Table S1. Summary of WRF parameterizations (http://wrf-model.org).

Parameter	Option
Microphysics	WRF Single-Moment 5-class (WSM5).
Longwave and Shortwave Radiation	Rapid Radiative Transfer Model for general circulation models (RRTMG).
Surface Layer	Revised version of the fifth generation Pennsylvania State University–National Center for Atmospheric Research Mesoscale Model (MM5) parameterization.
Land Surface	Noah land surface model (LSM).
PBL	Yonsei University non-local closure scheme (YSU).
Cumulus Parameterization	Kain-Fritsch.
Dynamics & FDDA	Recommendations of WRF user's guide.

Table S2. Description of SNAP source categories in TNO-MACC-III (as in Kuenen et al., 2014).

SNAP	SNAP sector description
1	Energy industries
2	Non-industrial combustion
34	Industry (combustion and processes)
5	Extraction and distribution of fossil fuels
6	Product use
7	Road transport
8	Non-road transport and other mobile sources
9	Waste treatment
10	Agriculture

Table S3. Overview of ozonesonde stations.

Station	Longitude (deg)	Latitude (deg)	Elevation (m)	Daily Records	UTC time
Valentia	10.25 W	51.93 N	14	7	11:00–12:00
Barajas	3.58 W	40.47 N	631	12	10:00–11:00
Legionowo	20.97 E	52.40 N	96	9	11:00–12:00
Hohenpeissenberg	11.00 E	47.80 N	976	25	04:00–06:00
Payerne	6.57 E	46.49 N	491	36	11:00–12:00
Uccle	4.35 E	50.80 N	100	40	11:00–12:00
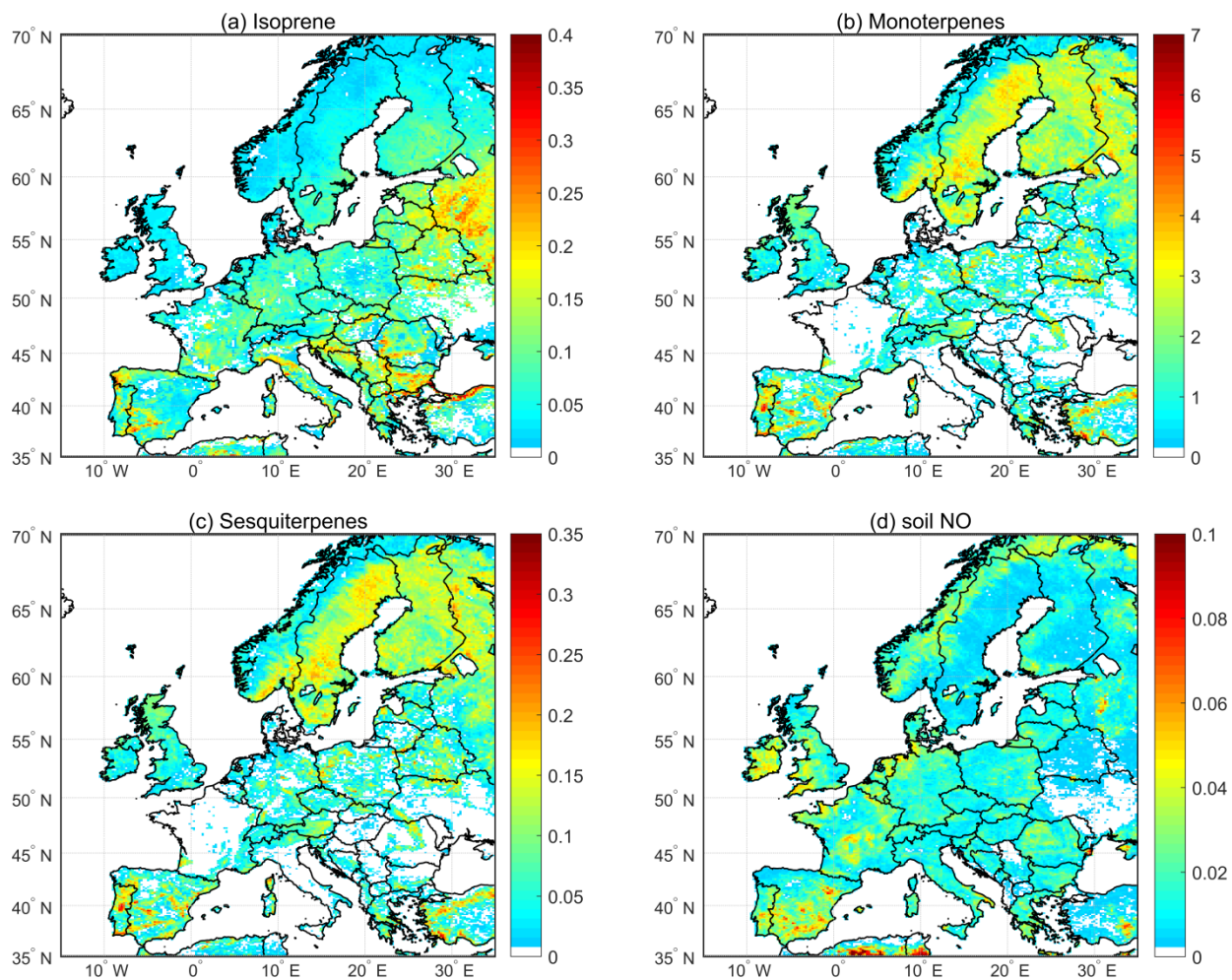
Figure S1. Spatial distributions of total (a) isoprene, (b) monoterpene, (c) sesquiterpene and (d) soil NO emissions (t km^{-2}) in Europe for the summer of 2010.
