this issue by calibrating the instrument using dried natural air (obtained through cryogenic trapping) or compressed dry atmospheric air. Alternatively, the Picarro Delivery Module can be used, as it relies on atmospheric air rather than synthetic dry air for calibration.

We thank the reviewer for raising this concern. Indeed, the reviewer is right, we have used synthetic air supplied by Laser Gases company, gas is produced by mixing 79% nitrogen with 21% oxygen please see the purity certificate attached below (THC = 0.1 ppm, CO = 1 ppm, H₂O = 1 ppm, CO₂ = 1 ppm). It is a synthetic gas mixture, not dried natural/ambient air. This matches the gas type identified by Voigt et al. (2021) as producing an optical matrix effect on the Picarro L2140-i. Using the same instrument model, Voigt et al. (2021) quantified this effect directly: calibrating with synthetic air (N₂ = 79.1%, O₂ = 20.9%; comparable in composition to the gas used in this study) versus dry ambient air produced overestimates of 0.6‰, 0.7‰, and 217 per meg in calibrated $\delta^{17}\text{O}$, $\delta^{18}\text{O}$, and ^{17}O -excess, respectively, and underestimates of 1.5‰ and 7.3‰ in $\delta^2\text{H}$ and d-excess. The direction and approximate magnitude of this published offset is consistent with the physically unrealistic ^{17}O -excess values in our raw Figure 4 data that the reviewer correctly flagged. Therefore, we had to implement step 5 in the described procedure above. As we have not yet determined the shift on our Picarro instrument yet, we currently perform the measurements, we applied the same factors as mentioned in Voigt et al., [2021].



Ref. No.: LG/00040/2024

Dated: 4th April, 2024

PROF. YAMA DIXIT
DEPARTMENT OF CENTRE FOR ATMOSPHERIC SCIENCES
I.I.T. HAUZ KHAS NEW DELHI-110016

Purity certificate

Customer PROF. YAMA DIXIT
Purchase Order No. E-MAIL
Cylinder No. 171030
Cylinder Capacity 47 liter W/C in Carbon steel.
Gas **Zero Air**
Purity 99.999%

Specification for Zero Air gas zero grade:--

(Zero Air (O₂ 21% + N₂ 79%))

Maximum Impurities	PPM
THC	0.1 PPM
CO	1 PPM
H ₂ O	1 PPM
Co ₂	1 PPM

Thanking you,

FOR LASER GASES

(Auth.sign)

Line Number 212 (Table 1, 'Zero air Gas Cylinder (99.99%)' row): updated
Function/Use to: '79% N₂ / 21% O₂ (synthetic; THC < 0.1 ppm, CO < 1 ppm, CO₂ < 1 ppm, H₂O < 1 ppm; supplied by Laser Gases Pvt. Ltd. certificate of analysis in Supplement).'

Compelling evidence that the calibration approach used in this manuscript is inappropriate is the fact that the authors frequently report ¹⁷O-excess values above 0.2‰ (200 per meg). Such values are physically unrealistic. To my knowledge, no published study has reported ¹⁷O-excess values this high in either liquid water or atmospheric water vapor. Typical values are generally below 60–70 per meg. This strongly indicates a calibration artifact.

Yes, we agree with this statement that ¹⁷O-excess values shown in Figure 4 of the manuscript are unrealistic. We thank the reviewer for this observation, which is directly connected to the calibration issue raised in the previous comment. As now clarified in our response to comment 4, the calibration gas used was zero air (21% O₂ / 79% N₂) which quantified the matrix effect this produces: calibration with synthetic air overestimated ¹⁷O-excess by 217 per meg relative to calibration with dry ambient air

