

Supporting Information of:

**Exploring the influence of land use on the urban carbonyl sulfide budget: a case study of the
Metropolitan Area of Barcelona**

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Table S1. Table with values of OCS (ppt) at every date, location and group in campaign May and October 2020: Wind direction in degrees; Wind speed m/s; temperature in °C, and Pressure is QFE in hPa; relative humidity (RH) is in %; PBLH is extracted from WRF model and is in m.

| Date m-d | Location | Group (Altitude) | Lat,lon | Hour UTC | OCS ppt | WRF PBLH | WRF temp | Wind direction | Wind Speed | Air temperature | Air pressure | RH |
|-------------|-----------|---------------------|------------------|-------------|-------------|-------------|-------------|-------------------|---------------|--------------------|-----------------|----|
| 05-18 | Gavà | Downwind (2) | 41.268, 2.032 | 7:00 | 407.1±8.5 | 534 | 19.45 | 2 | 0.87 | 23.4 | 1019 | - |
| | | | | 9:30 | 600.9±30.3 | 1380 | 23.10 | 180 | 2.29 | 22.8 | 1019 | - |
| | | | | 11:00 | 477.5±17.9 | 816 | 21.64 | 140 | 1.15 | 26.2 | 1019 | - |
| | | Target (1) | 41.279, 2.010 | 7:00 | 432.7±13.3 | 577 | 19.69 | - | 0.00 | 21.2 | 1018 | - |
| | | | | 9:30 | 522.9±15.7 | 1498 | 23.49 | 180 | 1.88 | 23.8 | 1019 | - |
| | | southern (-2) | 41.285, 2.002 | 7:00 | 452.9±9.3 | 592 | 19.81 | 225 | 0.73 | 23.1 | 1018 | - |
| | | | | 9:30 | NA | 1578 | 23.66 | 215 | 2.43 | 27.4 | 1018 | - |
| | Tibidabo | Downwind (442) | 41.418, 2.116 | 4:00 | 443.3±4.4 | 18 | 13.59 | - | 0.00 | 13.7 | 966 | - |
| 05-19 | Tibidabo | southern (442) | 41.418, 2.116 | 7:00 | 491.9±12.8 | 19 | 17.39 | 340 | 2.34 | 18.9 | 964 | - |
| | | | | 9:30 | 482.0±30.3 | 224 | 20.84 | 12 | 1.17 | 23.9 | 964 | - |
| | | Target (320) | 41.435, 2.087 | 7:00 | 620.2±260.2 | 193 | 17.74 | 270 | 0.92 | 17.3 | 978 | - |
| | | | | 9:30 | 499.4±16.1 | 406 | 21.59 | 280 | 0.04 | 24.5 | 977 | - |
| | | Downwind (175) | 41.457, 2.065 | 7:00 | 463.4±1.3 | 210 | 18.19 | 275 | 1.43 | 19.9 | 994 | - |
| | | | | 9:30 | 499.8±NA | 441 | 21.98 | 270 | 1.52 | 29.6 | 994 | - |
| 05-20 | Montjuic | southern (58) | 41.369, 2.171 | 7:00 | 471.1±12.6 | 327 | 21.93 | - | 0.00 | 22.02 | 1007 | - |
| | | | | 9:30 | 496.1±0.9 | 567 | 23.20 | - | 0.00 | 22.5 | 1007 | - |
| | | Target (98) | 41.367, 2.165 | 7:00 | 483.9±2.3 | 327 | 21.93 | - | 0.00 | 19.7 | 1001 | - |
| | | | | 9:30 | 484.3±0.4 | 567 | 23.20 | 185 | 0.32 | 25.5 | 1001 | - |
| | | Downwind (42) | 41.369, 2.156 | 7:00 | 500.1±5.5 | 430 | 22.51 | - | 0.00 | 22.6 | 1008 | - |
| | | | | 9:30 | 489.7±1.1 | 883 | 24.96 | 80 | 1.34 | 29.9 | 1008 | - |
| 05-21 | Poble Nou | southern (4) | 41.395, 2.206 | 7:00 | 451.8±0.3 | 44 | 21.65 | 180 | 0.56 | 26.4 | 1016 | - |
| | | Target (0) | 41.400, 2.199 | 7:00 | 448.1±7.9 | 142 | 23.27 | 130 | 0.28 | 23.1 | 1016 | - |

| | | | | | | | | | | | | |
|-------|--------------------|-------------------|-------------------|-------|------------|------|-------|-----|------|------|------|---|
| | Gavà | Downwind (4) | 41.403, 2.192 | 7:00 | 484.6±22.6 | 234 | 24.17 | - | 0.00 | 27.2 | 1016 | - |
| | | southern (2) | 41.268, 2.032 | 4:00 | 482.1±NA | 11 | 18.43 | 347 | 1.05 | 16.2 | 1016 | - |
| | | | | 11:00 | 495.5±10.7 | 249 | 23.97 | 240 | 3.40 | 31.1 | 1017 | - |
| | Tibidabo | southern (442) | 41.418, 2.116 | 4:00 | 472.7±2.6 | 9 | 19.42 | 347 | 0.94 | 19.9 | 965 | - |
| 05-25 | Prat | southern (0) | 41.315, 2.063 | 7:00 | 509.2±16.8 | 737 | 21.70 | 170 | 0.30 | 26.7 | 1025 | - |
| | | | | 9:30 | 501.7±12.4 | 1084 | 23.23 | 180 | 3.57 | 25.1 | 1025 | - |
| | | Target (0) | 41.318, 2.057 | 7:00 | 521.9±30.2 | 736 | 21.62 | 230 | 1.14 | 26.9 | 1024 | - |
| | | | | 9:30 | 528.9±37.9 | 1085 | 23.27 | 180 | 3.29 | 26.5 | 1024 | - |
| | Gavà | Downwind (2) | 41.326, 2.051 | 7:00 | 506.4±2.5 | 827 | 21.87 | 167 | 0.88 | 27.6 | 1024 | - |
| | | | | 9:30 | 657.6±57.3 | 1182 | 24.35 | 178 | 2.07 | 32.6 | 1024 | - |
| | | southern (2) | 41.268, 2.032 | 4:00 | 486.0±31.8 | 9 | 16.91 | 20 | 0.00 | 19.8 | 1024 | - |
| | | | | 11:00 | 495.8±4.4 | 960 | 22.76 | 240 | 3.50 | 28 | 1025 | - |
| | Tibidabo | southern (442) | 41.418, 2.116 | 4:00 | 496.4±7.9 | 27 | 16.73 | - | 0.00 | 21.5 | 972 | - |
| | | | | 11:00 | 507.9±2.2 | 958 | 21.47 | 252 | 1.20 | 29.6 | 973 | - |
| 05-26 | Sagrada Familia | southern (37) | 41.395, 2.181 | 7:00 | 462.4±1.0 | 771 | 23.21 | 240 | 0.00 | 22.5 | 1023 | - |
| | | Target (54) | 41.401, 2.174 | 7:00 | 452.3±7.6 | 806 | 23.21 | 140 | 0.69 | 24.2 | 1020 | - |
| | | Downwind (55) | 41.404, 2.169 | 7:00 | 464.9±20.6 | 806 | 23.21 | - | 0.00 | 24.2 | 1018 | - |
| 05-27 | Collserola | southern (155) | 41.434, 2.140 | 7:00 | 468.9±22.8 | 441 | 20.96 | 80 | 0.00 | 24.4 | 1003 | - |
| | | | | 9:30 | 493.1±NA | 1079 | 22.96 | - | 0.00 | 23.4 | 1004 | - |
| | | Target (107) | 41.459, 2.2.12 | 7:00 | 412.6±13.8 | 395 | 20.28 | - | 0.72 | 19.4 | 1006 | - |
| | | | | 9:30 | 498.3±12.6 | 1133 | 23.24 | 355 | 1.94 | 24 | 1006 | - |
| | | Downwind (442) | 41.435, 2.087 | 7:00 | 451.0±37.4 | 289 | 19.82 | - | 0.00 | 23.7 | 1011 | - |
| | | | | 9:30 | 499.5±4.2 | 1076 | 23.06 | 120 | 3.62 | 29.7 | 1012 | - |
| 05-28 | Guinardó | southern (115) | 41.419, 2.171 | 7:00 | 495.0±8.7 | 590 | 21.30 | 180 | 0.00 | 21 | 1011 | - |
| | | | | 9:30 | 594.3±5.6 | 1524 | 24.52 | 80 | 0.31 | 25.8 | 1010 | - |

