



## Supplement of

# Impact of land–water sensitivity contrast on MOPITT retrievals and trends over a coastal city

Ian Ashpole and Aldona Wiacek

*Correspondence to:* Ian Ashpole (ian.ashpole@smu.ca)

The copyright of individual parts of the supplement might differ from the CC BY 4.0 License.

10

## Section S1

Figure S1 shows that a land-water sensitivity contrast also exists at lower troposphere levels of the profile in the MOPITT TIR-only product ( $\mathbf{a}$  – equivalent to Fig. 3 in the main article), with the biggest difference in the JJA season, as is the case with the TIR-NIR product. Consequently, the temporal trend identified using WLS regression (method outlined in Section 3.2.2) is underestimated over water compared to land in this season (b – equivalent to Fig. 9 in the main article). In DJF, the sensitivity contrast is much smaller and the identified temporal trends are more similar. Note that we only show WLS results for days when the L3 surface index is mixed, and we do not compare regression results to those identified using the original L3 timeseries here, since the purpose of SM1 is just to demonstrate that land-water differences exist in the TIR-only product.



**Figure S1 (a)** Mean retrieval averaging kernel (AK) rows and DFS values for L3L ("Land", left column) and L3W ("Water", right column) in DJF (top row) and JJA (bottom row), selected profile levels only, from the TIR-only product. Filled diamonds indicate diagonal value location for that AK. Numbers in legend indicate corresponding retrieval level of AK and show the mean rowsum for that AK level. See main text for definition of L3L and L3W. This figure is equivalent to Figure 3 in the main article.



Figure S1 (b) WLS regression best-fit lines calculated from seasonal mean retrieved surface VMR timeseries in DJF (top row) and JJA (bottom row), from the TIR-only product. Lines correspond to L3L (green) and L3W (blue). The daily observations corresponding to each seasonal mean value are represented by colour-coded boxplots each year, and the seasonal mean value is represented by filled squares. Note that only days when the original L3 surface index is "mixed" are considered in this analysis (see explanation in the main text). The dashed red line is the mean of the corresponding seasonal mean a priori data from each of the timeseries in the respective panel. Colour-coded values below the top x axis correspond to the number of observations each season. Values in the legend are the value, standard error, and probability of zero value of

75

the trend, respectively.

Figure S2 shows that sensitivity differences in retrievals over land (L3L) and water (L3W) are greater and more persistent (i.e. sensitivity is regularly greater over one surface than the other) in the LT than MT and UT; and more pronounced in JJA than in DJF (as discussed in Section 3.1.2).



**Figure S2** Boxplots for the distribution of differences (L3W - L3L) in (a) AK rowsums and DFS values; and (b) AK diagonal values, that correspond to the mean AK plots shown in Figure 3. Squares = mean differences, with the p value associated with each mean difference (from a 2-tailed Student's t test) given on the right-hand side y axis. Plus symbols = outliers<sup>†</sup>.

<sup>†</sup>Outliers defined as: above (below) percentile 75 (25) + (-) 1.5 \* interquartile range

85

Figure S3 corresponds to Figure 6, discussed in Section 3.1.4. The difference here is that, while Figure 6 is a seasonal median plot, including all L2 retrievals made over the plotted region in DJF and JJA, Figure S3 only consists of retrievals made on days when L3L and L3W are both present, i.e., only the days analysed in Section 3.1.

DJF, Surface, retrieved DJF, Surface, RET-APR DJF, Surface, a priori 4.16, (p = 0.001) 479 439 JA, Surface, retrieved JA, Surface, RET-APR IIA, Surface, a priori 6.52.(p = 0.000)47°N -12 -16 45 ° -20 43°N 41°N 69 61°W 61°V 65°W 63°W 125 140 155 170 185 200 215 230 245 260 275 ppb

Figure S3 Median L2<sup>†</sup> retrieved VMR (left column), a priori VMR (centre column), and RET-APR (right 95 column) at the surface profile level for DJF (top row) and JJA (bottom row), for days when L3L and L3W are both present. Values to the right above RET-APR plots = (L2 retrievals over water) – (L2 retrievals over land) for plotted area (data were first binned according to L2 surface index); numbers in brackets correspond to significance of mean difference using a 2-tailed Student's t test. Blue or green dashed square = outline of L3 grid box that contains Halifax. White shading = gridboxes with no retrievals 100 on the days analysed.

<sup>†</sup>These maps were created from L2 data that were interpolated to a regular 0.25° x 0.25° grid for ease of plotting.