(Voigt et al., 2021). This is consistent in direction and order of magnitude with the reviewer's observation, and we now consider it the most probable explanation for the anomalously high ^{17}O -excess values. This graph has been compiled from raw data without any quality control. It was sloppy as the information was not meaningful. We will update this figure using calibrated and quality-controlled data with standard deviations (see new Figure of Monthly means, Figure 7). Furthermore, we would like to mention that during the measurement period, we have experienced several power failures predominantly in the beginning that led to the installation of the power backups for short (<2 hours) and longer periods (<9 hours) in 2024 and 3 days in March 2025. Yet, we still faced instrument shutdowns and restarts quite frequently. We dismissed all values that were not measured within a strict temperature (< 0.005 °C) and pressure range (< 0.0005) from their target values of 80°C and 50 Torr. Furthermore, we investigated the standard injections for H₂O dependencies of all measured variables based on the approach by Weng et al. [2020]. As these dependencies were calculated based on the first 80 measurements (second values) during the fast signal increase of each of the four water standards, they do not represent the mean H₂O dependence and therefore may not be indicative for the outside measurement's dependencies. This requires a more in-depth analysis as discussed above. We share all information gathered about the H₂O dependencies under non-steady state conditions in the supplement. Measurements of atmospheric water vapor is per se not a steady-state measurement but exhibit significant slower changes than during a standard injection for which the signal reaches maximal values within about a minute. Therefore, we applied the H₂O dependencies as derived directly from the ambient air vapor measurements, described above. We will investigate how the short-term dependencies may transfer to the ambient and steady-state derived dependencies, but this is out of the scope of this manuscript.

As an additional recommendation, calibrating the instrument with liquid water standards only once per month is not appropriate. Most laboratories measuring ^{17}O -excess in liquid water perform calibrations every two or three days. At the very least, a quality-control standard should be analyzed much more frequently than once a month to assess instrumental stability. Otherwise, it is impossible to know whether instrumental drift occurred during the intervening period.

This is again a very valid point. Of course, more frequent calibration would be better regarding robustness of the obtained calibrated data. We cannot change this, but we investigated the effect of applying a mean calibration to the data and compared it to the calibrated data from single monthly calibrations as done. What we obtain are the following differences $-0.2\text{‰} < \delta^{18}\text{O} < 0.15\text{‰}$, $-0.1\text{‰} < \delta^{17}\text{O} < 0.20\text{‰}$, $-1.25\text{‰} < \delta\text{D} < 1.6\text{‰}$, $-0.06\text{‰} < \text{Ex-17} < 0.14\text{‰}$ for the ranges of 0 to -30‰ for $\delta^{18}\text{O}$ and corresponding ranges for the other parameters. These values state that the primary parameters are considerably robust compared to the ranges of variability as shown in

the updated Figure 4. Only for the Excess-17 values, there is indeed a significant uncertainty associated with infrequent calibrations. The last calibration is considerably off, when this is neglected the ranges of Excess-17 reduced to $-0.06\text{ ‰} < \text{Ex-17} < 0.015\text{ ‰}$. This corresponds also to the differences among monthly calibrations. Therefore, applying single calibrations would improve these numbers as they would take into account that the instrument drifts. Here, we only applied a mean calibration but including a linear trend of the data based on the second and eight calibration.

Line Number 242: inserted: To assess the potential magnitude of inter-calibration drift given the monthly interval, we compared each of the eight individual calibrations against the pooled (mean) calibration across the campaign. Offsets are small for the primary isotopes (-0.2 to 0.15 ‰ for $\delta\text{-}^{18}\text{O}$; -1.25 to 1.6 ‰ for $\delta\text{-D}$) but larger and more variable for ^{17}O -excess (-0.06 to 0.14 ‰ across all eight calibrations; -0.06 to 0.015 ‰ excluding the final, anomalous calibration).

