



*Supplement of*

**Vertical and horizontal variability and representativeness of the water vapor isotope composition in the lower troposphere: insight from ultralight aircraft flights in southern France during summer 2021**

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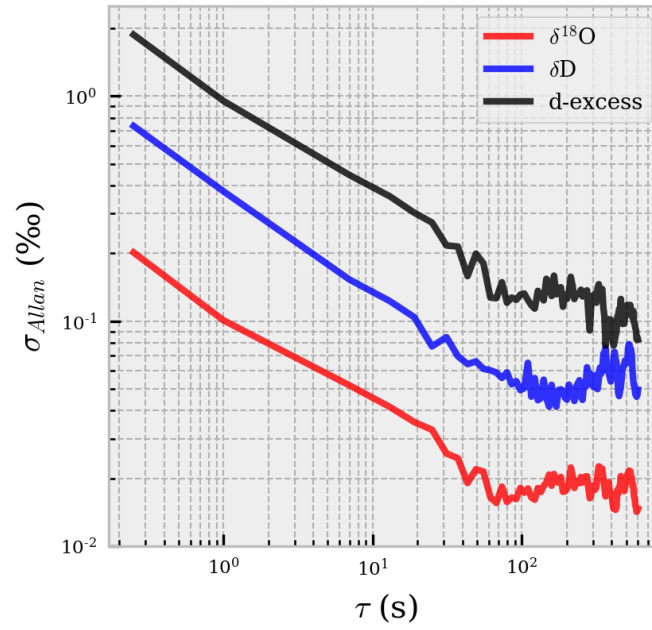
## Standards

**Table S1:** Water isotope standards used for calibrating the CRDS analyzer. A single “X” means single humidity level calibration ( $\sim 10\text{--}12\text{ g kg}^{-1}$ ) while “XX” means multiple humidity level calibration for humidity-isotope characterization (between 0.6 and  $12.5\text{ g kg}^{-1}$ ). \*Used also for specific humidity calibration with reference chilled mirror hygrometer (Panametrics Optisonde).

Standard	Campaign day						
	17.09	18.09	19.09	20.09	21.09	22.09	23.09
<b>BERM</b> $\delta^{18}\text{O} = 0.58 \pm 0.01\text{‰}$ $\delta\text{D} = 6.5 \pm 0.1\text{‰}$	X	X	XX	X	X	X	X
<b>FIN</b> $\delta^{18}\text{O} = -11.65 \pm 0.02\text{‰}$ $\delta\text{D} = -81.1 \pm 0.1\text{‰}$					XX*		
<b>GLW</b> $\delta^{18}\text{O} = -40.06 \pm 0.02\text{‰}$ $\delta\text{D} = -308.1 \pm 0.2\text{‰}$	X	X	XX	XX	X	X	X

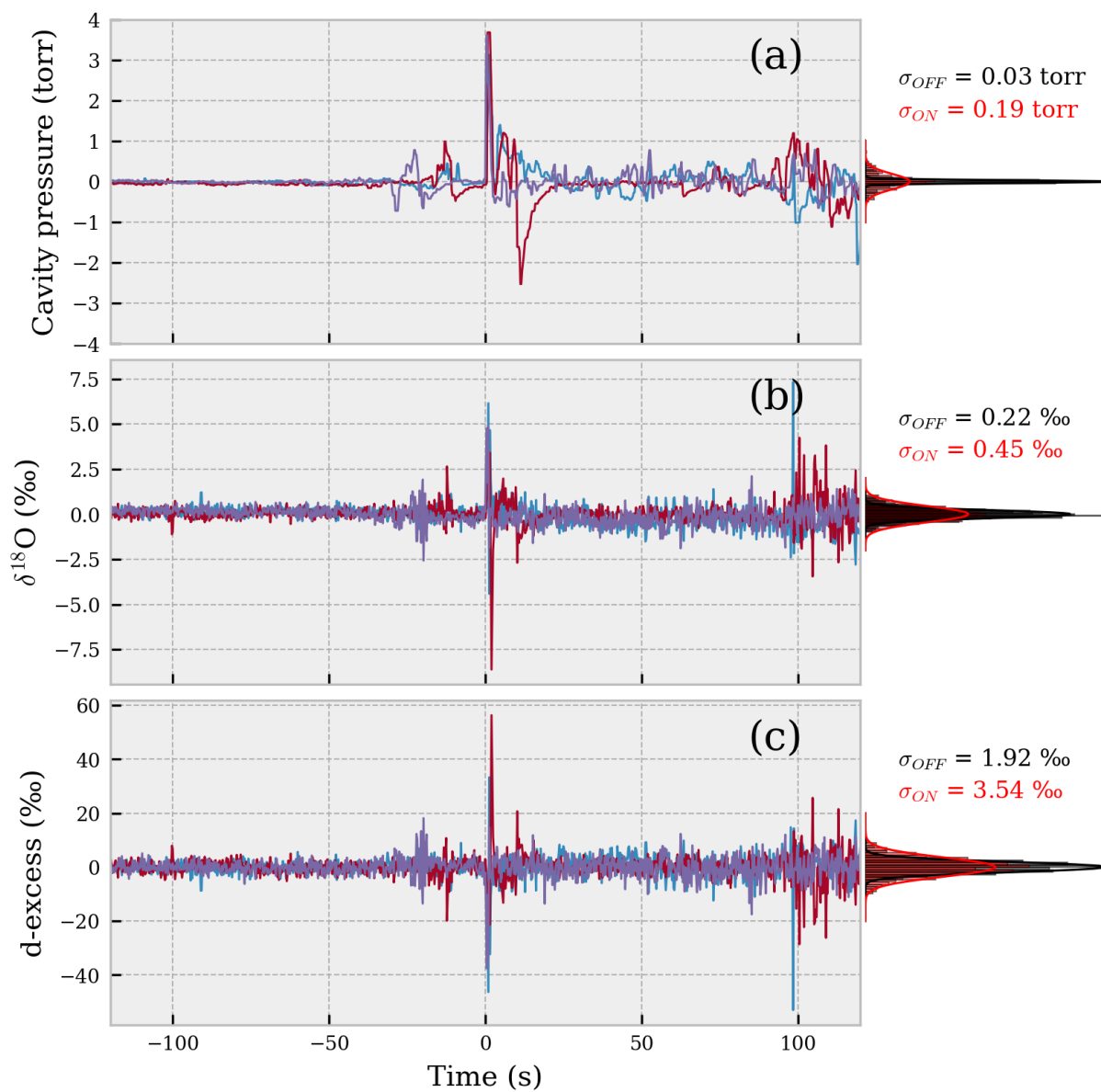
The VSMOW-SLAP slope of the calibration line varied between 1.118 - 1.132 and 0.914 - 0.928 for  $\delta^{18}\text{O}$  and  $\delta\text{D}$ , respectively, with no visible trend during the study period. Such slope values are consistent with the long-term slope variability of the instrument estimated between 2016-2022 ( $\delta^{18}\text{O}$  slope =  $1.1305 \pm 0.0095$ ,  $\delta\text{D}$  slope =  $0.9253 \pm 0.0027$ ), thus ensuring reliable instrument performances during the field operations.