| | | | | | | | | | | | | |
|-------|-----------|-------------------|------------------|--------------|-------------------------|-------------|----------------|------------|--------------|--------------|--------------|--------|
| 10-13 | Gavà | Target (169) | 41.420, 2.168 | 7:00 9:30 | 512.2±8.2 612.8±13.7 | 590 1524 | 21.30 24.52 | 140 160 | 0.00 1.60 | 21.3 22.6 | 1003 1002 | - - |
| | | Downwind (127) | 41.422, 2.166 | 7:00 9:30 | 506.7±2.6 628.8±20.5 | 590 1524 | 21.30 24.52 | - 272 | 0.00 1.36 | 24.5 26.1 | 1008 1007 | - - |
| | | southern (2) | 41.268, 2.032 | 4:00 | 400.5±3.0 | 31 | 14.31 | 5 | 0.83 | 12.9 | 1011 | 75 |
| | | | | 7:00 | 398.3±6.1 | 62 | 14.12 | 5 | 1.88 | 13 | 1011 | 60 |
| | | | | 9:30 | 410.8±6.0 | 846 | 18.35 | 25 | 2.00 | 17.8 | 1011 | 49 |
| | | | | 11:00 | 420.1±5.8 | 1004 | 19.84 | 10 | 0.77 | 22.6 | 1010 | 41 |
| | Poble Nou | southern (4) | 41.395, 2.206 | 4:00 | 405.3±5.8 | 181 | 17.36 | 310 | 2.00 | 15.4 | 1011 | 49 |
| | | | | 7:00 | 447.6±3.1 | 210 | 17.45 | 340 | 1.31 | 17.1 | 1011 | 50 |
| | | | | 9:30 | 392.9±8.8 | 639 | 18.46 | 340 | 2.35 | 19.3 | 1011 | 42 |
| | | | | 11:00 | 426.1±19.8 | 1202 | 19.97 | 60 | 1.70 | 21.5 | 1011 | 52 |
| | Tibidabo | southern (442) | 41.418, 2.116 | 4:00 | 392.1±5.5 | 23 | 10.36 | 355 | 1.40 | 8.4 | 959 | 70 |
| | | | | 7:00 | 412.2±10.2 | 69 | 11.07 | 350 | 2.44 | 11.3 | 954 | 57 |
| | | | | 9:30 | 404.5±5.8 | 302 | 13.56 | 340 | 0.04 | 17.9 | 960 | 40 |
| | | | | 11:00 | 416.9±12.9 | 879 | 17.30 | 298 | 0.70 | 22.3 | 959 | 39 |
| 10-15 | Poble Nou | southern (4) | 41.395, 2.206 | 7:00 | 390.4±15.7 | 287 | 15.61 | 310 | 0.00 | 11.8 | 1010 | 59 |
| | | | | 9:30 | 407.9±2.4 | 525 | 16.82 | 360 | 2.48 | 17.5 | 1011 | - |
| | | Target (0) | 41.400, 2.199 | 7:00 | 391.6±3.1 | 226 | 12.42 | 265 | 0.51 | 11 | 1012 | 55 |
| | | | | 9:30 | 391.1±2.9 | 530 | 14.96 | 90 | 0.46 | 18.8 | 1010 | 37 |
| | | Downwind (4) | 41.403, 2.192 | 7:00 | 432.1±3.9 | 199 | 12.50 | 56 | 0.00 | 13.1 | 1010 | 63 |
| | | | | 9:30 | 477.6±19.5 | 571 | 15.08 | 260 | 0.45 | 19.4 | 1010 | 50 |
| 10-19 | Guinardó | southern (115) | 41.419, 2.171 | 7:00 | 374.1±8.6 | 69 | 14.82 | 340 | 0.00 | 14.8 | 1007 | 82 |
| | | | | 9:30 | 392.4±9.8 | 934 | 18.23 | 340 | 0.00 | 17.7 | 1008 | 66 |
| | | Target (169) | 41.420, 2.168 | 7:00 | 403.4±7.2 | 69 | 14.82 | - | 0.00 | 15 | 999 | 75 |
| | | | | 9:30 | 375.6±NA | 934 | 18.23 | 330 | 0.56 | 17.6 | 1000 | 66 |
| | | Downwind (127) | 41.422, 2.166 | 7:00 | 520.8±112.8 | 69 | 14.82 | - | 0.00 | 13 | 1003 | 75 |
| | | | | 9:30 | 409.1±7.5 | 934 | 18.23 | - | 0.00 | 17.6 | 1004 | 66 |
| 10-20 | Sagrada | southern (37) | 41.395, 2.181 | 7:00 | 397.3±5.2 | 29 | 17.60 | - | 0.00 | 16.7 | 1017 | 77 |
| | | | | 9:30 | 442.5±6.3 | 1084 | 19.91 | 120 | 1.08 | 20.5 | 1017 | 69 |
| | | Target | 41.401, | 7:00 | 414.3±19.9 | 32 | 17.03 | 190 | 0.11 | 17 | 1014 | 78 |

| | | | | | | | | | | | | | | | |
|----------|----------|----------|----------|----------|-----------|-------------|-----------|------------|-------|-------|------|------|------|------|----|
| 10-21 | Montjuic | | (54) | 2.174 | 9:30 | 426.0±3.0 | 1085 | 19.66 | 260 | 0.38 | 21.6 | 1014 | 64 | | |
| | | Downwind | 41.404, | | 7:00 | 567.5±1.5 | 32 | 17.03 | - | 0.00 | 17.6 | 1012 | 77 | | |
| | | | (55) | 2.169 | 9:30 | 464.9±22.7 | 1085 | 19.66 | 180 | 0.50 | 20.9 | 1014 | 66 | | |
| | | southern | 41.369, | | 7:00 | 665.1±54.5 | 92 | 20.87 | - | 0.00 | 19.7 | 1004 | 85 | | |
| | | | (58) | 2.171 | 9:30 | 550.3±71.1 | 561 | 22.10 | 350 | 0.00 | 21.1 | 1004 | 77 | | |
| | | Target | 41.367, | | 7:00 | 423.1±10.9 | 92 | 20.87 | - | 0.00 | 18.9 | 998 | 91 | | |
| | | | (98) | 2.165 | 9:30 | 425.0±10.7 | 561 | 22.10 | 260 | 0.00 | 23.3 | 998 | 70 | | |
| | | Downwind | 41.369, | | 7:00 | 546.4±12.3 | 90 | 20.82 | - | 0.00 | 19.3 | 1005 | 91 | | |
| 10-22 | Prat | | (42) | 2.156 | 9:30 | 635.3±164.6 | 734 | 22.45 | - | 0.00 | 23.5 | 1005 | 72 | | |
| | | southern | 41.315, | | 7:00 | 477.4±20.7 | 108 | 19.76 | 350 | 1.34 | 18.5 | 1014 | 87 | | |
| | | | (0) | 2.063 | 9:30 | 454.0±2.8 | 435 | 21.52 | 345 | 1.07 | 21.6 | 1014 | 70 | | |
| | | Target | 41.318, | | 7:00 | 440.6±5.6 | 98 | 19.74 | 340 | 0.58 | 18.4 | 1012 | 86 | | |
| | | | (0) | 2.057 | 9:30 | 529.3±8.5 | 458 | 21.59 | 40 | 0.30 | 20.8 | 1013 | 71 | | |
| | | Downwind | 41.326, | | 7:00 | 487.3±8.5 | 103 | 19.66 | 325 | 1.95 | 18.8 | 1013 | 82 | | |
| | | | (2) | 2.051 | 9:30 | 486.5±9.2 | 495 | 21.69 | 330 | 1.12 | 21 | 1013 | 70 | | |
| | | 10-23 | Gavà | southern | 41.268, | | 7:00 | 489.3±24.9 | 84 | 18.79 | 240 | 1.74 | 18.9 | 1016 | 87 |
| | (2) | | | 2.032 | 9:30 | 456.2±0.5 | 380 | 20.27 | 210 | 1.85 | 20.8 | 1016 | 84 | | |
| Target | 41.279, | | | | 7:00 | 460.1±9.7 | 21 | 18.38 | 250 | 1.51 | 17.9 | 1015 | 92 | | |
| | (0) | | | 2.010 | 9:30 | 462.8±6.1 | 427 | 20.54 | 230 | 2.00 | 22.3 | 1015 | 77 | | |
| Downwind | 41.285, | | | | 7:00 | 447.0±15.6 | 38 | 18.53 | 240 | 0.81 | 20.1 | 1015 | 78 | | |
| | (-2) | | | 2.002 | 9:30 | 461.7±8.5 | 519 | 20.96 | 230 | 1.86 | 21.4 | 1016 | 66 | | |
| 10-27 | Gavà | | | southern | 41.268, | | 4:00 | 396.6±6.8 | 13 | 14.03 | 0.5 | 0.45 | 9.8 | 1012 | 64 |
| | | | | | (2) | 2.032 | 7:00 | 399.8±4.1 | 89 | 12.21 | 0 | 0.14 | 10.9 | 1014 | 64 |
| | | | | | 9:30 | 393.9±11.8 | 805 | 15.62 | 120 | 0.80 | 16.2 | 1014 | 50 | | |
| | | | | | 11:00 | 396.3±7.7 | 1291 | 18.44 | 120 | 1.55 | 21.6 | 1015 | 45 | | |
| | | Poble | southern | 41.395, | | 4:00 | 402.8±NA | 472 | 16.04 | 320 | 0.00 | 10.7 | 1013 | 60 | |
| | | | | (4) | 2.206 | 7:00 | 400.9±8.8 | 216 | 15.05 | 270 | 0.10 | 13 | 1015 | 52 | |
| | | | | | | 9:30 | 407.5±9.8 | 633 | 16.57 | 320 | 1.71 | 17.3 | 1016 | 42 | |
| | | | | | | 11:00 | 406.1±7.4 | 908 | 17.94 | 270 | 0.72 | 21.6 | 1015 | 43 | |
| Tibidabo | southern | 41.418, | | 4:00 | 406.3±3.7 | 180 | 10.02 | 0 | 5.11 | 7.9 | 960 | 63 | | | |
| | | (442) | 2.116 | 7:00 | 407.1±1.7 | 139 | 9.423 | 100 | 1.32 | 8.9 | 961 | 63 | | | |

| | | | | | | | | | |
|--|-------|-----------|-----|-------|-----|------|------|-----|----|
| | 9:30 | NA | 172 | 11.03 | 20 | 1.19 | 11.3 | 963 | 57 |
| | 11:00 | 413.3±0.3 | 343 | 13.21 | 255 | 1.26 | 18.7 | 963 | 38 |

Table S2. Summary of climatic conditions during the days of both campaigns. Psfc, T(°C) and Accumulated Precipitation (mm) obtained from the Meteorological Service of Catalonia.