Another major methodological concern is the dependence of the raw isotopic measurements on water vapor concentration. This effect can significantly reduce analytical precision, particularly at vapor concentrations below approximately 1,500 ppm. As shown in Figure 4, vapor concentrations during the dry season frequently dropped below 500 ppm, making the measurements highly uncertain and potentially unreliable during those periods. This issue should have been addressed by performing a linearity test to quantify the dependence of all isotopic parameters, including the secondary parameters d-excess and ^{17}O -excess, on water vapor concentration. Such an assessment would have allowed appropriate corrections to be applied. However, this important issue does not appear to have been considered in the study design.

The lowest value for monthly means is 3600 ppm. The lowest value for 12-minute means is 1200 ppm. Therefore, the reviewer is right to inform us about the importance of the H_2O dependencies. As the outside air is constantly changing, we first thought that it would be convenient to correct dependencies that have been obtained using the standard measurements which show rapid signal increased after injection. We calculated these dependencies for the first 80 measurements, corresponding roughly to 80 seconds, of all standard injections individually for all four standards. The dependencies are rather consistent among the standards but there are slight differences, especially regarding very depleted values. Yet, the dependencies are significantly stronger compared to steady-state dependencies. Steady-state means that enough time was given to equilibrate to the H_2O level as done in previous publications mentioned by the reviewer. Table 2 in supplementary information section below in this reply lists our non-steady state dependencies and Figure 8-11 displays the second

values and their evolution over the 80 seconds as a function of increasing H₂O values. When applying these values to the raw data, all parameters are strongly out of range for a large water vapor amount range. As these dependencies are strongly amplified due to non-steady state conditions compared to generally perform dependency series made under steady state conditions, we scaled our dependencies as outlined above. However, we finally ended up in correcting the values based on deriving the H₂O dependencies directly from the air vapor measurements. A detailed analysis of these procedures and its comparison the two other approaches (rapid standard increase measurements as well as steady state standard measurements at different levels) will be part of a separate manuscript as it is out of the scope of this work.

Line Number 242: insertd: The lowest monthly-mean H₂O concentration recorded was 3,600 ppmv; at 12-minute resolution the lowest value was 1,200 ppmv. Mixing-ratio dependence of all isotopic parameters was characterized following the functional form of Weng et al. (2020). Dependencies derived from the rapid signal rise during standard injections over-corrected relative to steady-state behaviour and were not applied directly; dependencies derived from the ambient vapor record itself were used instead. A full comparison of these approaches, and their transfer to steady-state conditions, will be presented in a forthcoming manuscript; this correction should be regarded as a first-order treatment.

Other methodological aspects also remain unclear. For example, it is not specified whether a quality-control standard was measured regularly to evaluate instrumental drift, particularly for the secondary isotope parameters. Furthermore, the authors do not explain how 17O-excess values were assigned to the calibration standards, most of which appear to be in-house standards. Did they use the approach proposed by Schoenemann et al. 2013, to assign 17O-excess values?

No, we have not measured a quality control standard. The four in-house standards have been used since decades at the laboratory of Climate and Environmental Physics, Physics Institute of the University of Bern, Switzerland. They have been extensively calibrated to the international scale V-SMOW, SLAP and GISP for the primary isotope parameters. For the secondary parameters comparison with other institutions were performed, such as Laboratory for Climate and Environmental Sciences, LSCE at Saclay near Paris. These values have been used in publications such as (Affolter et al., 2014, 2015, 2019; Affolter & Leuenberger, 2021; Leuenberger & Ranjan, 2021; Ranjan & Leuenberger, 2021)

References:

- Affolter, S., Fleitmann, D., & Leuenberger, M. (2014). New online method for water isotope analysis of speleothem fluid inclusions using laser absorption spectroscopy (WS-CRDS). *Climate of the Past*, 10(4). <https://doi.org/10.5194/cp-10-1291-2014>
- Affolter, S., Häuselmann, A. D., Fleitmann, D., Häuselmann, P., & Leuenberger, M. (2015). Triple isotope (δD , $\delta^{17}O$, $\delta^{18}O$) study on precipitation, drip water and speleothem fluid inclusions for a Western Central European cave (NW Switzerland). *Quaternary Science Reviews*, 127. <https://doi.org/10.1016/j.quascirev.2015.08.030>
- Affolter, S., Häuselmann, A., Fleitmann, D., Lawrence Edwards, R., Cheng, H., & Leuenberger, M. (2019). Central Europe temperature constrained by speleothem fluid inclusion water isotopes over the past 14,000 years. *Science Advances*, 5(6). <https://doi.org/10.1126/sciadv.aav3809>
- Affolter, S., & Leuenberger, M. C. (2021). Challenges in the Direct Determination of ^{17}O excess in Microliter Amount of Water Extracted From Speleothem Fluid Inclusions. *Frontiers in Earth Science*, 9. <https://doi.org/10.3389/feart.2021.612436>
- Leuenberger, M. C., & Ranjan, S. (2021). Disentangle Kinetic From Equilibrium Fractionation Using Primary ($\delta^{17}O$, $\delta^{18}O$, δD) and Secondary ($\Delta^{17}O$, $\delta^{18}O$) Stable Isotope Parameters on Samples From the Swiss Precipitation Network. *Frontiers in Earth Science*, 9. <https://doi.org/10.3389/feart.2021.598061>
- Ranjan, S., & Leuenberger, M. C. (2021). Comparison of Three Measurement Principles on Water Triple Oxygen Isotopologues. *Frontiers in Earth Science*, 9. <https://doi.org/10.3389/feart.2021.598616>

We have described the approach in Affolter et al., 2015.

The authors also state that instrument calibration could not be performed during part of the monitoring period. This effectively invalidates all measurements collected after calibration ceased. Although this issue is mentioned in the abstract, it is surprisingly scarcely discussed in the remainder of the manuscript.

This is a severe shortcoming; we are aware of. There have been limited standard measurements done during part of the period, however very infrequently as well as incomplete compared to previous calibration sequences. Based on the information given above, the measurements need to be flagged as measurements without proper calibration. The fact that the mean calibration yields relatively good and consistent values over the complete record over an extended range that surpasses the range of

values at Manali and Sissu, we would not call these values invalid data. Especially for the primary data.

Finally, it is noteworthy that although the instrument likely acquired measurements every 2–3 seconds, resulting in a dataset comprising lots of observations, the authors only present monthly averages. It would have been much more informative to present the complete time series, perhaps together with appropriate moving averages or smoothing techniques, so that the temporal variability and trends throughout the monitoring period could be properly evaluated.

This manuscript intends to publish the instrumental setup at high elevation stations in the Himalayas. Of course, we have obtained high resolution data, as shown above, that will be part of upcoming submissions. Based on the reviewers' suggestions, we have used the high-resolution data for calculating the H₂O dependence and noticed a much stronger effect on H₂O for rapid H₂O amount changes. They lead to unexpectedly high corrections for low water amounts that are unrealistic. We conclude from these measurements and evaluation that dependency parameters based on fast H₂O amount changes are not directly representative for ambient water vapor measurements even though they represent also non-steady state conditions. But they require scaling. This highlights the importance of investigating an ideal correction procedure as the generally applied steady state dependencies, in which measurements are used at different H₂O levels which were taken after a long equilibration time to settle to the corresponding H₂O level, may also not be ideal. We suspect that this correction originates from adsorption/desorption processes at the surfaces of the complete system as well as the laser optic dependencies on different water vapor amounts and is strongly dependent on the equilibration required. The lower the water amount in the system the more important these adsorption/desorption effects become as its relative weight increases (limited adsorption sites). This is negligible for higher water amounts (low relative weight). Similarly, the spectral behavior and laser-cavity interactions under changing water vapor conditions is strong. For low water vapor amounts laser signal stability is hampered by the frequency lock stability and is taken care of by the factor (a/x) in the equation taken by Weng et al., [2020]. Whereas under high water vapor amounts the signal stability must be corrected for collision broadening among water molecules, spectral cross talk and flank overlap in some cases. Here the counter-act is the factor $(b \cdot x)$.