## Allan deviation test



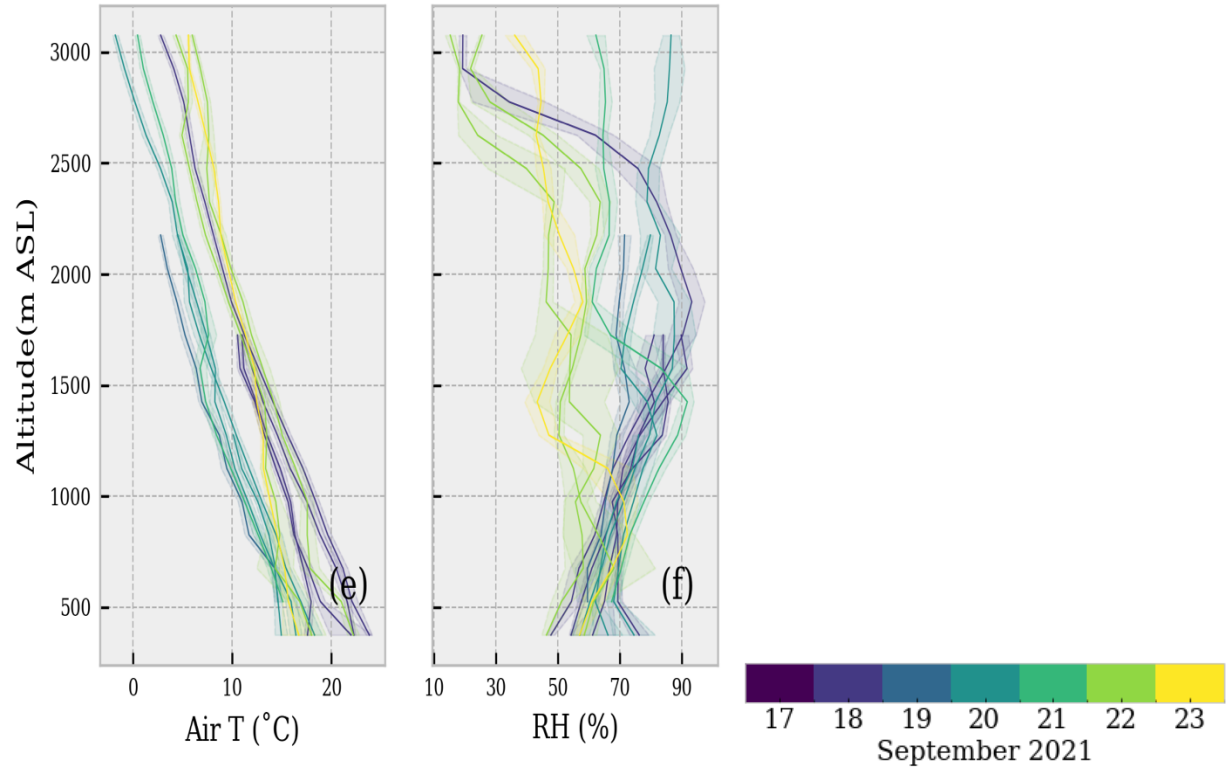
**Fig. S1:** The Allan deviation test was performed injecting BER standard for 90 minutes. The humidity level was set to  $q = 8.3 \pm 0.3 \text{ g kg}^{-1}$ . The first 30 minutes of the injection were discarded to minimize memory effect.