May 2020 Campaign

| Dates (2020) | Psfc (hPa) | T (°C) | Precipitation accumulated (mm) | CWTs | Synoptic Wind component |
|--------------|------------|--------|--------------------------------|------|-------------------------|
| 05-18 | 1019.2 | 19.75 | 0.0 | CNE | NE |
| 05-19 | 1015.73 | 21.43 | 0.0 | C | N |
| 05-20 | 1015.17 | 21.45 | 0.0 | C | N |
| 05-21 | 1018.27 | 23.63 | 0.0 | U | N |
| 05-25 | 1025.6 | 20.93 | 0.0 | E | E |
| 05-26 | 1024.97 | 22.23 | 0.0 | E | E |
| 05-27 | 1026.1 | 21.08 | 0.0 | E | E |
| 05-28 | 1023.7 | 20.5 | 0.0 | A | SE |
| 06-01 | 1015.5 | 20.56 | 2.6 | U | SE |
| 06-15 | 1019.03 | 20.98 | 0.0 | U | NW |

October 2020 Campaign

| Dates (2020) | Psfc (hPa) | T (°C) | Precipitation accumulated (mm) | CWTs | Synoptic Wind component |
|--------------|------------|--------|--------------------------------|------|-------------------------|
| 10-13 | 1012.7 | 16.3 | 0.0 | C | NW |
| 10-15 | 1013.0 | 13.5 | 0.0 | CN | N |
| 10-19 | 1021.2 | 16.1 | 0.0 | ASW | SW |
| 10-20 | 1017.1 | 18.5 | 0.0 | SW | SW |
| 10-21 | 1012.4 | 20.2 | 0.0 | CSW | SW |
| 10-22 | 1015.2 | 19.2 | 1.3 | SW | SW |
| 10-23 | 1017.7 | 18.9 | 0.3 | U | W |
| 10-27 | 1016.2 | 14.3 | 0.0 | ANW | NW |

Method description

Objective and automatic classifications are based on the application of algorithms that use ratios derived from atmospheric pressure fields and allow objective comparisons to be made. Jenkinson and Collison (JC) classification (Jenkinson and Collison, 1977), which was adapted from the Lamb catalogue and uses surface pressure data. This classification has been adapted to many regions and is one of the most commonly used (e.g. Jones et al., 1993; Trigo and DaCamara, 2000; Cortesi et al., 2014). JC classification has the advantage of being a universal and standardized method that allows comparison between different regions. JC method was included in the framework of COST7333 Action, which involved researchers from 23 countries working to find the most suitable automatic classifications in Europe (Huth et al., 2008). The JC classification is an objective scheme based on 9 daily grid-point mean sea-level pressure data. It

is an extension of the Lamb classification from 10 to 27 circulation weather types (CWTs). The method was adapted to the Iberian Peninsula in accordance with the proposal by Spellman (2000) and moved 5° to the east (Gilabert and Llasat, 2017). The CWTs are calculated using the surface pressure of the NCEP/NCAR reanalysis dataset.

CWTs: Cyclonic (C), Anticyclonic (A), pure advectives (N, S, E, W, NE, SE, NW, SW), hybrid cyclone- advectives (CN, CNE, CE, CSE, CS, CSW, CW, CNW), hybrid anticyclone-advectives (AN, ANE, AE, ASE, AS, ASW, AW, ANW).

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Table S3. Model characteristics and experiment configurations.

| Resolution and initial conditions | |
|-----------------------------------|--|
| Horizontal resolution | 9 km x 9 km; 3 km x 3 km; 1 km x 1 km |
| Domain dimensions | 150 x 145; 118 x 118; 121 x 121 |
| Vertical layers | 57 (16 between the surface and 100 m) |
| Top of the atmosphere | 50 hPa |
| Initial conditions | ERA5 (C3S 2017) with 31 km horizontal resolution, 137 vertical levels and 6-hour separation |
| Physics parameterizations | |
| Microphysics | WRF Single-Moment 6-class scheme (Hong and Lim 2006) |
| Shortwave and longwave radiation | RRTMG scheme (Iacono et al. 2008) |
| Cumulus | Kain-Fritsch scheme (Kain and Fritsch 2004) (only the outermost domain) |
| PBL Scheme | Bougeault (Bougeault and Lacarrere 1989) |
| Surface / UCP | Noah Land Surface Model (pervious areas) (Chen and Dudhia 2001) / BEP+BEM (impervious areas) (Salamanca and Martilli 2010) |
| Surface layer | Monin-Obukhov Eta similarity scheme with Zilitinkevich thermal roughness length |

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Table S4. Validation and statistical analyses of the root-mean-square error (RMSE), mean bias (MB) and correlation factor (R).

| Campaign | Parameter | root-mean-square error (RMSE) | mean bias (MB) | r |
|----------|-----------|-------------------------------|----------------|------|
| May | RH | 13.17% | -6.19% | 0.74 |
| May | T | 1.74°C | 0.44°C | 0.91 |
| May | WS | 1.35ms-1 | 0.13ms-1 | 0.59 |
| October | RH r | 14.13% | 7.72% | 0.80 |
| October | T | 1.94°C | 1.16°C | 0.94 |
| October | WS | 1.65 ms-1 | 0.45 ms-1 | 0.52 |

$$RMSE = \left(\frac{1}{N} \sum_{i=1}^N (M_i - O_i)^2 \right)^{1/2}$$

$$MB = \frac{1}{N} \sum_{i=1}^N (M_i - O_i)$$

$$R = \frac{\sum_{i=1}^N (M_i - \bar{M})(O_i - \bar{O})}{\left(\sum_{i=1}^N (M_i - \bar{M})^2 \right)^{1/2} \left(\sum_{i=1}^N (O_i - \bar{O})^2 \right)^{1/2}}$$

Where M_i represents the modelled value and O_i the observed value at each time step. The overbar represents the mean between all the available time steps (N) for all meteorological station.

Figure S1. Plot of the Zumkehr et al. (2018) gridded anthropogenic emissions inventory of OCS in Catalonia (left panel) and Barcelona metropolitan area (right panel). Below are the histograms with the emission values of the different sources for Barcelona and the rest of Europe according to Zumkehr et al. (2018).

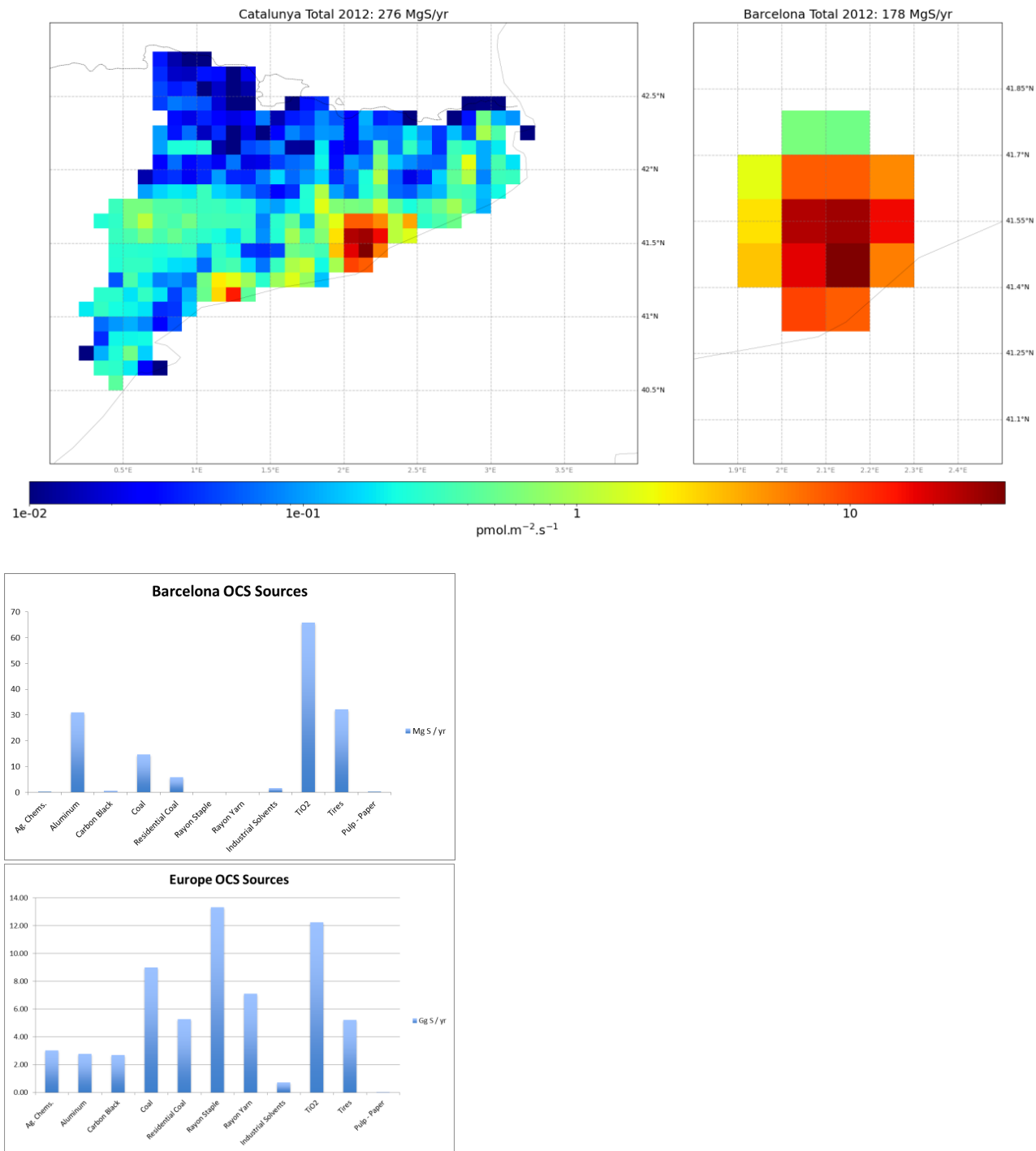


Figure S2. Domains of the WRF model for this study with 9, 3 and 1km grid size for d01, d02 and d03, respectively.