Supplementary information:

Table of H2O dependency of all parameters

Table 2: H2O dependency parameters for the four water standards				
Standards	ST-1 (ST-08)	ST-2 (B-Slap)	ST-3 (Dye-III)	ST-4 (MW)
Excess_17_a	-4043.888	-5561.617	-5272.412	-4935.954
Excess_17_b	0.00007632	0.00006594	0.00006672	0.00006849
Excess_17_c	-1.394	-1.006	-1.197	-1.139
Excess_17_offset	175.384	-55.283	-72.23	9.611
Delta_17_16_a	1554.082	2034.844	1340.901	1027.446
Delta_17_16_b	0.00014502	0.00011815	0.00014204	0.00015968
Delta_17_16_c	-9.053	-26.764	-15.806	-5.021
Delta_17_16_offset	289.066	357.157	367.083	348.225
Delta_18_16_a	9249.89	11007.314	9113.778	8276.621
Delta_18_16_b	0.00011646	0.00007406	0.0001116	0.00013896
Delta_18_16_c	-14.171	-47.705	-26.848	-6.624
Delta_18_16_offset	260.013	246.21	240.039	247.769
Delta_D_H_a	-110306.35	11035.97	-156014.74	-217127.14
Delta_D_H_b	-0.0002569	-0.0004266	-0.0005237	-0.0004127
Delta_D_H_c	-62.44	-268.623	-200.496	26.461
Delta_D_H_offset	-2938.017	-592.405	-7021.263	-2797.994
$y_{corr} = y - [a/(x-offset) + b*(x-offset) + c]$				

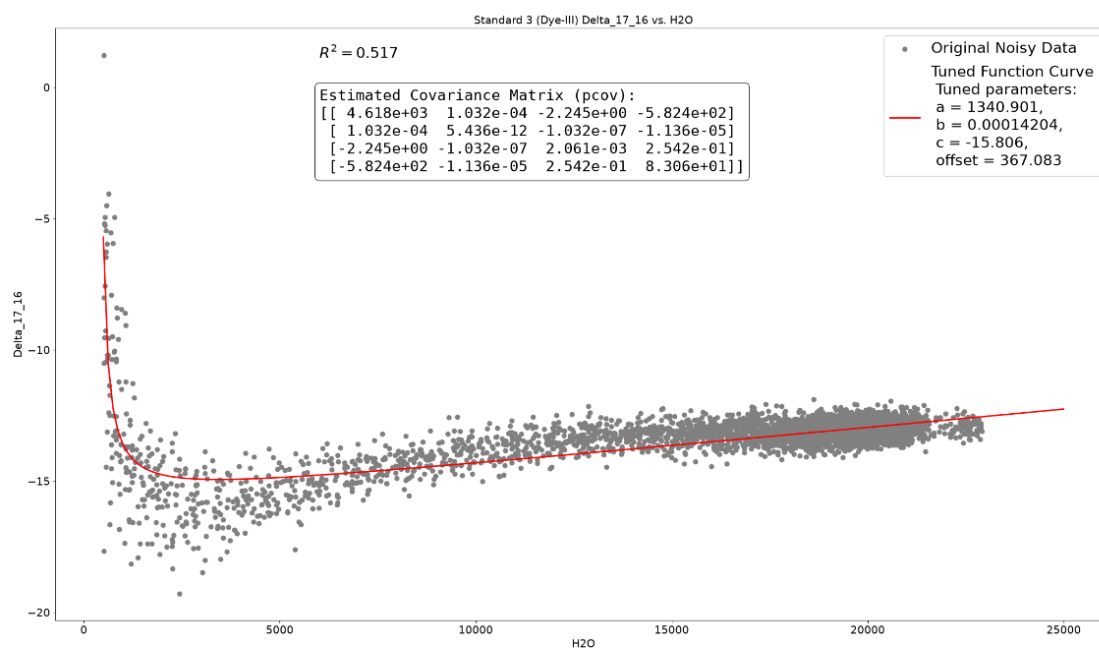


Figure 8: Delta_17_16 H2O dependence for non-steady state.