Engine vibrations impact on the CRDS analyzer performances.



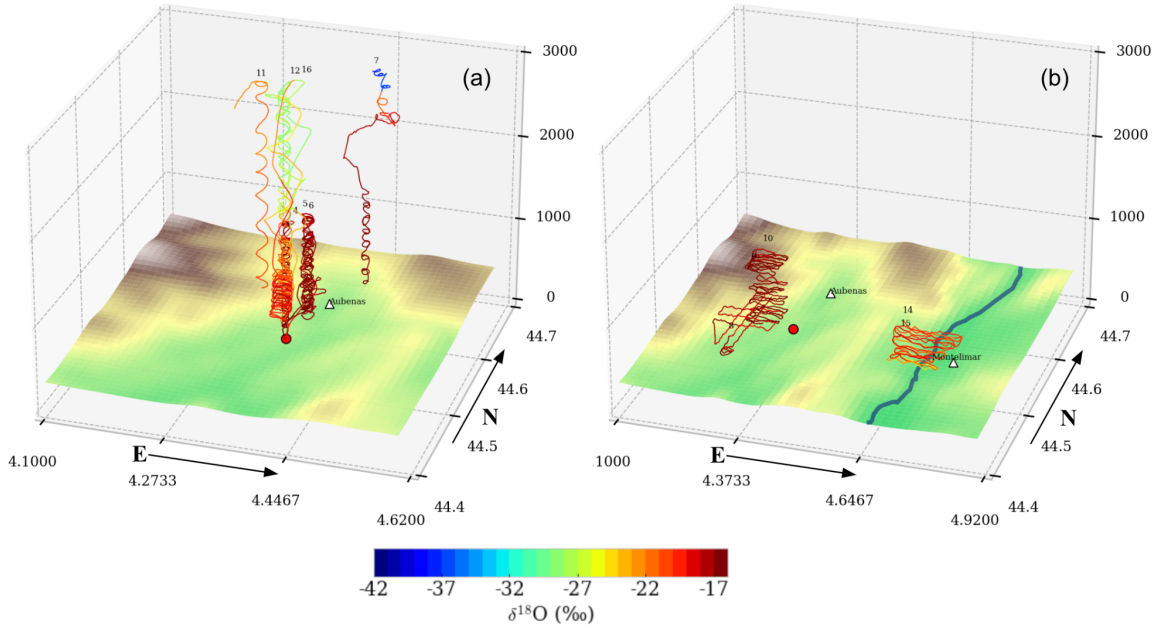
**Fig. S2:** Impact of engine vibrations on CRDS analyzer performances. Time series of deviations from mean values of cavity pressure (a), of  $\delta^{18}\text{O}$  (b) and of d-excess (c). Time = 0s when ULA engine was turned on. Blue lines for flight 7, red for flight 8, purple for flight 9. Best fit of normal distribution on same quantities are reported over marginal histograms on the right (black: engine OFF, red: engine ON). Standard deviation values are reported for reference ( $\sigma_{\text{OFF}}$ ,  $\sigma_{\text{ON}}$ ).

### Vertical profiles

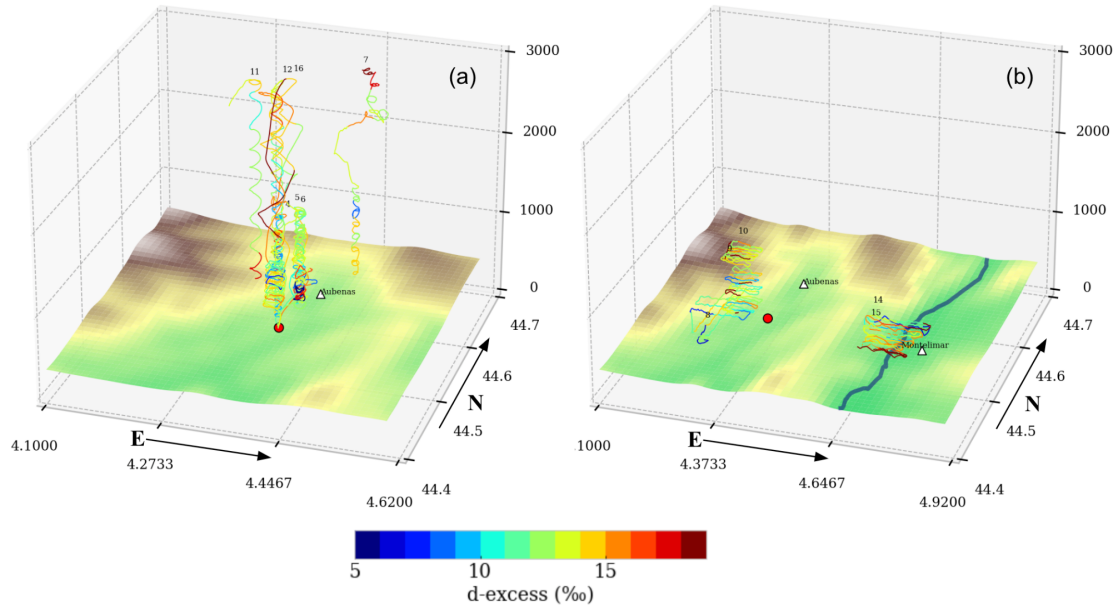


**Fig. S3:** Supplement to Fig. 6 in the main text: vertical profiles of air temperature **(e)** and relative humidity **(f)**. Solid line represents the average calculated over a 150m bin size. Shadings represent  $\pm 1\sigma$  interval around the mean.