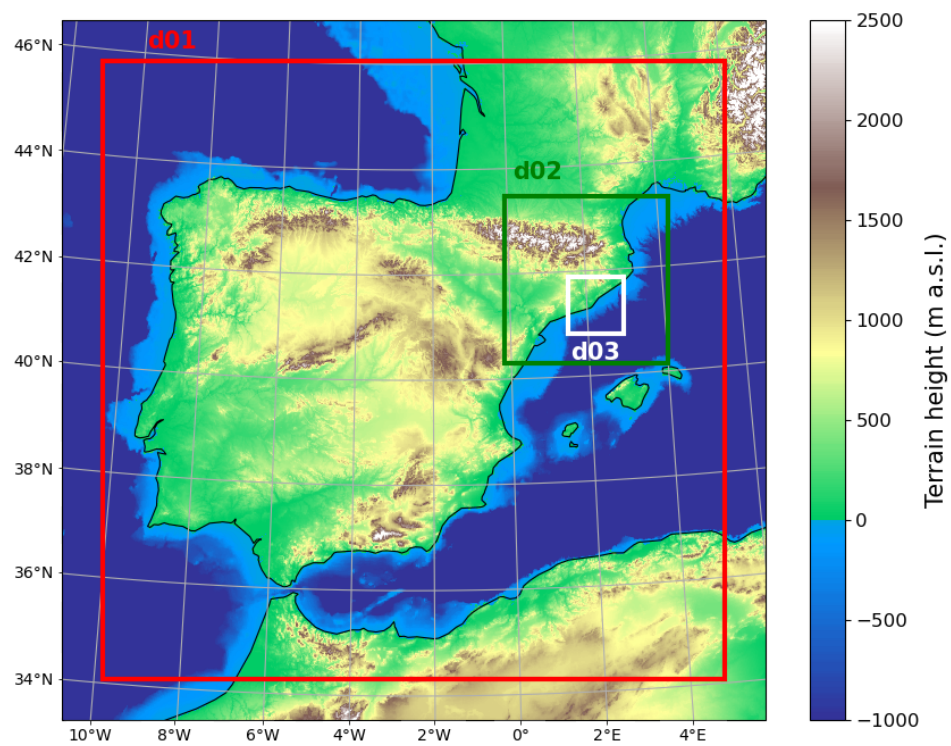


Figure S3 Relative humidity (RH), temperature (T) and wind speed (WD) analysis for the validation of WRF simulations. Root mean square error (RMSE), Mean bias and correlation are shown for each station during the campaign in May.

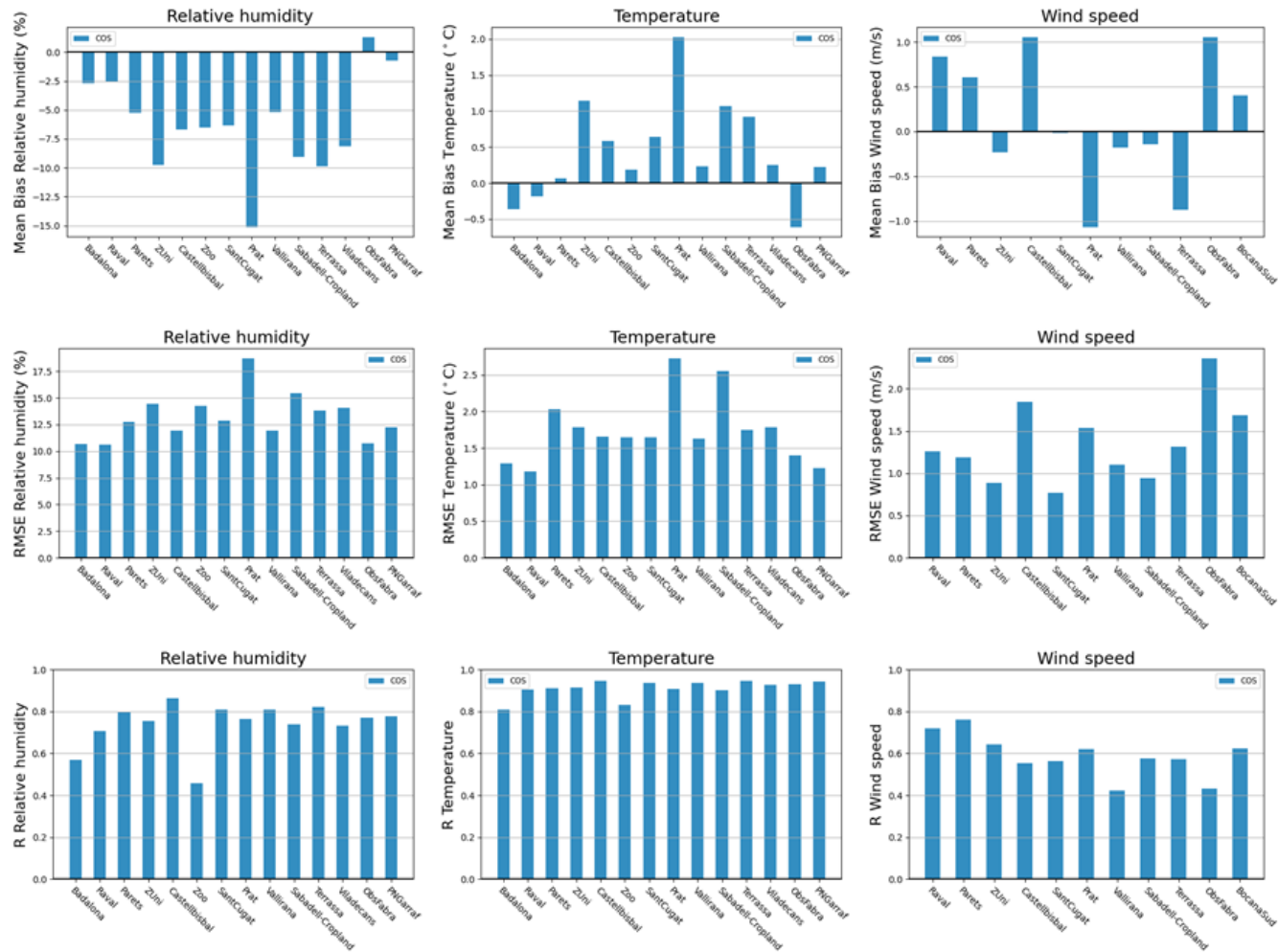


Figure S4 Relative humidity (RH), temperature (T) and wind speed (WD) analysis for the validation of WRF simulations. Root mean square error (RMSE), Mean bias and correlation are shown for each station during the campaign in October.

