



Supplement of

SI-traceable validation of a laser spectrometer for balloon-borne measurements of water vapor in the upper atmosphere

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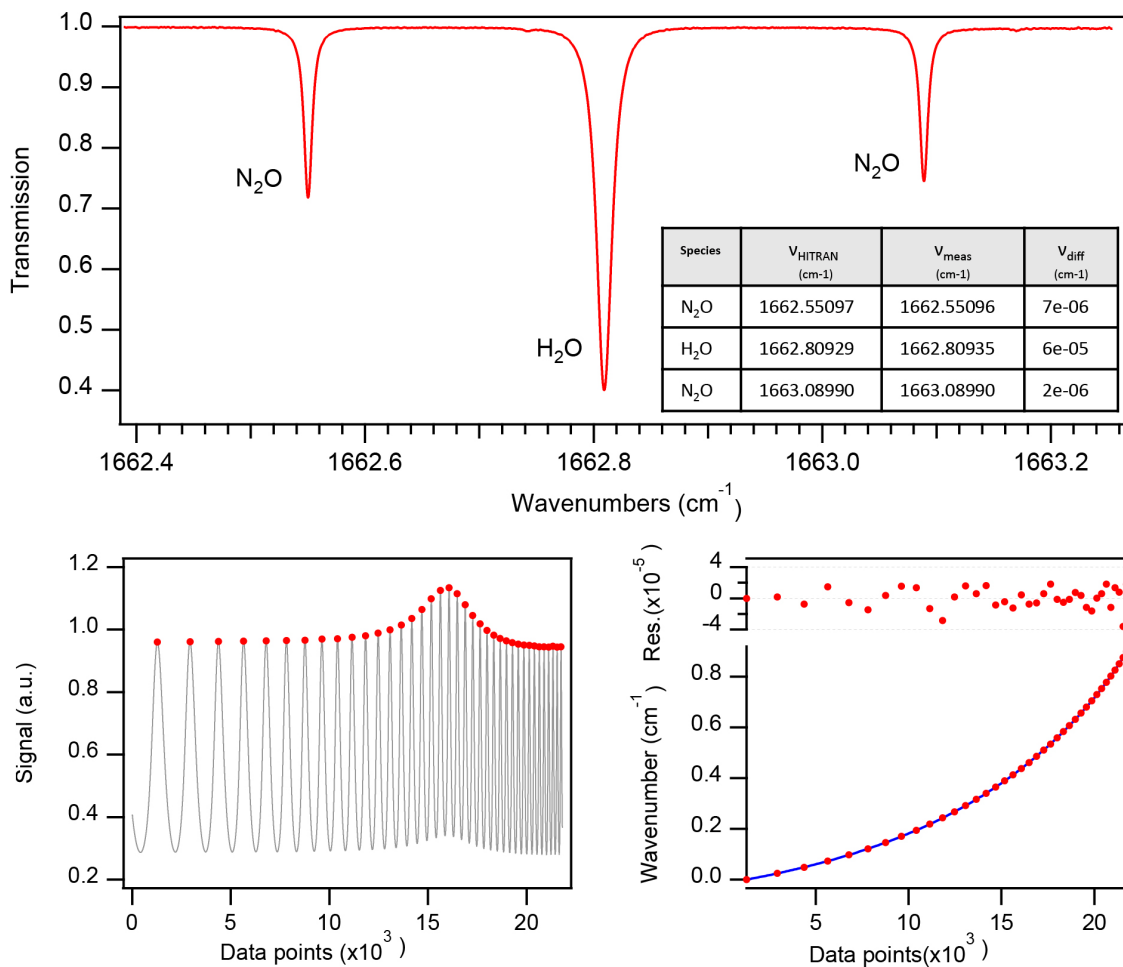


Figure S1: Absolute frequency scale determination. Top: measured normalized spectrum recorded using pure N₂O with 260 ppm H₂O content. The two neighboring N₂O transitions were used for the frequency scale determination. The measured position of each line along with their tabulated values in the HITRAN2020 database, as well as their differences, are listed in the inset table. Bottom, left panel: normalized Ge-etalon transmission spectrum as function of sampling points. The solid circles indicate the etalon fringe positions. Bottom, right panel: laser tuning curve (i.e., free spectral range (FSR) index vs. etalon fringe position index). The FSR value (0.02429 cm⁻¹) was first estimated using the definition $\text{FSR} = 1/(2n(\lambda, T)L)$, where $n(\lambda, T)$ is the refractive index and L (= 2 in) is the length of the Ge-etalon, followed by an iterative optimization using the N₂O transitions.