### 3D pattern for $\delta^{18}\text{O}$ and d-excess



**Fig. S4:**  $\delta^{18}\text{O}$  recorded during flights used to probe the spatial variability of the isotopic composition of water vapor. **(a)** Soundings recorded during flights 4-7, 11, 12, 16. **(b)** Observations recorded at different altitudes during flights 8-10, 14, 15. Ground height vertical exaggeration  $\sim 2$ .



**Fig. S5:** d-excess recorded during flights used to probe the spatial variability of the isotopic composition of water vapor. **(a)** Soundings recorded during flights 4-7, 11, 12, 16. **(b)** Observations recorded at different altitudes during flights 8-10, 14, 15. Ground height vertical exaggeration  $\sim 2$ .

## R<sup>2</sup> table

**Table S2:** Coefficient of determination of linear model  $\log(q)$  vs  $\delta$  for vertical and horizontal pattern flights.

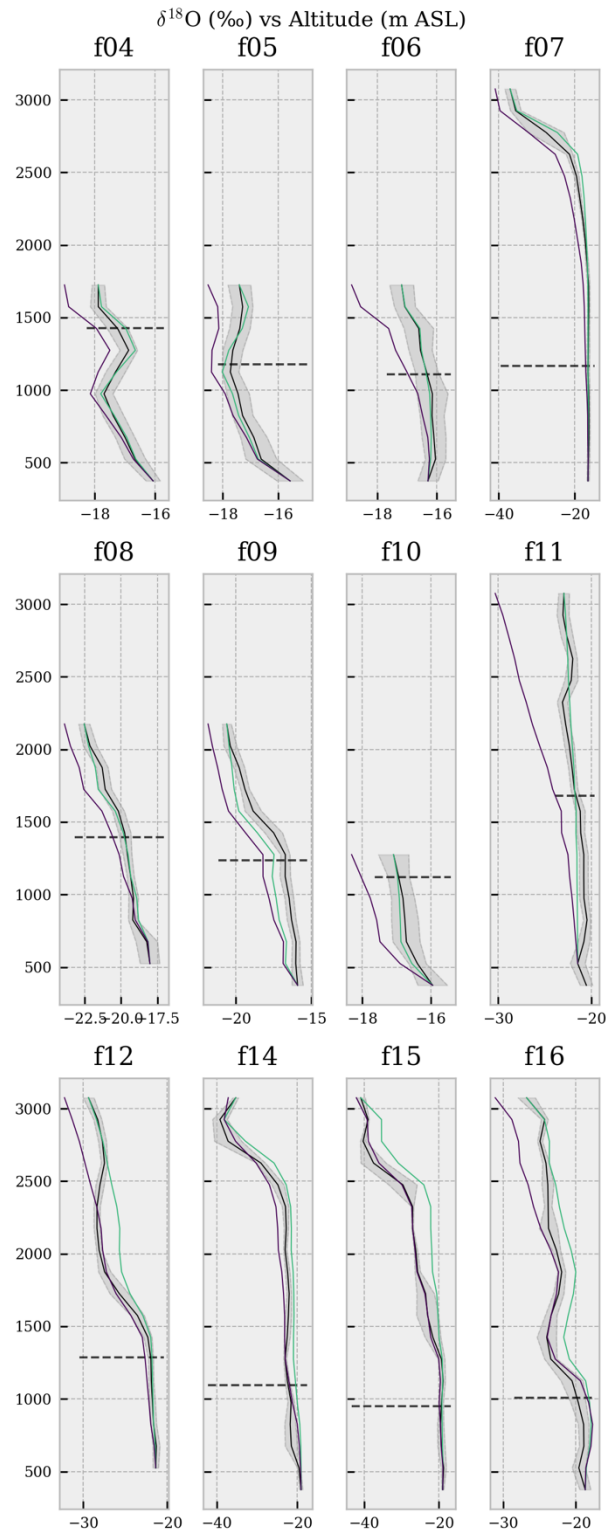
### Vertical pattern

Flight	$\delta^{18}\text{O}$	$\delta\text{D}$	d-excess
4	0.74	0.92	0.10
5	0.76	0.94	0.00
6	0.58	0.92	0.01
7	0.99	0.99	0.34
11	0.67	0.79	0.00
12	0.92	0.94	0.06
16	0.92	0.92	0.18
<b>Average</b>	<b>0.80</b>	<b>0.92</b>	<b>0.10</b>