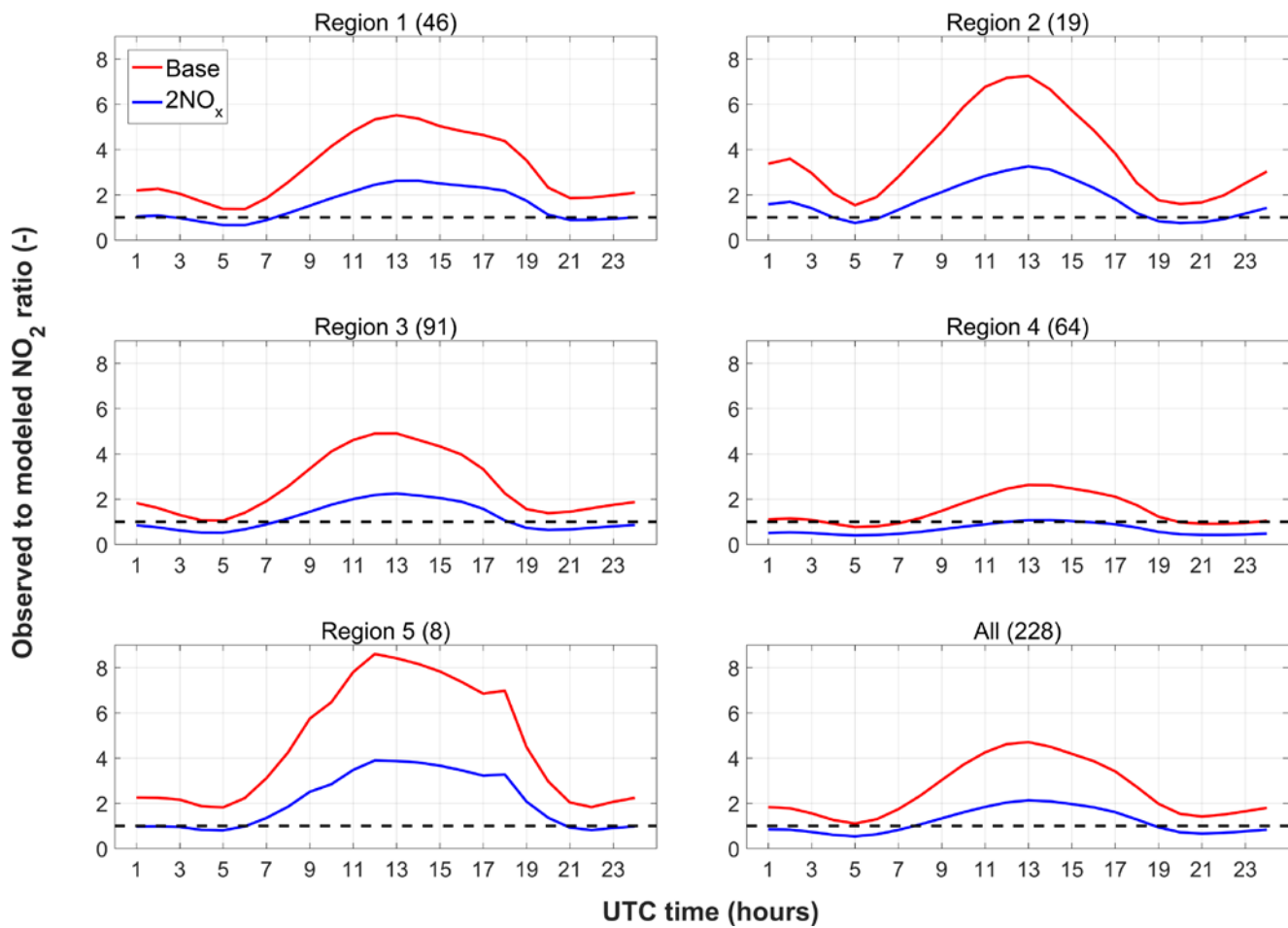
Figure S2. Diurnal profile of the ratio of observed to modeled surface NO$_2$ mixing ratios for Regions 1–5 as well as for the whole domain (All) in summer 2010. The number of stations available for each region is reported in parentheses at the top of each panel. The dashed black line is the ratio of 1. Red line shows the base case, blue line shows the scenario with doubled NO$_x$ emissions (2NO$_x$).