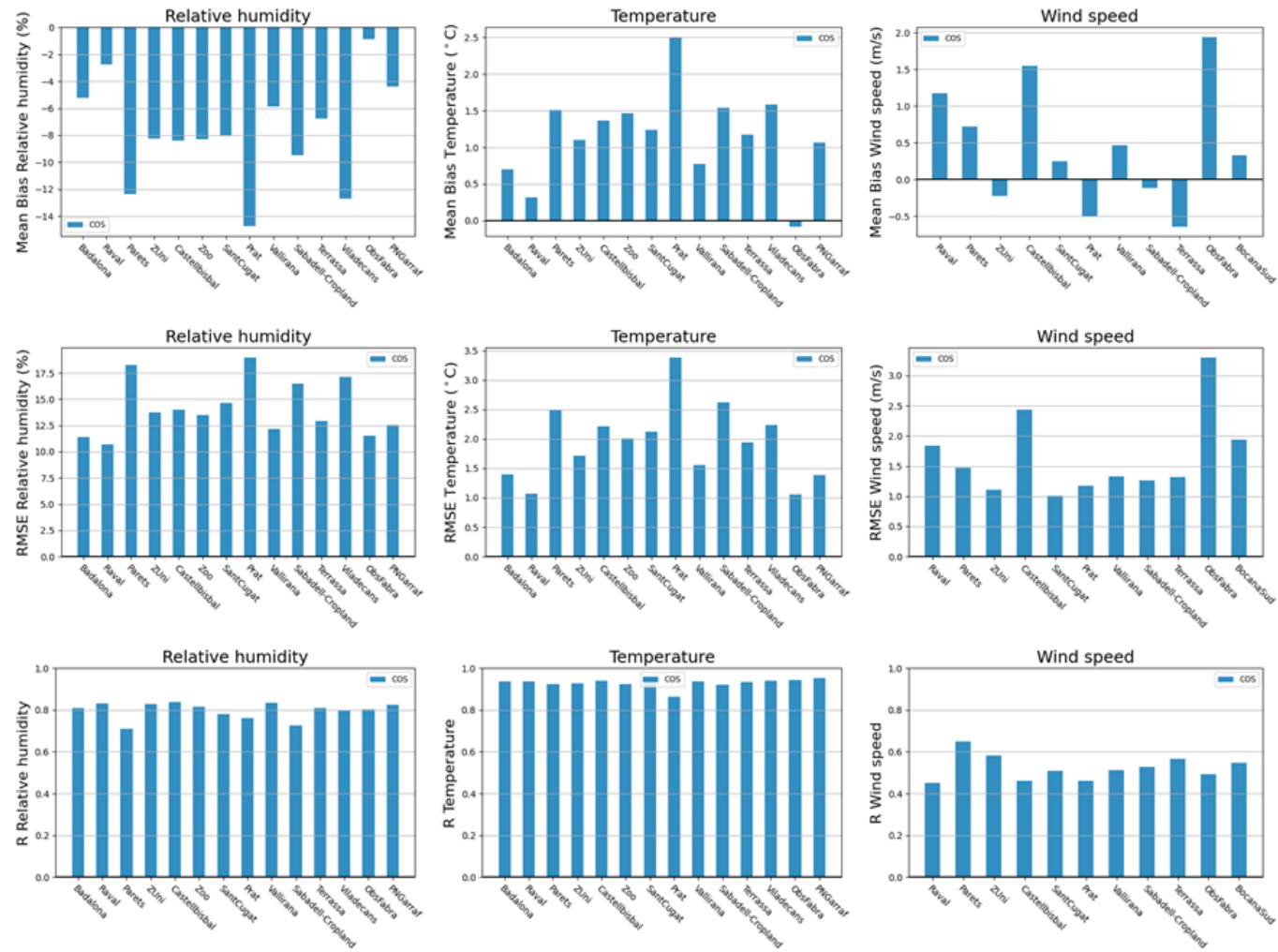


Figure S5 Upper panel: Hourly atmospheric radon concentrations (black dots) and PBLH estimations based on ceilometer (red dots) where dotted vertical lines represent 00:00 and solid lines 12:00 UTC; Bottom panel: Hourly ratio between radon concentration and PBLH estimates labeled per hour of the day.

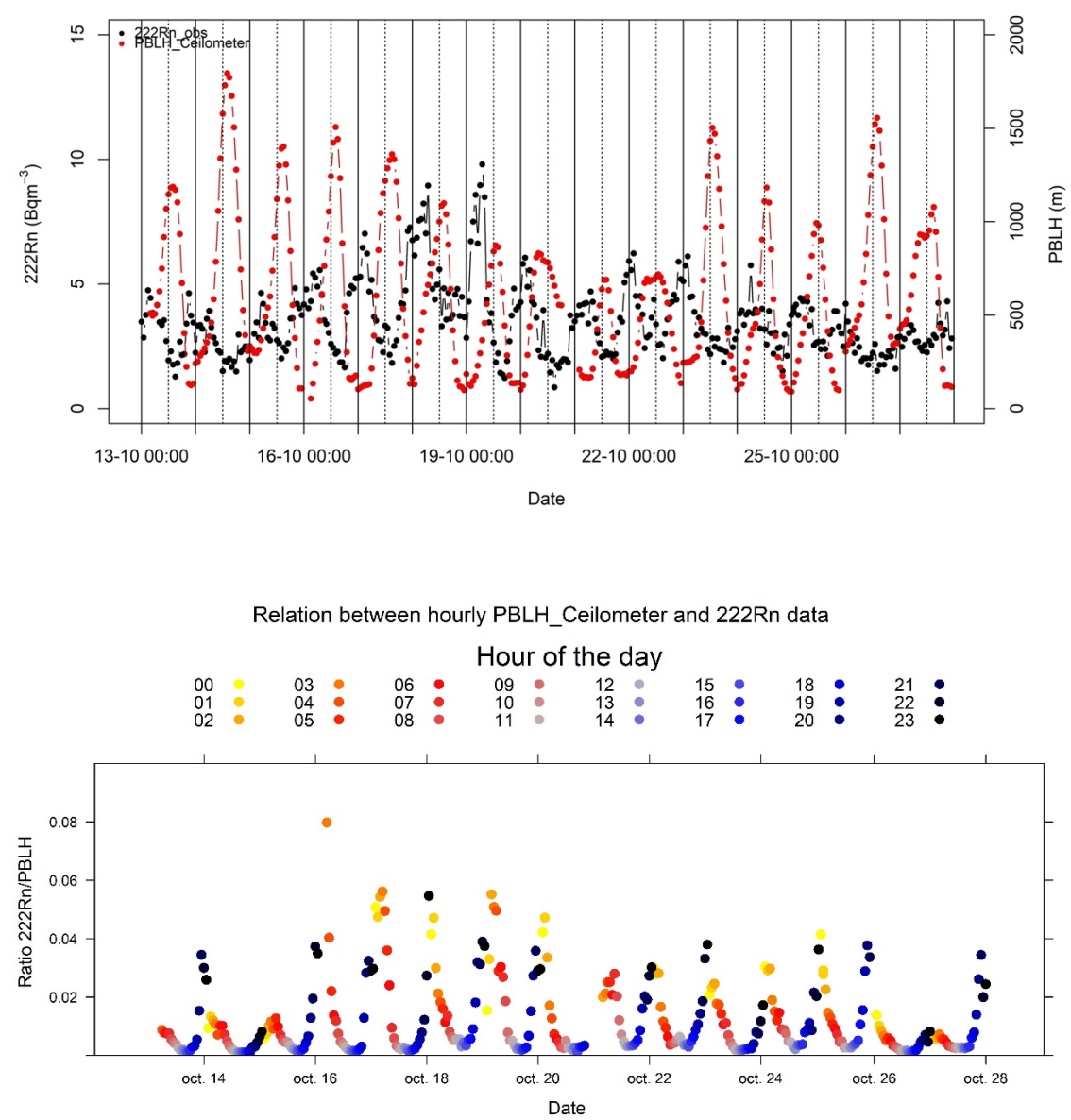


Figure S6. Pseudo gradient of radon (black line) and the wind speed (blue line) time series obtained at the same Target during October 2020. Lines indicating the values of the quartile 1, 2 and 3 used for the stability index analysis are also presented in red, violet and brown colors respectively.

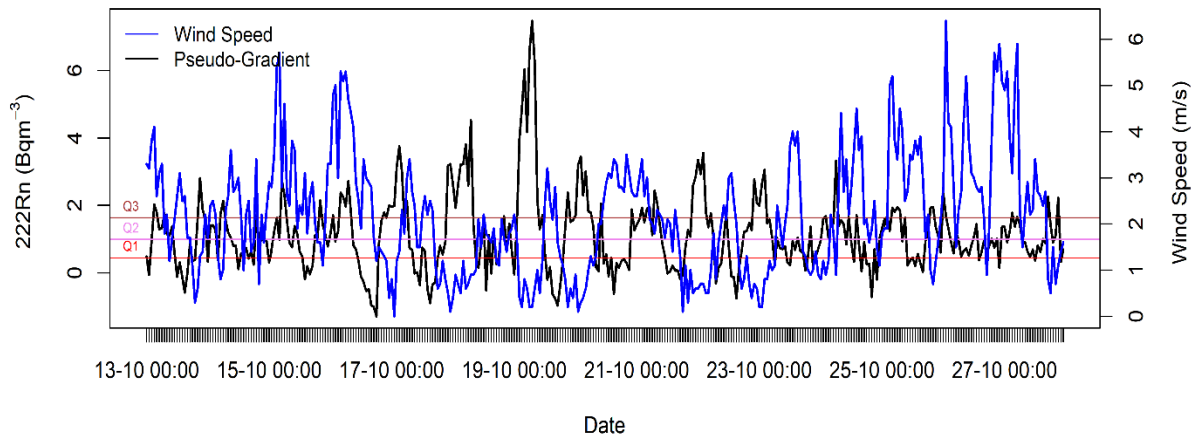


Figure S7. Maps (a,b) with mean ($\pm\sigma$) OCS Values from NOAA stations, GIF station, Utrecht station and our background value calculated from Tibidabo upwind site, in the northern hemisphere calculated using data from the periods in which the campaigns were done. Plots (c,d) show mean ($\pm\sigma$) from OCS values obtained during both campaigns in Barcelona area (BCN), and at the European Integrated Carbon Observation System (ICOS) station located in Gif-Sur-Yvette (Gif) and run by LSCE, and NOAA station Mace Head, Ireland (MHD) during the same period in a) May and b) October 2020.