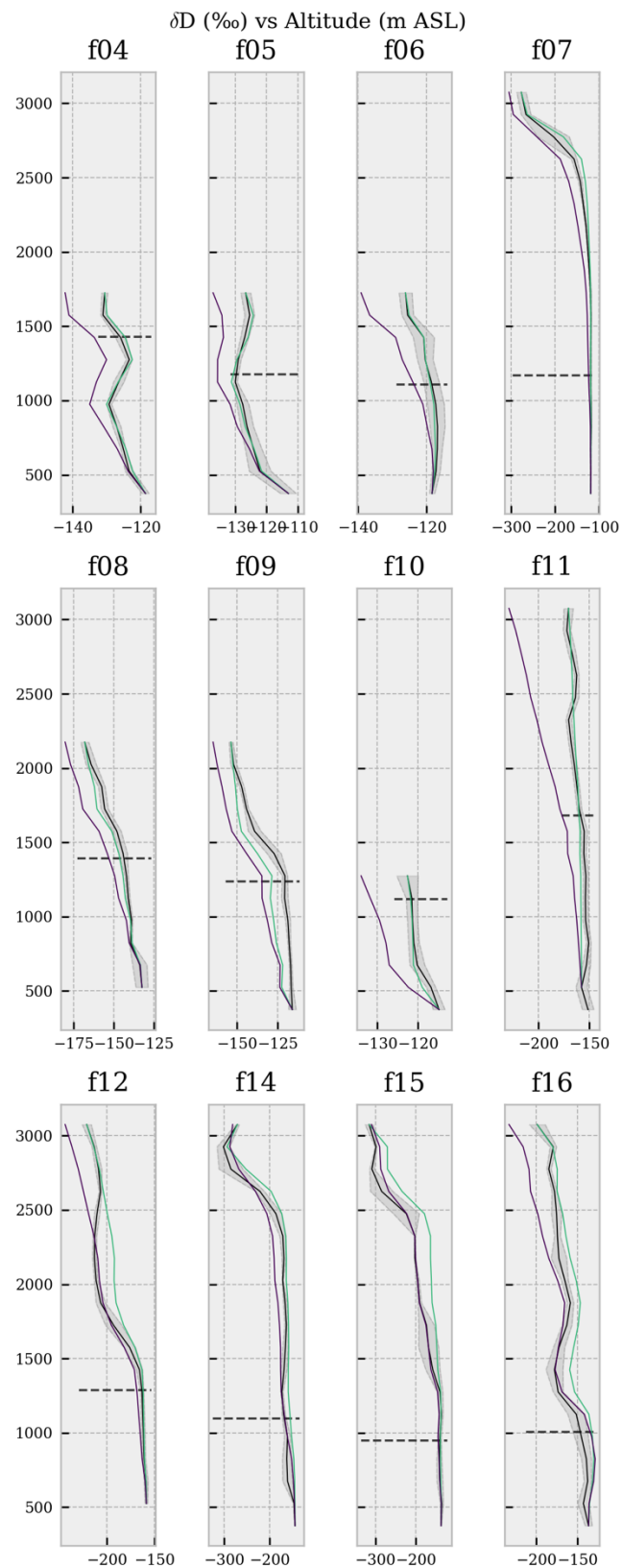
### Horizontal pattern

Flight	$\delta^{18}\text{O}$	$\delta\text{D}$	d-excess
8	0.72	0.86	0.10
9	0.62	0.71	0.28
10	0.35	0.67	0.03
14	0.52	0.53	0.07
15	0.83	0.90	0.02
<b>Average</b>	<b>0.61</b>	<b>0.74</b>	<b>0.10</b>