Figure S4 corresponds to Figure 6, discussed in Section 3.1.4, but for selected other profile levels (Figure S4a-d) and for CO Total Column (TCO) (Figure S4e).



110

Figure S4 (a) Seasonal median L2<sup>†</sup> retrieved VMR (left column), a priori VMR (centre column), and RET-APR (right column) at the 900 hPa profile level in DJF (top row) and JJA (bottom row). Values to the right above RET-APR plots = (L2 retrievals over water) - (L2 retrievals over land) for plotted area (data were first binned according to L2 surface index); numbers in brackets correspond to significance of mean difference using a 2-tailed Student's t test. Blue or green dashed square = outline of L3 grid box that contains Halifax.

<sup>†</sup>These maps were created from L2 data that were interpolated to a regular 0.25° x 0.25° grid for ease of plotting.







130 Figure S4(c) As Figure S4a, but for the 600 hPa profile level.







Figure S4(e) As Figure S4a, but for CO Total Column (TCO).

140 Figure S5 corresponds to Figure 7, discussed in Section 3.1.5, but for selected other profile and model wind levels.



145

**Figure S5 (a)** Mean ERA-Interim 850 hPa winds (vectors) and MOPITT  $L2^{\dagger}$  VMR at the 800 hPa profile level (shading) for days when retrieved surface level VMRs in L3W are greater than in L3L (L3W > L3L) and days when they are less (L3W < L3L). Top row = DJF; bottom row = JJA.

<sup>†</sup>These maps were created from L2 data that were interpolated to a regular 0.25° x 0.25° grid for ease of plotting.



**Figure S5 (b)** As Figure S5a but for mean ERA-Interim 500 hPa winds (vectors) and MOPITT L2<sup>†</sup> VMR at the 600 hPa profile level (shading).

160

Figure S6 corresponds to Figure 8, discussed in Section 3.2.1. The difference here is that, while Figure 8 includes *all* data in the L3L, L3W and L3O timeseries, Figure S6 only consists of retrievals made on days when L3L and L3W are both present and the L3O surface index is "mixed", i.e., only the days analysed in Section 3.1. (Note that there are no observations with a L3O surface index of "water" (L3O<sub>(water)</sub>) owing to the fact that there is only one L3O observation per day in the timeseries.)



#### 165

**Figure S6** Boxplots showing the distribution of retrieved surface level VMR values from L3W, L3L, L3O,  $L3O_{(mixed)}$  and  $L3O_{(water)}$  for days when L3L and L3W are both present and the L3O surface index = "mixed". Squares = mean values; Red triangles = corresponding mean a priori values. Sample sizes are given below the top x axis.

Figure S7 corresponds to Figure 8, discussed in Section 3.2.1, but for selected other profile levels (Figure S7a-d) and for CO Total Column (TCO) (Figure S7e).



175

**Figure S7 (a)** Boxplots showing the seasonal distribution of retrieved 900 hPa level VMR values from L3W, L3L, L3O,  $L3O_{(mixed)}$  and  $L3O_{(water)}$ . Squares = mean values; Red triangles = corresponding mean a priori values. Sample sizes are given below the top x axis.



Figure S7 (b) As Figure S7a but for the 800 hPa level of the profile.



Figure S7 (c) As Figure S7a but for the 600 hPa level of the profile.



Figure S7 (d) As Figure S7a but for the 300 hPa level of the profile.



Figure S7 (e) As Figure S7a but for CO Total Column (TCO).

Table S8 and Figure S8 correspond respectively to Table 3 and Figure 9, discussed in Section 3.2.2. The difference here is that, while Table 3 and Figure 10 are from analysis that considers *all* data in the L3L, L3W

and L3O timeseries, Table S8 and Figure S8 only consist of retrievals made on days when L3L and L3W are both present and the L3O surface index is "mixed", i.e., only the days analysed in Section 3.1 (thus, there are no data for L3O<sub>(water)</sub> because there is only one L3O observation per day in the timeseries).