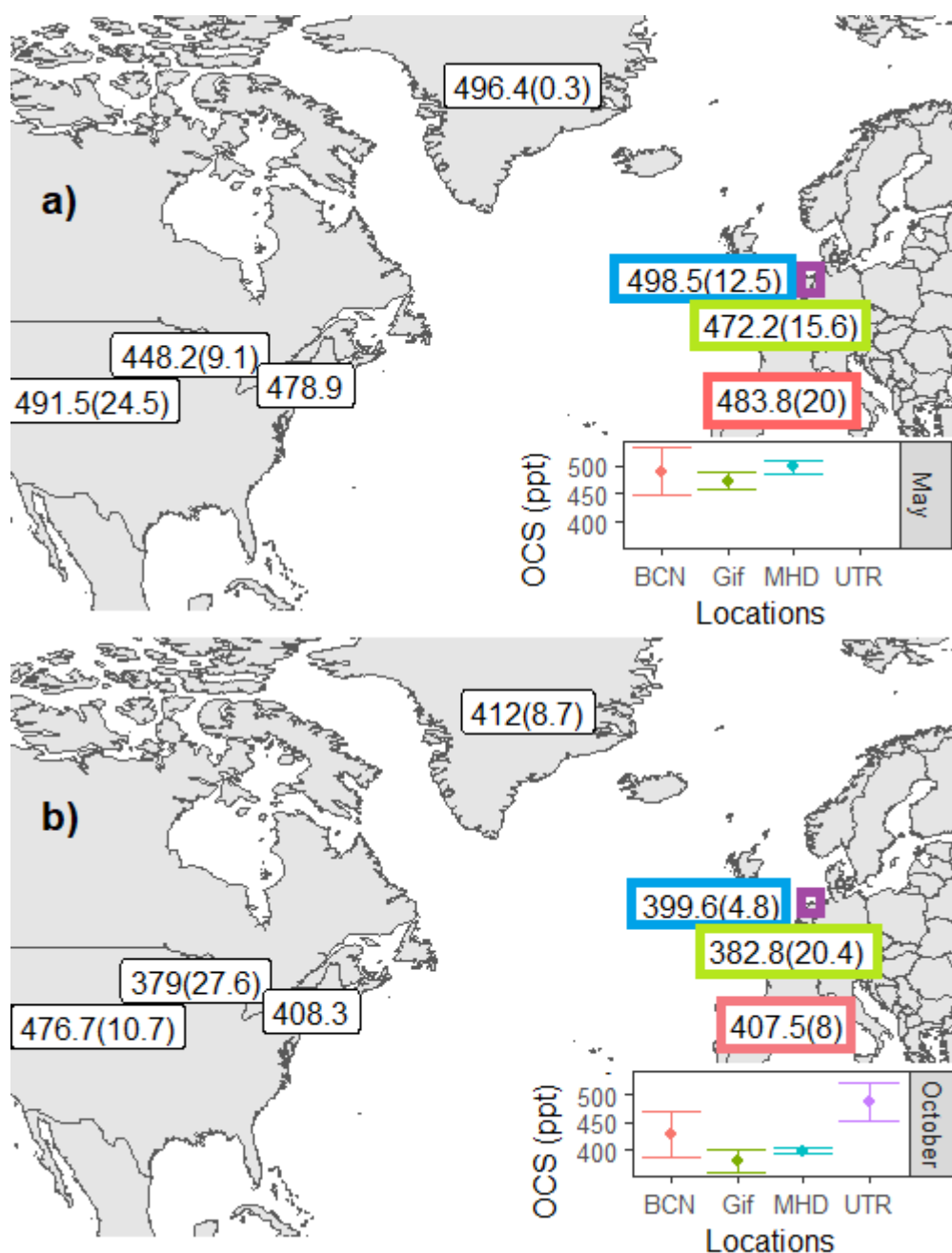
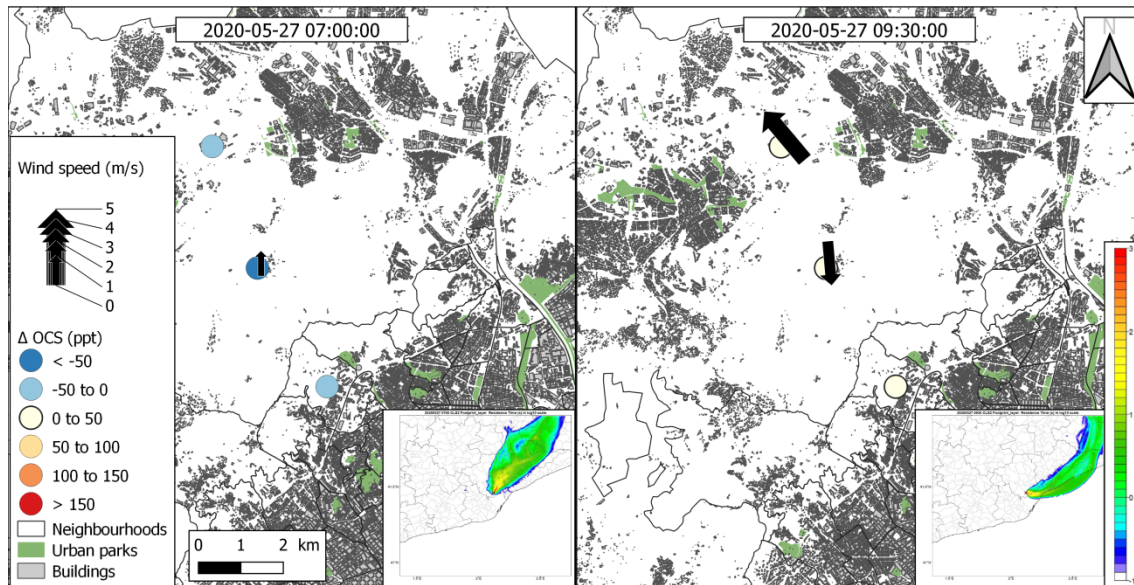
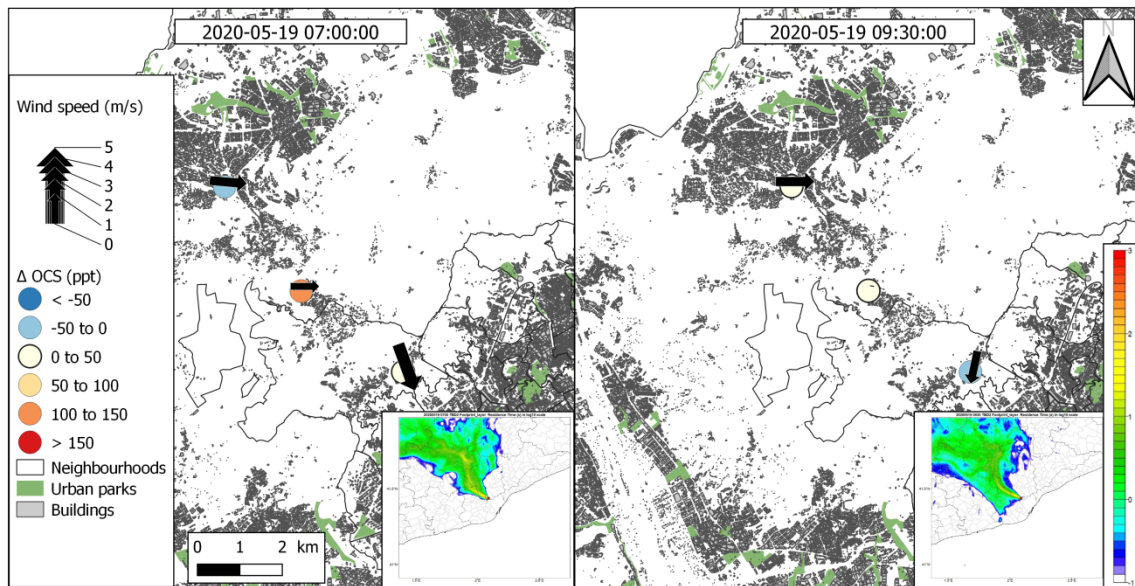


Figure S8. Maps showing the increase or decrease in OCS referred to background values (Delta OCS) for May at Forest: a) Collserola and b) Tibidabo. Delta OCS is in ppt. background values were 487 ppt. Black arrow shows the wind direction and speed measured on site, the absence of an arrow means that there was not wind blowing at the moment of measurement. Small map on the left is showing the residence time in logarithmic scale of the air masses arriving at the sampling site, determined with the FLEXPART model.