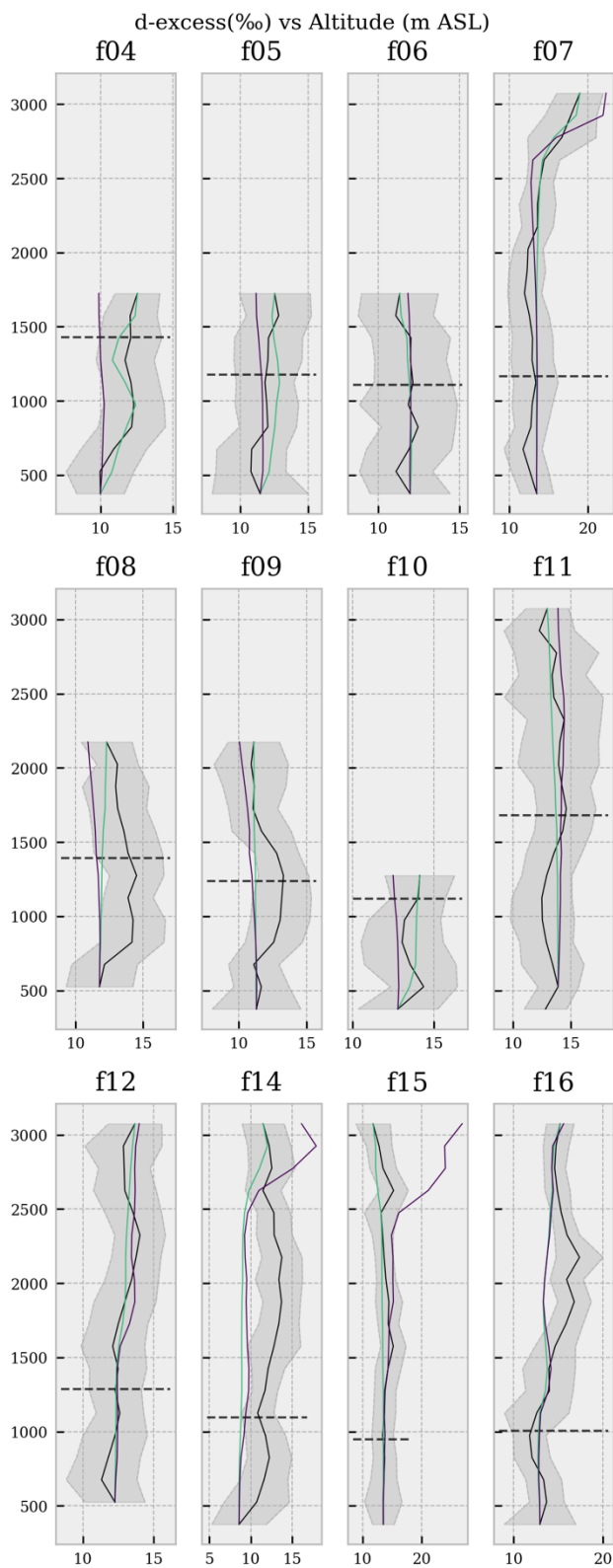
## Simulated vertical profiles



**Fig. S6.a:** Modeled water vapor  $\delta^{18}\text{O}$  (solid black line  $\pm 1$  standard deviation) compared to Rayleigh (purple) and binary mixing (green) model. Horizontal dashed black lines report BLH (ERA5) at the time of the flight.

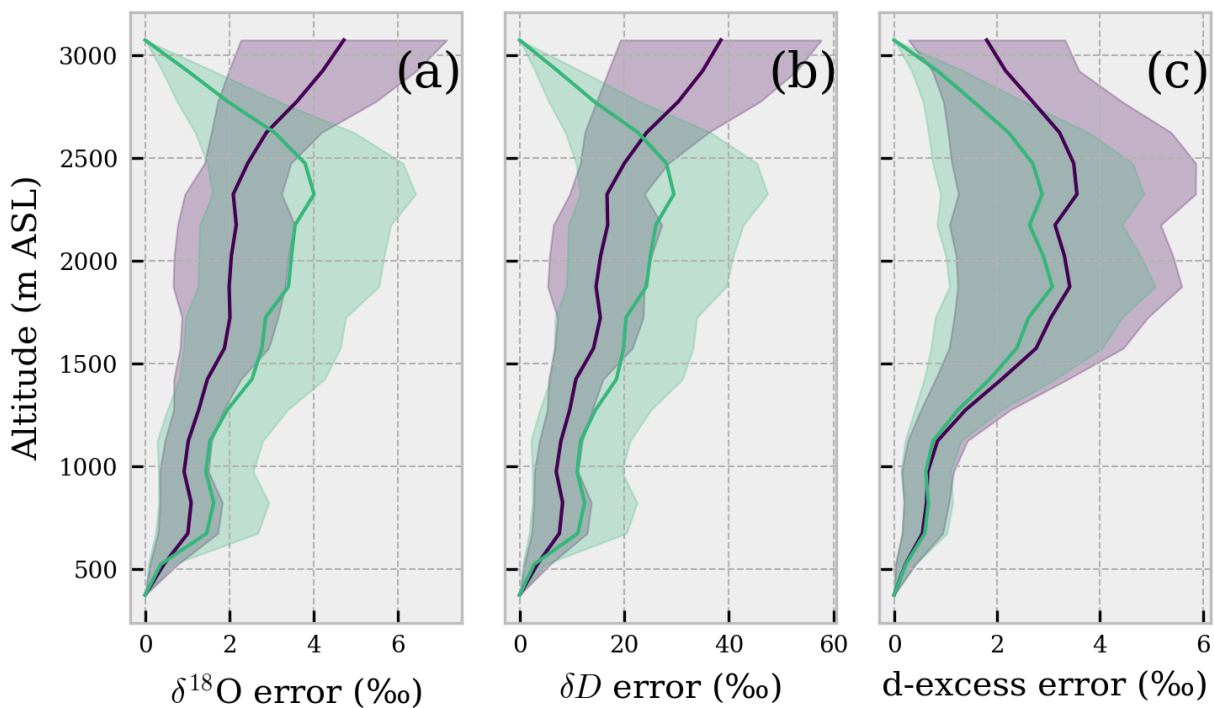


**Fig. S6.b:** Modeled water vapor  $\delta D$  (solid black line  $\pm 1$  standard deviation) compared to Rayleigh (red) and binary mixing (blue) model. Horizontal dashed black lines report blh (ERA5) at the time of the flight.



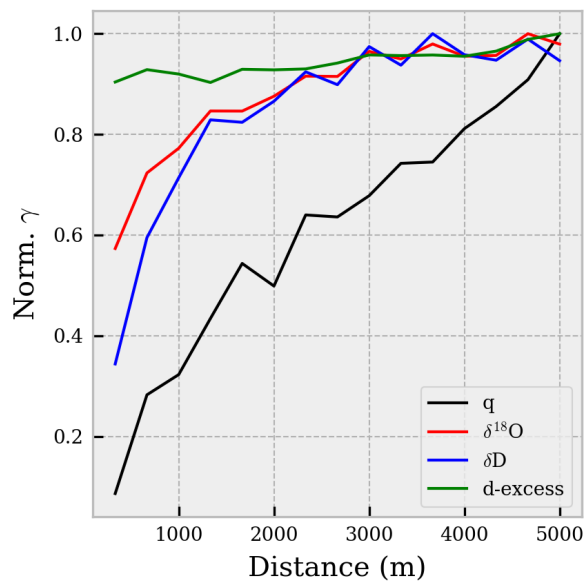
**Fig. S6.c:** Modeled water vapor d-excess (solid black line  $\pm$  1 standard deviation) compared to Rayleigh (red) and binary mixing (blue) model. Horizontal dashed black lines report blh (ERA5) at the time of the flight.