#### 205

Table S8 Results from WLS regression analysis of seasonal mean L3W, L3L, L3O, L3O<sub>(water)</sub> and L3O<sub>(mixed)</sub> timeseries for selected profile levels in DJF and JJA, only for days when L3L and L3W are both present and the L3O surface index is "mixed". Trend corresponds to the gradient of the WLS best-fit line; SE = standard error of trend; P-value = probability that the trend is zero; % change y<sup>-1</sup>= mean percentage change in retrieved CO per year, calculated from WLS regression model predicted values as follows:

% change y<sup>-1</sup> = { 
$$\left[ \left( \frac{\text{Predicted}_{\text{last}}}{\text{Predicted}_{\text{first}}} \right) * 100 \right] - 100$$
}/ny

where ny = number of years. The penultimate two columns correspond to the result of a significance test performed on the difference between that row's trend and the trend in L3L and L3W, respectively, as follows:

$$Z = \frac{Trend_1 - Trend_2}{\sqrt{SE_1^2 + SE_2^2}}$$

where SE<sub>1</sub> and SE<sub>2</sub> correspond to the standard errors of Trend<sub>1</sub> and Trend<sub>2</sub> respectively, and Z is the test statistic. Where Z is greater (less) than 1.645 (-1.645) the trend difference is statistically significant to at least 90 % (i.e. p < 0.1). Drift = the measurement drift values given in Deeter et al., (2017). No values are given for the 900 and 300 hPa levels of the profile: we therefore cite values for the 400 and 200 hPa levels to give context to the 300 hPa trends we show; and we expect that the 900 hPa level drift is somewhere between that of the surface and 800 hPa levels, which are both shown.