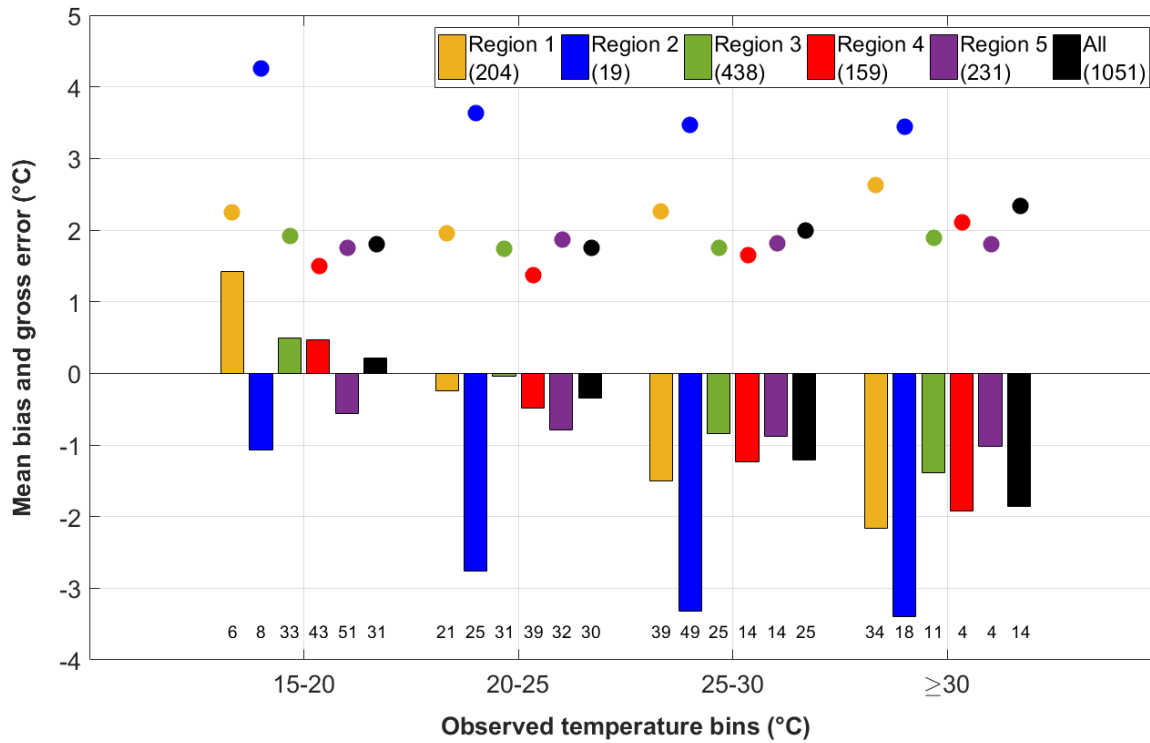
Figure S3. Temperature (2 m) mean bias (bars) and gross error (dots) for observed afternoon temperature bins for each region in Europe for the summer of 2010. Values below the bars indicate the fraction (%) of the values assigned to each bin for each region. The number of stations available for each region is reported in parentheses in the legend.