a) Collserola



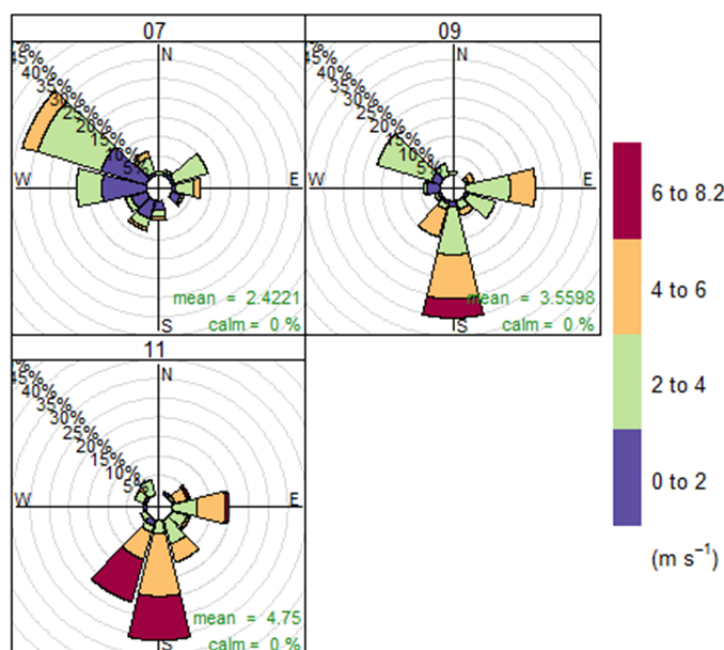
b) Tibidabo



Appendix S1

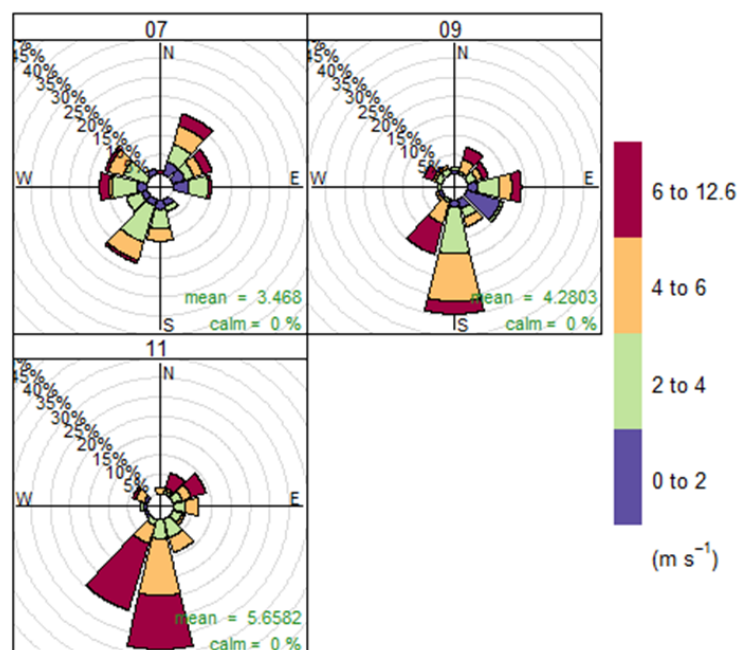
Wind rose showing typical winds direction and speed for our sampling period in Barcelona Metropolitan area. Data from May and June 2019, data provided from Meteocat (<https://www.meteo.cat/>). Each panel shows time in UTC at the upper part. Colour palette represent the wind velocity in m s^{-1} .

Prat de Llobregat



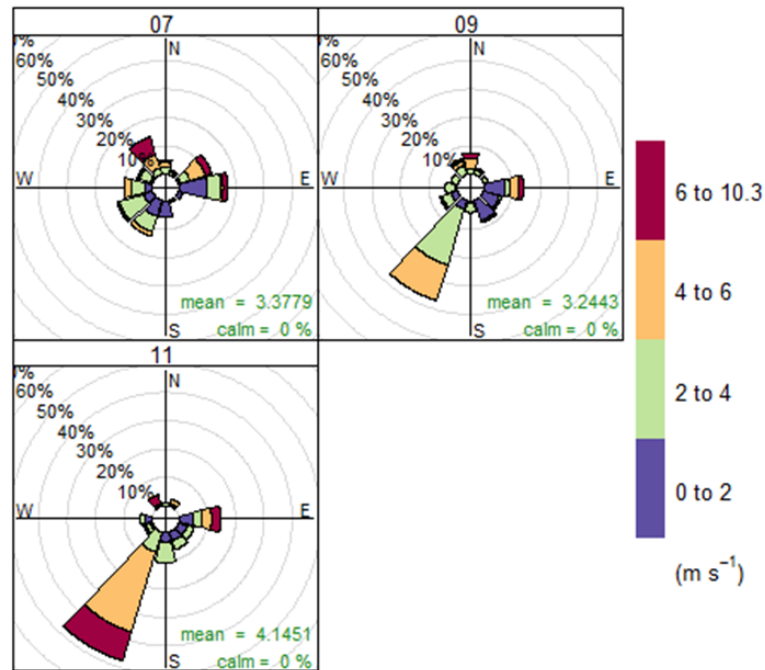
Frequency of counts by wind direction (%)

Port de Barcelona - Bocana Sud



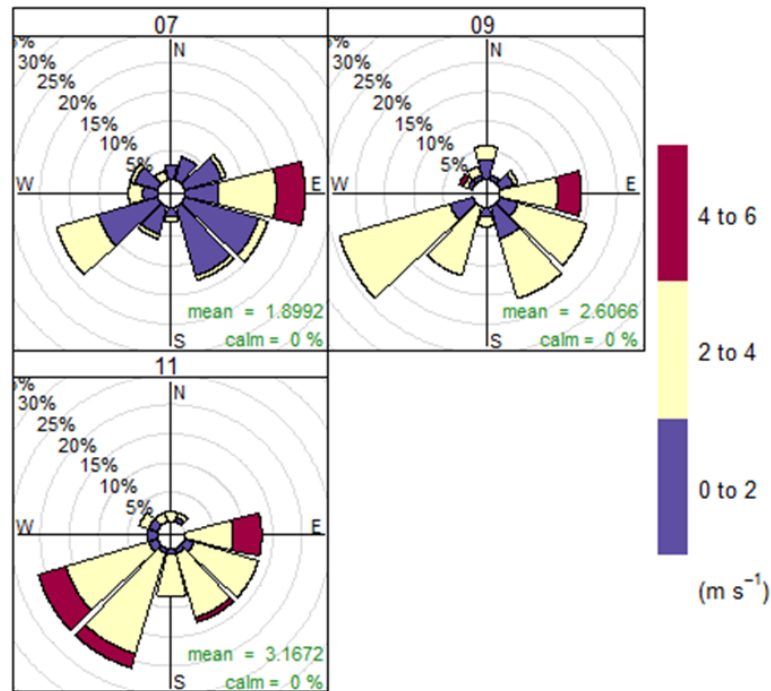
Frequency of counts by wind direction (%)

Barcelona - Observatori Fabra



Frequency of counts by wind direction (%)

Barcelona - el Raval



Frequency of counts by wind direction (%)

Meteocat stations locations: Prat de Llobregat is located near Prat and Gava Targets. Por the Barcelona is located near Montjuic, Observatori Fabra is located near Tibidabo background sampling Target nd elRaval is located in the vicinities of Sagrada and Poble Nou.

Appendix S2

Pictures and brief description showing the Targets measured during the campaigns:

- Agricultural (Gavà and Prat)



The two pictures on the left correspond to Gavà and two pictures on the right show Prat, both are similar agricultural Targets with Gava having one of the locations next to the sea and Prat was a couple km inland.

- forest (Tibidabo and Collserola)



Tibidabo and collserola have in common being forested areas and being topographically elevated.

- urban green (Montjuic and Guinardó)



Guinardó (Left) and Montjuic (Right) are both extensive urban parks within the city of Barcelona, but while Guinardó is surrounded by intensive anthropogenic constructions (buildings and roads), Montjuic is located next to the sea, however, Montjuic is near the Port of Barcelona, where an intensive activity is performed.

- Urban (Sagrada Familia and Poble Nou).



Those locations are representatives of Barcelona streets; they are characterized by high intensive traffic and commercial activities. The only difference between both Targets is that, Poble Nou area is next to the sea and has marine influence while Sagrada is located at the heart of the city.

Appendix S3

The structure of the PBL, part of the troposphere that is directly influenced by the presence of the earth's surface, can be complicated and variable (Stull, 1988). The PBL height is commonly used to characterize the vertical extension of the mixed layer and the level at which exchange with the free atmosphere occurs (Seibert et al., 2000). PBL height estimation methods can differ among them by several hundred meters. For these reasons, some studies compare methods and their uncertainties (Seidel, 2000; Seibert et al., 2000) or develop numerical procedures (Liu and Liang, 2010) to determine PBLH from available soundings.

PBL height on the campaign days was extracted from daily observations (at 0000 UTC and 1200 UTC) from the Barcelona radiosonde station (on the roof of the Physics Faculty, close to Palau Reial), part of the Global Meteorological Network. The sounding data includes the observed temperature, dew point, humidity, wind speed, and wind direction at different pressure levels. To determine the PBLH, we followed a robust numerical procedure proposed by Liu and Liang. (2010). this procedure begins by identifying for each sounding observation in which of the three major regimes of the PBL structure we are (Stull, 1988): unstable, stable, or neutral regime. To do so, we calculate the potential temperature between 5th and 2nd levels (chosen to remove raw data noises):

$$\theta_5 - \theta_2 = \begin{cases} < -\delta_s \text{ for CBL} \rightarrow \text{an unstable regime} \\ > +\delta_s \text{ for SBL} \rightarrow \text{a stable regime} \\ \text{else for NRL} \rightarrow \text{a neutral regime} \end{cases}$$

Where:

- θ = potential temperature (Kelvin) and its sub-index is the corresponding to the sounding data level (l=1 is on the surface)
- $\delta_s = \theta$ increment for the minimum strength of the stable (inversion) layer, above the CBL top or below the SBL top
($\delta_s = \theta$ for idealized cases but in practice is a small positive)

Unstable and neutral regime

Following the method from Liu and Liang (2010), for unstable and neutral regime, we scan upward twice: First, to find the lowest level $l=k$ that meets the condition $\theta_k - \theta_a \geq \delta_u$; where δ_u is the θ increment for the minimum strength of the unstable layer. And then, a second scan to search the occurrence of

$$\dot{\theta}_k \equiv \frac{\partial \theta_k}{\partial z} \geq \dot{\theta}_r$$

Where:

- θ = vertical thermal gradient per height z
- θ_r = minimum strength for the overlying inversion layer and can be considered as the overshooting threshold of the rising parcel.

