

Figure S1. Measured $\mathrm{O}_{3}$ (top) and RH (bottom) daily values at PRS (grey lines). Red (blue) dots indicate measurement periods identified as SI by the "Cui09" ("Trickl14") criterion.


Figure S2. Measured $\mathrm{O}_{3}$ (top) and RH (bottom) daily values at CUR (grey lines). Red (blue) dots indicate measurement periods identified as SI by the "Cui09" ("Trickl14") criterion.


Figure S3. Left column: average seasonal cycle of experimental SI detection ("Cui09": magenta, "Trick114": blue, "All detections": green) at PRS (2012-2016). Right column: average seasonal cycle of SI detection by STEFLUX for the original $3^{\circ} \times 3^{\circ}$ target box (blue area) and for the $1^{\circ} \times 1^{\circ}$ target box (white line).

Table S1. Agreement in percentage between in-situ detections (considering both "Cui09" and "Trickl14" criterions) and STEFLUX outputs, as a function of the horizontal extension of the box. For each station, the top lid pressure level used for the STEFLUX configuration is reported. SI events were defined according to Section 3.1 of the main manuscript.

| Horizontal Box Extension | PRS @ 660 hPa | CMN @ 790 hPa | CUR @ 820 hPa |
| :---: | :---: | :---: | :---: |
| $1^{\circ} \times 1^{\circ}$ | $14 \%$ | $5 \%$ | $7 \%$ |
| $2^{\circ} \times 2^{\circ}$ | $27 \%$ | $10 \%$ | $11 \%$ |
| $3^{\circ} \times 3^{\circ}$ | $38 \%$ | $19 \%$ | $11 \%$ |

Table S2. Overall trend estimates for the four measurement sites, obtained by considering seasonal SI frequency values. Bold indicates the SI frequency values calculated considering only "long" SI events (i.e., longer than three days). The significant trends ( $p<0.1$ ) are denoted by " + ".

| Season | CMN | PRS | CMP | CUR |
| :---: | :---: | :---: | :---: | :---: |
| DJF | 0.12 [-0.10, 0.28] | 0.04 [-0.28, 0.44] | 0.06 [-0.14, 0.35] | 0 [-0.12, 0.13] |
|  | 0.06 [-0.16, 0.27] | -0.11 [-0.52, 0.53] | 0.06 [-0.19, 0.33] | 0 [ $0.16,0.11$ ] |
| MAM | 0.10 [-0.07, 0.24] | 0.18 [-0.09, 0.38] | 0.16 [0, 0.36] + | 0.09 [-0.04, 0.21] |
|  | 0.08 [0, 0.25] | 0.31 [-0.06, 0.66] + | 0.15 [-0.08, 0.36] | 0.08 [0, 0.24] |
| JJA | 0 [0, 0.04] | -0.11 [-0.25, 0] | 0 [0, 0.06] | 0 [ $-0.05,0$ ] |
|  | 0 [0, 0] | 0 [-0.11, 0] | 0 [0, 0] | 0 [0, 0] |
| SON | 0.04 [-0.09, 0.2] | 0.13 [0, 0.31] | 0.08 [-0.06, 0.22] | 0 [-0.09, 0.1] |
|  | 0 [-0.1, 0.05] | 0.10 [-0.13, 0.37] | 0.07 [0, 0.21] | 0 [0, 0] |