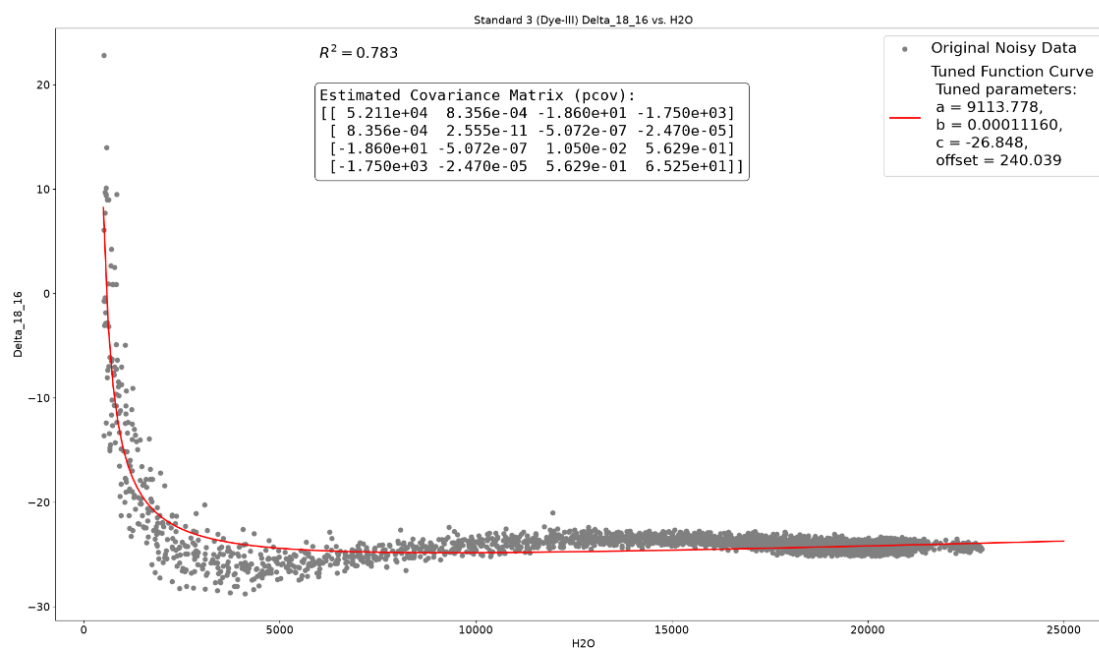


Figure 9: Delta_18_16 H2O dependence for non-steady state

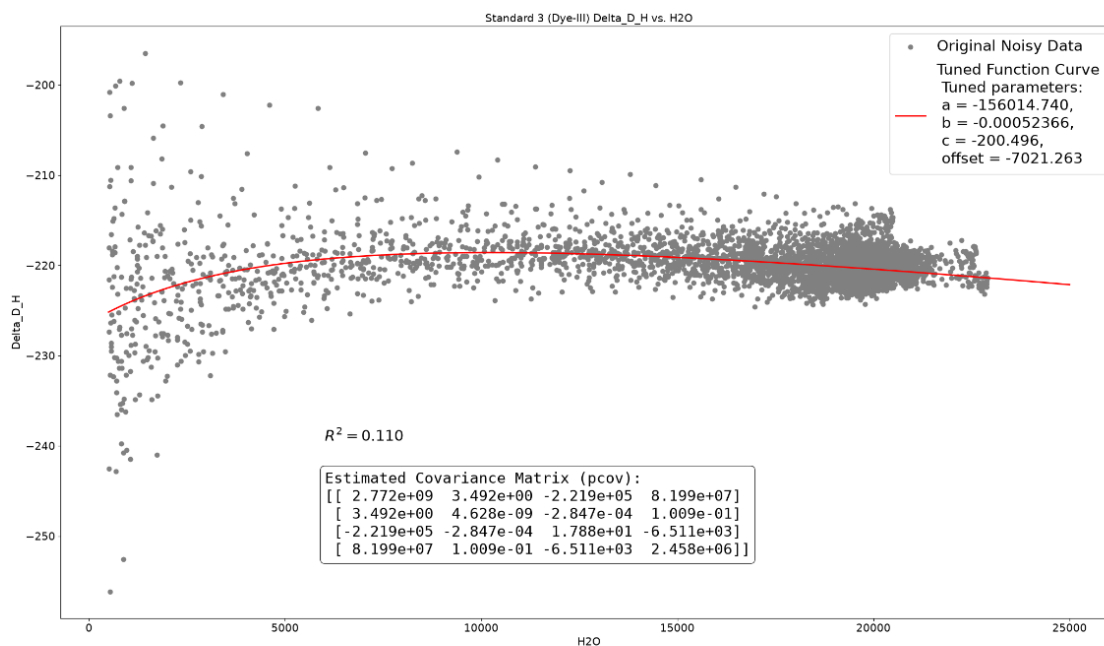