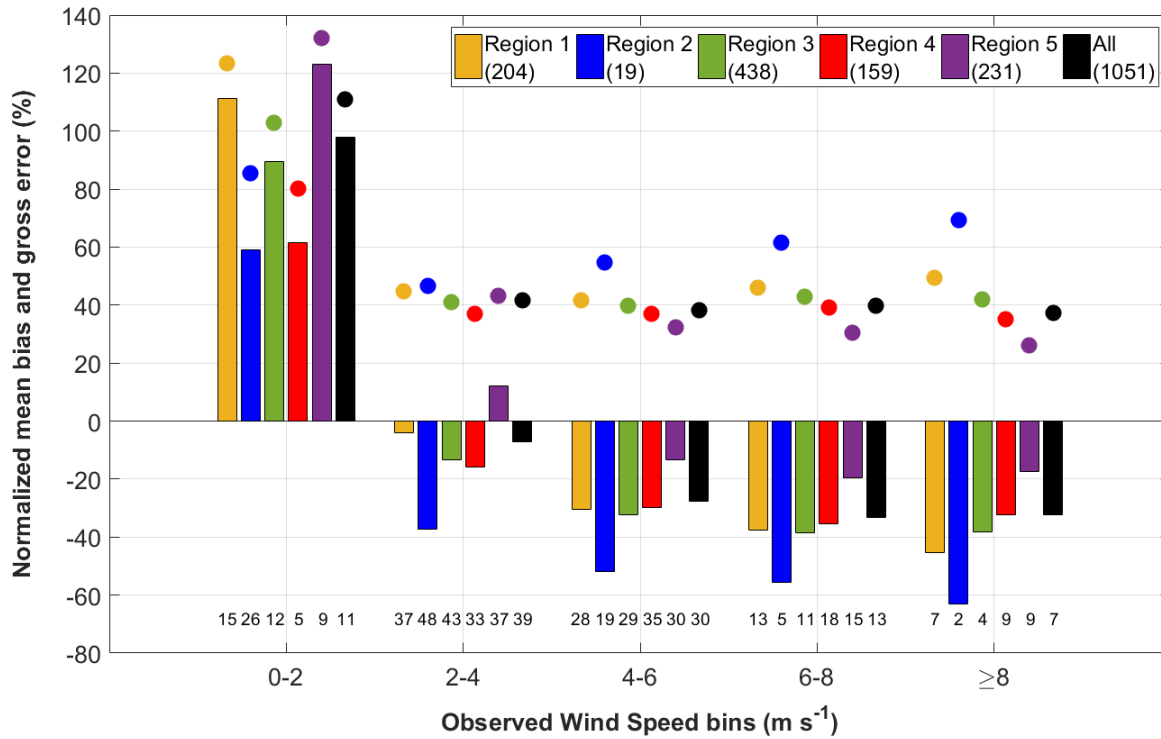
Figure S4. Wind speed (10 m) normalized mean bias (bars) and gross error (dots) for observed afternoon wind speed bins for each region in Europe for the summer of 2010. Values below the bars indicate the fraction (%) of the values assigned to each bin for each region. The number of stations available for each region is reported in parentheses in the legend.
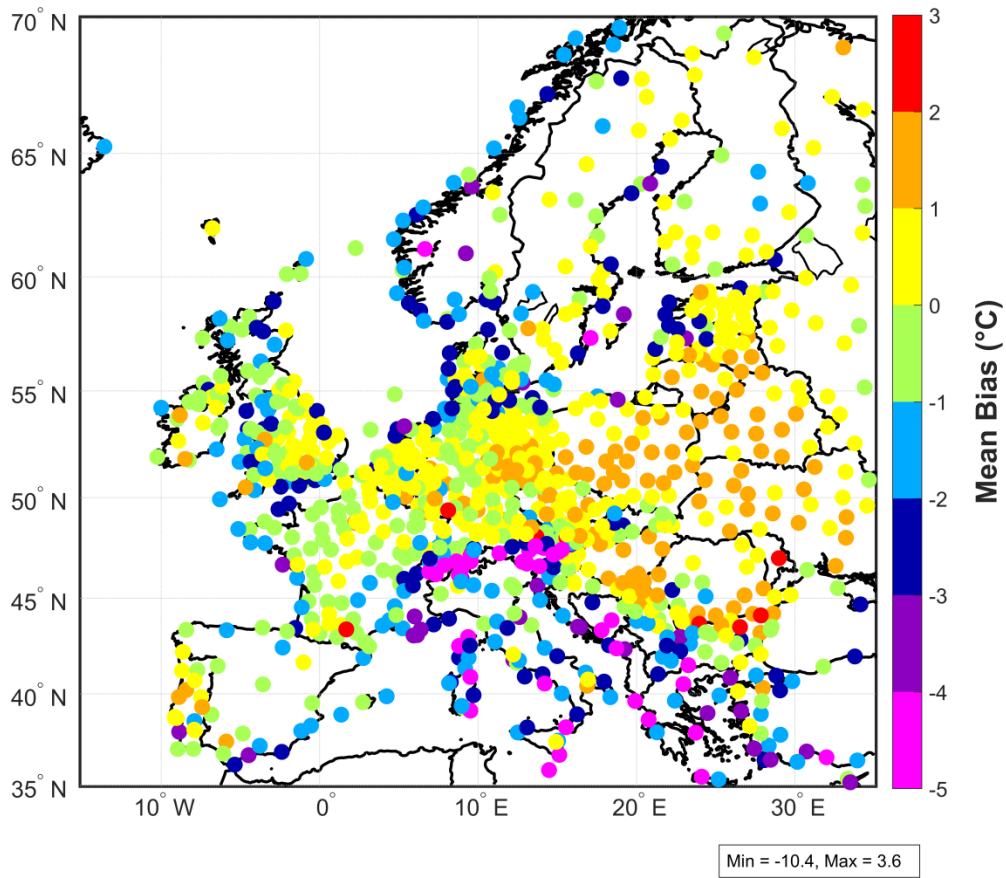
Figure S5. Temperature (2 m) afternoon mean bias for each station in Europe for the summer of 2010. There are 18 stations outside the color scale.