Stable regime

For the stable regime, the PBLH is more difficult to quantify and there is no unique algorithm to determine it accurately without actual observations of the turbulence kinetic energy profile in the boundary layer (Stull, 1988; Seibert et al., 2000). SBL turbulence can result from two dominant mechanisms: buoyancy forced and/or shear driven. When the SBL is buoyancy forced, Liu and Liang (2010) propose to scan upward to find the lowest level at which $\dot{\theta}_k$ reaches a minimum and then determine the PBLH at that level if either of the following conditions is met:

$$\begin{cases} \dot{\theta}_k - \dot{\theta}_{k-1} < -\dot{\delta} \\ \dot{\theta}_{k+1} < \dot{\theta}_r, \dot{\theta}_{k+2} < \dot{\theta}_r \end{cases}$$

Where:

- 1st condition ensures that θ_r is a local peak with a curvature parameter θ_r of $40 \text{ K}\cdot\text{km}^{-1}$
- 2nd condition constrains that an inversion layer is not evident in the upper two layers.

PBLH is defined at either the top of the bulk stable layer or at the level of the low-level jet (LLJ) nose if present, whichever is lower.

References:

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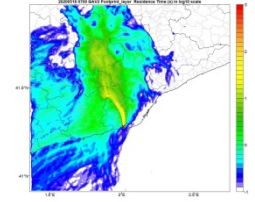
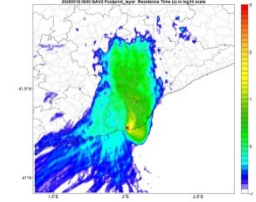
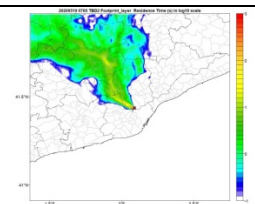
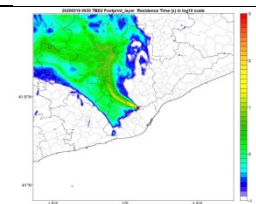
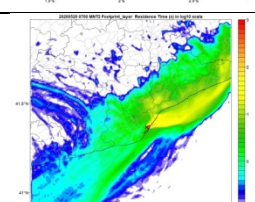
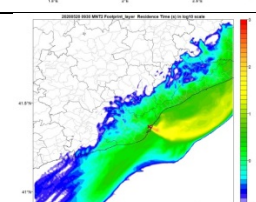
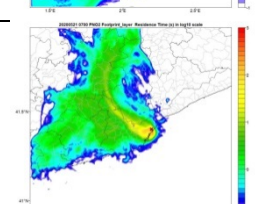
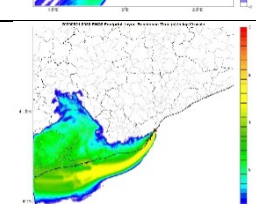
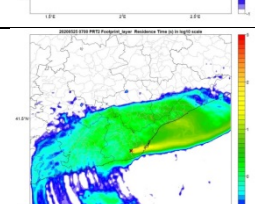
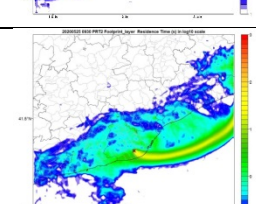
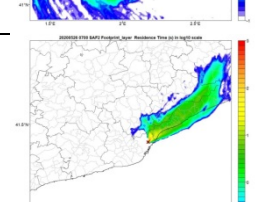
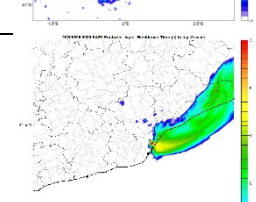
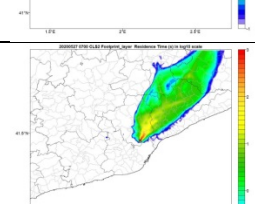
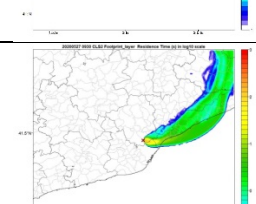
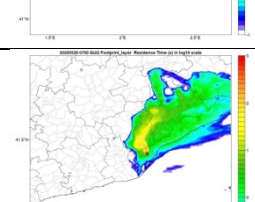
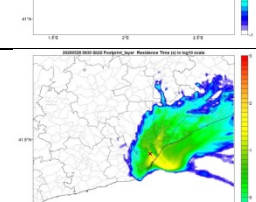
Seibert, P. et al., 2000. Review and intercomparison of operational methods for the determination of the mixing height. *Atmos. Environ.*, 34, 1001-1027

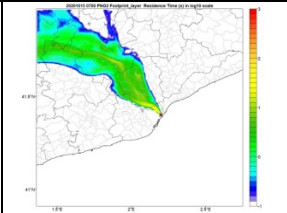
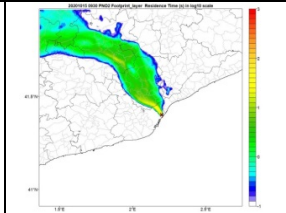
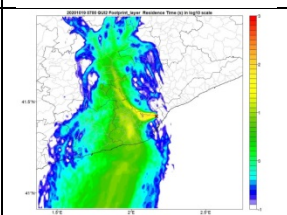
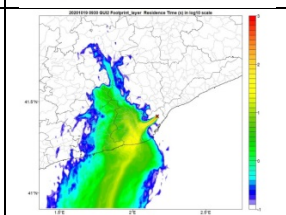
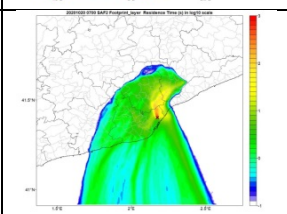
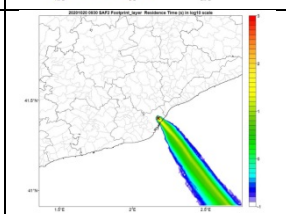
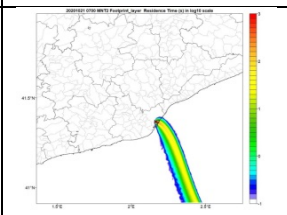
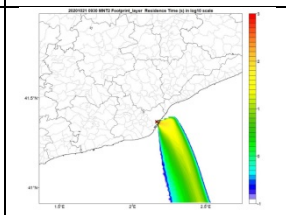
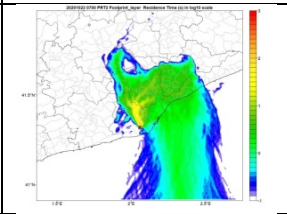
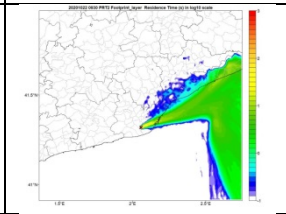
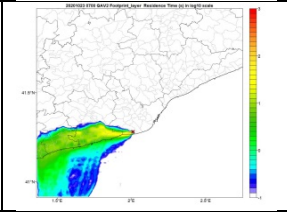
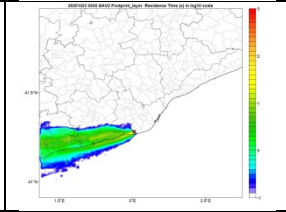
Seidel, D. J., C.O. Ao, and K. Li, 2010. Estimating climatological planetary boundary layer heights from radiosonde observations: Comparison of methods and uncertainty analysis, *J. Geophys. Res.*, 115, D16113, doi: 10.1029/2009JD013680.

Stull, R.B., 1988. *An Introduction to Boundary Layer Meteorology*. Kluwer Academic, 666 pp.

Appendix S4 Maps showing the results from FLEXPART models.

MEASUREMENTS AT 7 AND 9:30:

| Campaign | Date | 7 UTC | 930 UTC |
|----------|-------------------------------|---|--|
| May | 2020-05-18 Gavà |  |  |
| | 2020-05-19 Tibidabo |  |  |
| | 2020-05-20 Montjuic |  |  |
| | 2020-05-21 Poble Nou |  |  |
| | 2020-05-25 Prat |  |  |
| | 2020-05-26 Sagrada Familia |  |  |
| | 2020-05-27 Collserola |  |  |
| | 2020-05-28 Guinardó |  |  |

| | | | |
|---------|-------------------------|---|--|
| October | 2020-10-15 Poble Nou |  |  |
| | 2020-10-19 Guinardó |  |  |
| | 2020-10-20 Sagrada |  |  |
| | 2020-10-21 Montjuic |  |  |
| | 2020-10-22 Prat |  |  |
| | 2020-10-23 Gavà |  |  |

MEASURES AT 4 AND 11:

| | Date hour | Gavà | Tibidabo | Poble Nou |
|------|-------------------------------|------|----------|-----------|
| May | 2020-05-18 4:00 UTC | | | |
| | | | | |
| | 2020-05-21 4:00 UTC | | | |
| | | | | |
| | 2020-05-25 4:00 UTC | | | |
| | | | | |
| Oct. | 2020-10-13 4:00 UTC | | | |
| | 07:00 UTC | | | |