220

of the surface and 800 hPa levels, which are

<sup>†</sup> Units for TCO are mol cm<sup>-2</sup>

DJF	Level	Timeseries	Trend (ppbv)	Standard Error (ppbv)	P-value	% change y <sup>-1</sup>	Sig. di L3L	iff. to: L3W	Drift (% y⁻
nL3W = 18	Surface	L3W	-1.93	0.45	0.003	-0.99	No	n/a	
nL3L = 18		L3L	-0.945	0.42	0.055	-0.52	n/a	No	
nL3O = 18 nL3O <sub>(water)</sub> = 0		L3O L3O <sub>(water)</sub>	-1.21	0.33	0.007	-0.65	No	No	-0.69±0.1
nL3O <sub>(mixed)</sub> = 18		L3O <sub>(mixed)</sub>	-1.21	0.33	0.007	-0.65	No	No	
	900hPa	L3W	-2.29	0.52	0.002	-1.25	No	n/a	
		L3L	-2.69	0.57	0.001	-1.51	n/a	No	
		L30	-2.3	0.47	0.001	-1.29	No	No	Not given
		L3O <sub>(water)</sub>							nor given
		L3O <sub>(mixed)</sub>	-2.3	0.47	0.001	-1.29	No	No	
	800hPa	L3W	-1.95	0.31	0.000	-1.27	No	n/a	
		L3L	-2.84	0.57	0.001	-1.77	n/a	No	
		L3O L3O <sub>(water)</sub>	-3.11	0.51	0.000	-1.90	No	Yes	-1.04±0.1
		L3O <sub>(mixed)</sub>	-3.11	0.51	0.000	-1.90	No	Yes	
	600hPa	L3W	-1.07	0.26	0.004	-0.88	Yes	n/a	
		L3L	-2.64	0.63	0.003	-1.97	n/a	Yes	
		L30	-1.48	0.41	0.006	-1.20	No	No	-0.33±0.0
		L3O <sub>(water)</sub>							0.00 - 0.0
		L3O <sub>(mixed)</sub>	-1.48	0.41	0.006	-1.20	No	No	
	300hPa	L3W	0.861	0.35	0.039	1.12	No	n/a	
		L3L	-0.583	0.92	0.542	-0.64	n/a	No	400 hPa:
		L30	0.271	0.71	0.714	0.32	No	No	1.15 ± 0.1 200 hPa:
		L3O <sub>(water)</sub>							1.49±0.1
		L3O <sub>(mixed)</sub>	0.271	0.71	0.714	0.32	No	No	1119 - 011
	тсо	L3W	-2.88E+16	4.00E+15	0.000	-1.08	Yes	n/a	
		L3L	-1.45E+16	7.40E+15	0.085	-0.60	n/a	Yes	
		L3O L3O <sub>(water)</sub>	-3.24E+16	4.00E+15	0.000	-1.20	Yes	No	0.001±0.0
		L3O <sub>(water)</sub> L3O <sub>(mixed)</sub>	-3.24E+16	4.00E+15	0.000	-1.20	Yes	No	
JJA									
nL3W = 84	Surface	L3W	-0.72	0.38	0.084	-0.35	Yes	n/a	
nL3L = 84	oundee	L3L	-2.90	0.68	0.001	-1.46	n/a	Yes	
nL3O = 84		L30	-1.43	0.39	0.003	-0.72	Yes	No	-0.69±0.1
$nL3O_{(water)} = 0$ $nL3O_{(mixed)} = 84$		L3O <sub>(water)</sub> L3O <sub>(mixed)</sub>	-1.43	0.39	0.003	-0.72	Yes	No	
nL3O <sub>(water)</sub> = 0 nL3O <sub>(mixed)</sub> = 84		L3O <sub>(mixed)</sub>	-1.43	0.39	0.003	-0.72	Yes	No	
	900hPa	L3O <sub>(mixed)</sub> L3W	-1.67	0.44	0.002	-0.99	No	n/a	
	900hPa	L3O <sub>(mixed)</sub> L3W L3L	-1.67 -2.84	0.44 0.69	0.002 0.001	-0.99 -1.71	No n/a	n/a No	
	900hPa	L3O <sub>(mixed)</sub> L3W L3L L3O	-1.67	0.44	0.002	-0.99	No	n/a	Not giver
	900hPa	L3O <sub>(mixed)</sub> L3W L3L	-1.67 -2.84	0.44 0.69	0.002 0.001	-0.99 -1.71	No n/a	n/a No	Not giver
		L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub>	-1.67 -2.84 -1.97 -1.97	0.44 0.69 0.54 0.54	0.002 0.001 0.003 0.003	-0.99 -1.71 -1.19 -1.19	No n/a No No	n/a No No	Not giver
	900hPa 800hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W	-1.67 -2.84 -1.97 -1.97 -1.72	0.44 0.69 0.54 0.54 0.66	0.002 0.001 0.003 0.003 0.023	-0.99 -1.71 -1.19 -1.19 -1.24	No n/a No No	n/a No No No	Not giver
		L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97	0.44 0.69 0.54 0.54 0.66 0.60	0.002 0.001 0.003 0.003 0.023 0.006	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47	No n/a No No No n/a	n/a No No n/a No	Not given
		L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O	-1.67 -2.84 -1.97 -1.97 -1.72	0.44 0.69 0.54 0.54 0.66	0.002 0.001 0.003 0.003 0.023	-0.99 -1.71 -1.19 -1.19 -1.24	No n/a No No	n/a No No No	-
		L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97	0.44 0.69 0.54 0.54 0.66 0.60	0.002 0.001 0.003 0.003 0.023 0.006	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47	No n/a No No No n/a	n/a No No n/a No	-
	800hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub>	-1.67 -2.84 -1.97 -1.97 -1.72 -1.76 -1.76	0.44 0.69 0.54 0.54 0.66 0.60 0.56	0.002 0.001 0.003 0.003 0.023 0.006 0.008	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29	No n/a No No n/a No	n/a No No n/a No No	-
		L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W	-1.67 -2.84 -1.97 -1.97 -1.72 -1.76 -1.76 -1.76 -1.15	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63	0.002 0.001 0.003 0.003 0.023 0.006 0.008 0.008 0.008	-0.99 -1.71 -1.19 -1.19 -1.24 -1.27 -1.29 -1.29 -1.04	No n/a No No n/a No No	n/a No No n/a No No	-
	800hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O	-1.67 -2.84 -1.97 -1.97 -1.72 -1.76 -1.76	0.44 0.69 0.54 0.54 0.66 0.60 0.56	0.002 0.001 0.003 0.003 0.023 0.006 0.008	-0.99 -1.71 -1.19 -1.19 -1.24 -1.27 -1.29 -1.29	No n/a No No n/a No	n/a No No n/a No No	-1.04±0.1
	800hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub>	-1.67 -2.84 -1.97 -1.97 -1.72 -1.76 -1.76 -1.15 -0.72 -0.90	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.008 0.008 0.090 0.166 0.102	-0.99 -1.71 -1.19 -1.19 -1.24 -1.27 -1.29 -1.29 -1.04 -0.69 -0.84	No n/a No No n/a No No No No	n/a No No n/a No No No No	-1.04±0.1
	800hPa 600hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(water)</sub> L3O <sub>(mixed)</sub>	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.008 0.090 0.166 0.102	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84	No n/a No No No No No No	n/a No No No No No No	Not given -1.04 ±0.1 -0.33 ±0.0