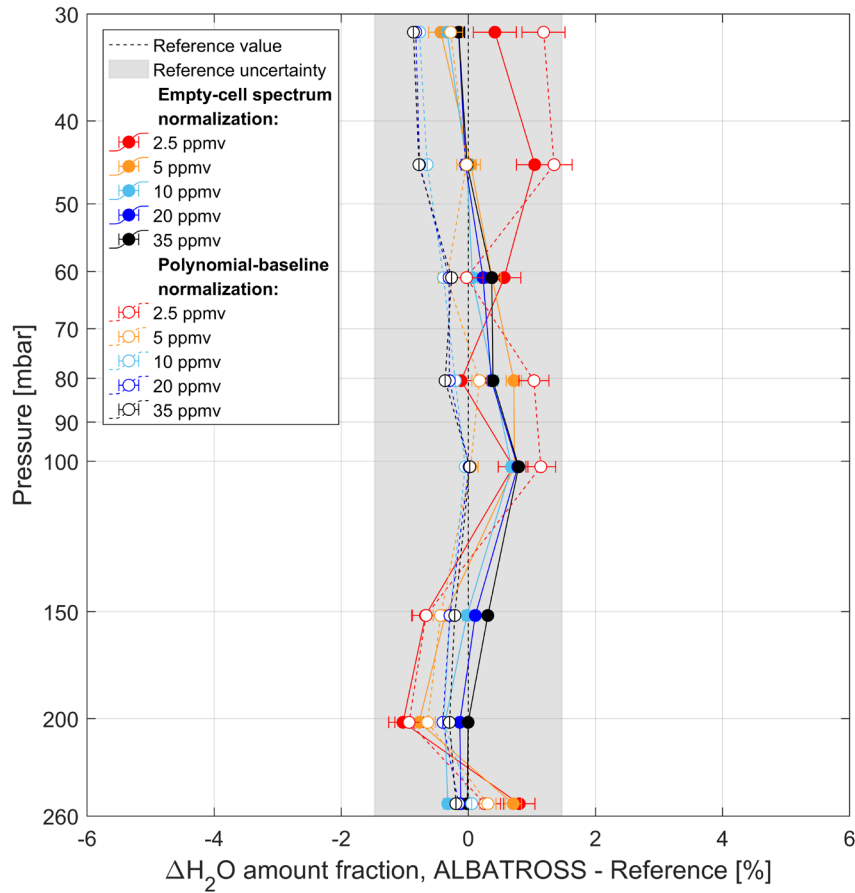


Figure S2. Comparison of accuracy assessed using the polynomial-baseline (open circles/dashed line) and the empty-cell spectrum (solid circles/solid line) normalization approaches. The relative differences in H_2O amount fraction ($\Delta\text{H}_2\text{O}$), between the ALBATROSS retrievals and the reference values are shown as function of pressure and H_2O amount fraction (color-coded as in Figure 8).

5 All measurements are retrieved using the qSDVP line shape model with the molecular parameters derived by the multi-spectrum fitting (MSF) method (see Table 2). For the polynomial-baseline normalization, the laser intensity is described by a 4th order polynomial function (see e.g. Graf et al., 2021).

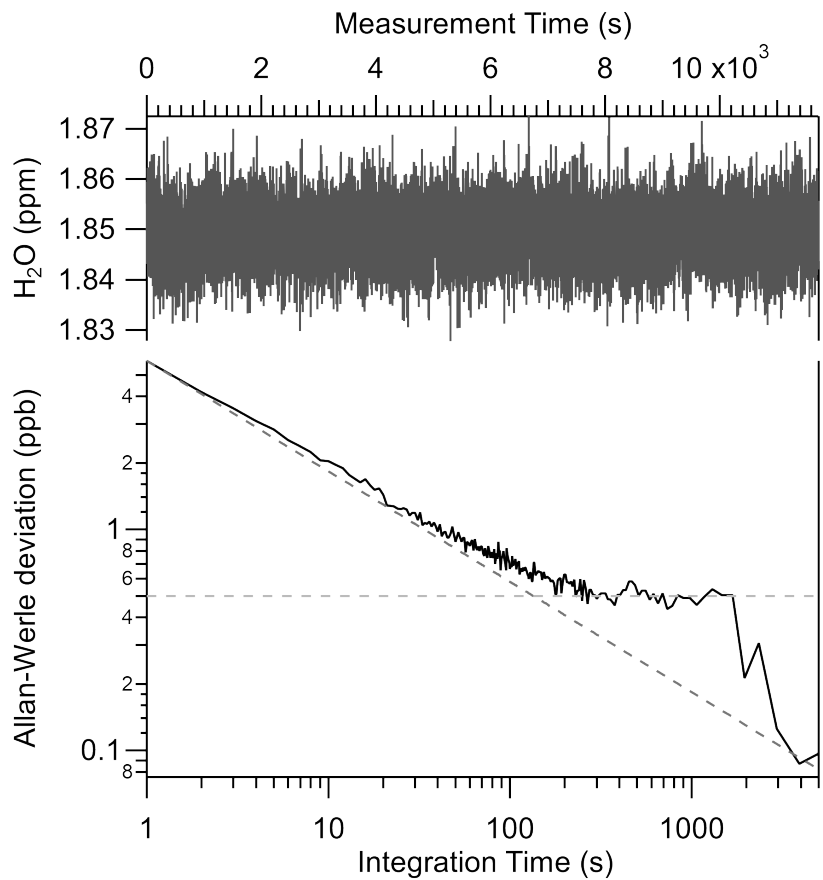


Figure S3. Allan-Werle deviation plot of a ~1.5 h interval of zero-air (i.e., dry synthetic air) measurement performed at a gas pressure of 73 mbar. Top panel: time-series of the measured H₂O amount fraction (retrieved using the polynomial-baseline normalization approach). Bottom panel: Allan-Werle deviation as function of integration time.