### Conceptual models applied on COSMO<sub>iso</sub> simulations.



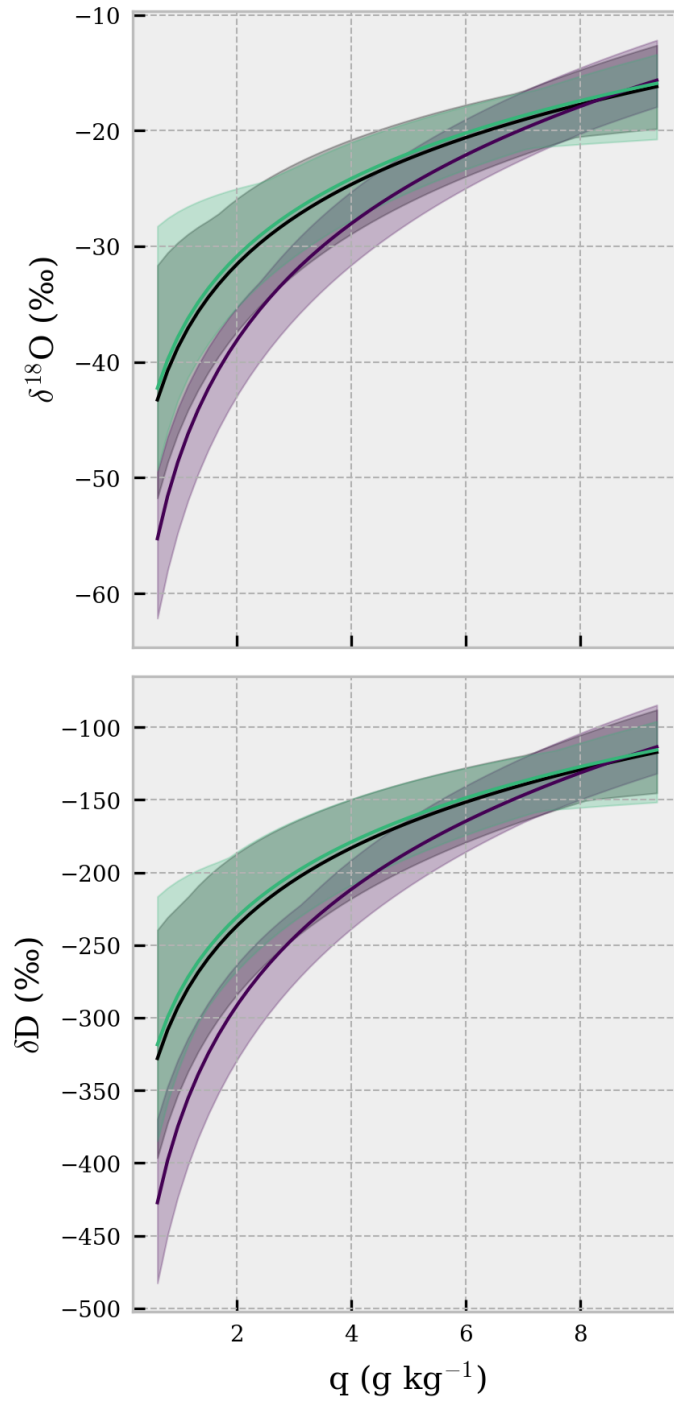
**Fig. S7:** Root Mean Square Error (RMSE) between conceptual models and COSMO<sub>iso</sub> simulations averaged per height levels for  $\delta^{18}\text{O}$  (a),  $\delta\text{D}$  (b) and d-excess (c). This figure has the same colors of Fig. 12 in the manuscript: purple is Rayleigh model, green is binary mixing model. Solid lines represent the average error calculated over a 150m bin size for all the flights and shadings represent the standard error of the mean.

### Normalized variograms



**Fig. S8:** Normalized semivariance of water vapor mixing ratio and isotopic composition for spatially resolved flights (horizontal pattern flights).

### Models best fit



**Fig. S8:** Comparison of  $\log(q)$  vs  $\delta$  best fits for observations (black), Rayleigh distillation model (purple) and binary mixing model (green). The solid lines indicate the ensemble means while shadings indicate the min-max ranges.