	800hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3W	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.090 0.166 0.102 0.102 0.102	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 -0.84 2.01	No n/a No No No No No No No	n/a No No No No No No No	-1.04±0.1 -0.33±0.0
	800hPa 600hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3W L3L	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86 2.90	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55 1.20	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.090 0.166 0.102 0.102 0.102	-0.99 -1.71 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 2.01 3.48	No n/a No No No No No No No No	n/a No No No No No No No No	-1.04±0.1
	800hPa 600hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3D L3O <sub>(mixed)</sub> L3W L3L L3O	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.090 0.166 0.102 0.102 0.102	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 -0.84 2.01	No n/a No No No No No No No	n/a No No No No No No No	-1.04 ±0.2 -0.33 ±0.0 400 hPa 1.15 ±0.1 200 hPa
	800hPa 600hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3O <sub>(mixed)</sub> L3W L3L	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86 2.90	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55 1.20	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.090 0.166 0.102 0.102 0.102	-0.99 -1.71 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 2.01 3.48	No n/a No No No No No No No No	n/a No No No No No No No No	-1.04 ±0.2 -0.33 ±0.0 400 hPa 1.15 ±0.1 200 hPa
	800hPa 600hPa 300hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3W L3L L3O L3O <sub>(water)</sub>	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 1.86 2.90 2.18 2.18	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55 1.20 0.65	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.008 0.008 0.102 0.102 0.102 0.102 0.005 0.027 0.005 0.005	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 2.01 3.48 2.44 2.44	No n/a No No No No No No No No No	n/a No No No No No No No No No	-1.04 ±0.1 -0.33 ±0.0 400 hPa 1.15 ±0.1 200 hPa
	800hPa 600hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3U L3D L3O <sub>(water)</sub> L3W	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86 2.90 2.18 2.18 2.18 -1.64E+16	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55 1.20 0.65 0.65 5.50E+15	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.008 0.008 0.102 0.102 0.102 0.005 0.027 0.005 0.005 0.005	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 2.01 3.48 2.44 2.44 2.44 -0.64	No n/a No No No No No No No No No No	n/a No No No No No No No No No No	-1.04 ±0.1 -0.33 ±0.0 400 hPa 1.15 ±0.1 200 hPa
	800hPa 600hPa 300hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3W L3L L3O L3O <sub>(water)</sub>	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86 2.90 2.18 2.18 2.18 -1.64E+16 -1.64E+16	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.56 0.63 0.49 0.51 0.51 0.55 1.20 0.65 0.65 5.50E+15 1.20E+16	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.008 0.008 0.102 0.102 0.102 0.102 0.005 0.027 0.005 0.005 0.005	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 2.01 3.48 2.44 2.44 2.44 -0.64 -0.69	No n/a No No No n/a No No No No No No No	n/a No No No No No No No No No No	-1.04±0.1 -0.33±0.0 400 hPa 1.15±0.1 200 hPa 1.49±0.1
	800hPa 600hPa 300hPa	L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3O <sub>(mixed)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3W L3L L3O L3O <sub>(water)</sub> L3W	-1.67 -2.84 -1.97 -1.97 -1.72 -1.97 -1.76 -1.76 -1.76 -1.15 -0.72 -0.90 -0.90 1.86 2.90 2.18 2.18 2.18 -1.64E+16	0.44 0.69 0.54 0.54 0.66 0.60 0.56 0.56 0.63 0.49 0.51 0.51 0.55 1.20 0.65 0.65 5.50E+15	0.002 0.001 0.003 0.023 0.006 0.008 0.008 0.008 0.008 0.102 0.102 0.102 0.005 0.027 0.005 0.005 0.005	-0.99 -1.71 -1.19 -1.19 -1.24 -1.47 -1.29 -1.29 -1.29 -1.04 -0.69 -0.84 2.01 3.48 2.44 2.44 2.44 -0.64	No n/a No No No No No No No No No No	n/a No No No No No No No No No No	-1.04 ±0.1 -0.33 ±0.0



Figure S8 WLS regression best-fit lines calculated from seasonal mean retrieved surface VMR timeseries in 230 DJF (top row) and JJA (bottom row), only for days when L3L and L3W are both present and the L3O surface index is "mixed". Left column = L3L (green) and L3W (blue); Right column = L3O (black), L3O<sub>(mixed)</sub> (brown) and L3O(water) (blue). The daily observations corresponding to each seasonal mean value are represented by colour-coded boxplots each year, and the seasonal mean value is represented by filled squares. The dashed red line is the mean of the corresponding seasonal mean a priori data from each of the timeseries in the respective panel. Colour-coded values below the top x axis correspond to the number of observations 235 each season. Values in the legend are the value, standard error, and probability of zero value of the trend, respectively.