Figure 10: Delta_D_H H2O dependence for non-steady state

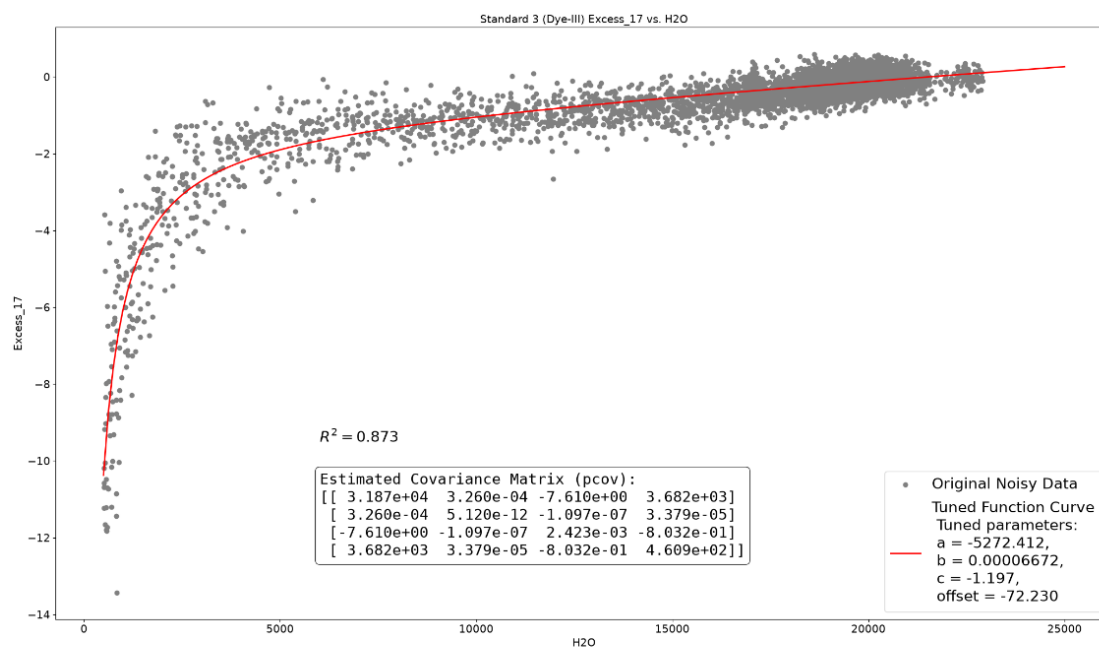


Figure 11: Excess_17 H2O dependence for non-steady state