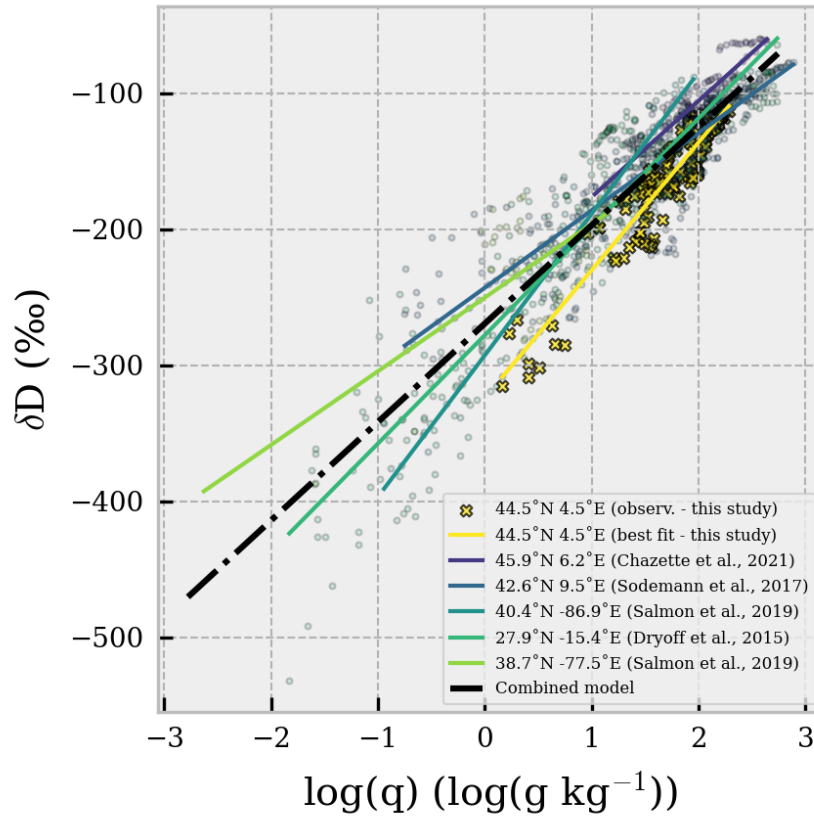
**Table S3.a:** Best fits parameters of the model  $\delta^{18}O = \beta_0 \log(q) + \beta_1$  for each flight.

	OBS			Rayleigh			Mix model		
Flight	beta0	beta1	r <sup>2</sup>	beta0	beta1	r <sup>2</sup>	beta0	beta1	r <sup>2</sup>
4	11.002	-40.746	0.969	16.219	-52.400	0.843	11.587	-41.934	0.999
5	10.456	-39.486	0.961	14.192	-48.023	0.824	11.464	-41.717	0.999
6	6.954	-31.538	0.986	16.276	-52.451	0.965	6.298	-30.147	1.000
7	10.162	-38.342	0.995	12.141	-43.751	0.996	9.652	-36.994	0.958
8	10.786	-38.145	0.976	15.459	-46.713	0.998	11.989	-40.211	0.997
9	13.725	-43.280	0.944	15.970	-48.688	0.995	13.265	-43.032	0.998
10	7.425	-31.078	0.958	15.332	-47.199	0.952	7.724	-31.742	1.000
11	4.358	-29.582	0.792	16.395	-54.356	0.998	2.778	-26.954	0.996
12	12.262	-45.935	0.942	14.895	-51.212	0.996	10.917	-42.907	0.993
14	12.668	-44.609	0.966	12.789	-45.215	0.994	12.303	-42.407	0.969
15	12.391	-44.492	0.995	12.062	-44.130	0.999	10.423	-39.085	0.949
16	7.759	-35.041	0.933	13.789	-45.717	0.990	8.291	-34.538	0.978

**Table S3.b:** Best fits parameters of the model  $\delta D = \beta_0 \log(q) + \beta_1$  for each flight.

	OBS			Rayleigh			Mix model		
Flight	beta0	beta1	r <sup>2</sup>	beta0	beta1	r <sup>2</sup>	beta0	beta1	r <sup>2</sup>
4	72.670	-281.544	0.971	129.717	-409.067	0.833	76.442	-289.184	0.999
5	79.586	-295.366	0.984	113.364	-372.325	0.806	85.061	-307.095	0.999
6	59.959	-249.920	0.996	130.991	-409.347	0.966	54.919	-239.203	1.000
7	78.248	-287.401	0.994	93.022	-328.089	0.994	74.622	-276.939	0.958
8	86.757	-292.617	0.969	125.926	-365.969	0.998	94.533	-307.349	0.997
9	112.673	-339.685	0.926	130.895	-384.464	0.994	106.661	-334.068	0.998
10	54.061	-224.676	0.916	124.066	-367.629	0.945	52.699	-222.564	1.000
11	34.819	-223.062	0.829	131.010	-420.426	0.997	23.992	-205.220	0.996
12	95.837	-350.886	0.945	116.661	-392.378	0.995	85.404	-327.173	0.993
14	100.890	-344.071	0.963	96.579	-342.309	0.991	96.224	-326.495	0.969
15	99.558	-342.827	0.996	90.051	-327.033	0.997	84.210	-300.748	0.949
16	58.912	-260.662	0.930	107.902	-348.087	0.989	64.008	-258.924	0.978

# $\delta D$ vs $q$ in semi-log space



**Fig. S10:** Same as Fig.14 in the manuscript but in semi-log space:  $\delta D$  vs of  $q$  over 150 m binned vertical profiles estimated for different airborne campaigns. The legend reports the coordinates of the flights and the reference study. Symbols are observations, solid lines are average best-fit curves. The black dot-dashed line is the best-fit curve combining all the observations. The best fit model for all the curves is  $\delta D = \beta_0 * \log(q) + \beta_1$ .