Min = -5.8, Max = 7
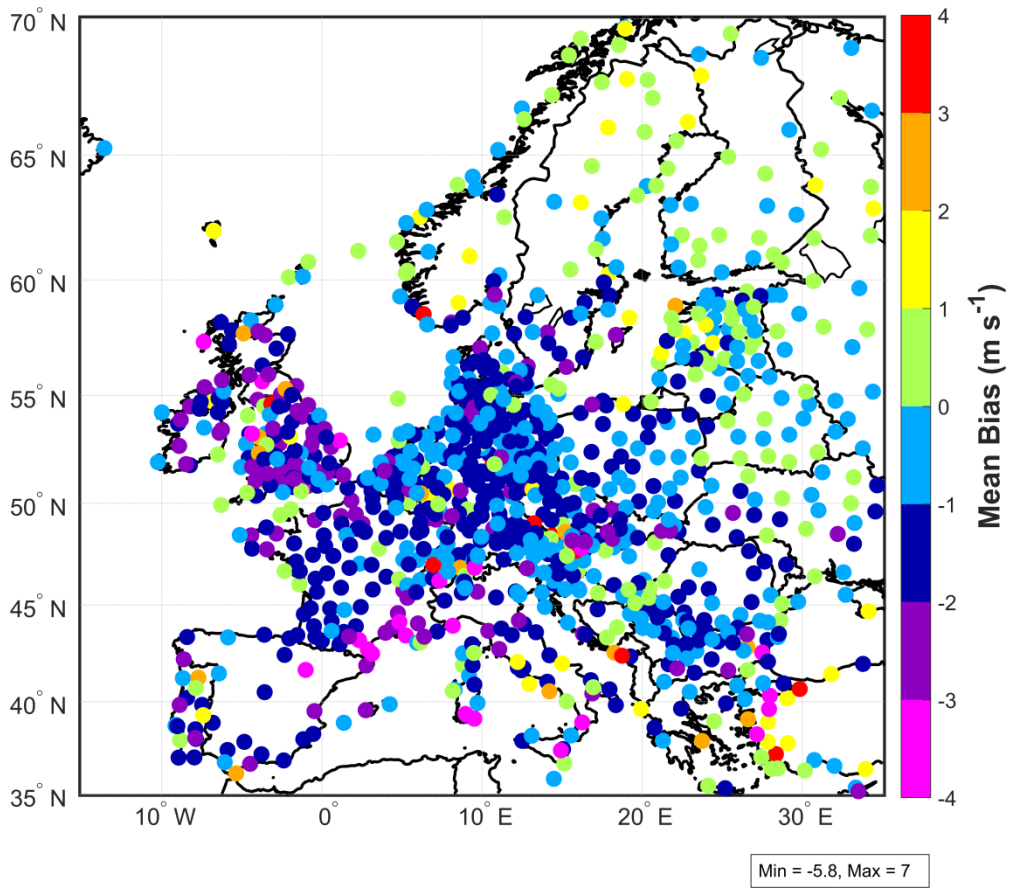
Figure S6. Wind speed (10 m) afternoon mean bias for each station in Europe for the summer of 2010. There are 8 stations outside the color scale.

Min = -591, Max = 856
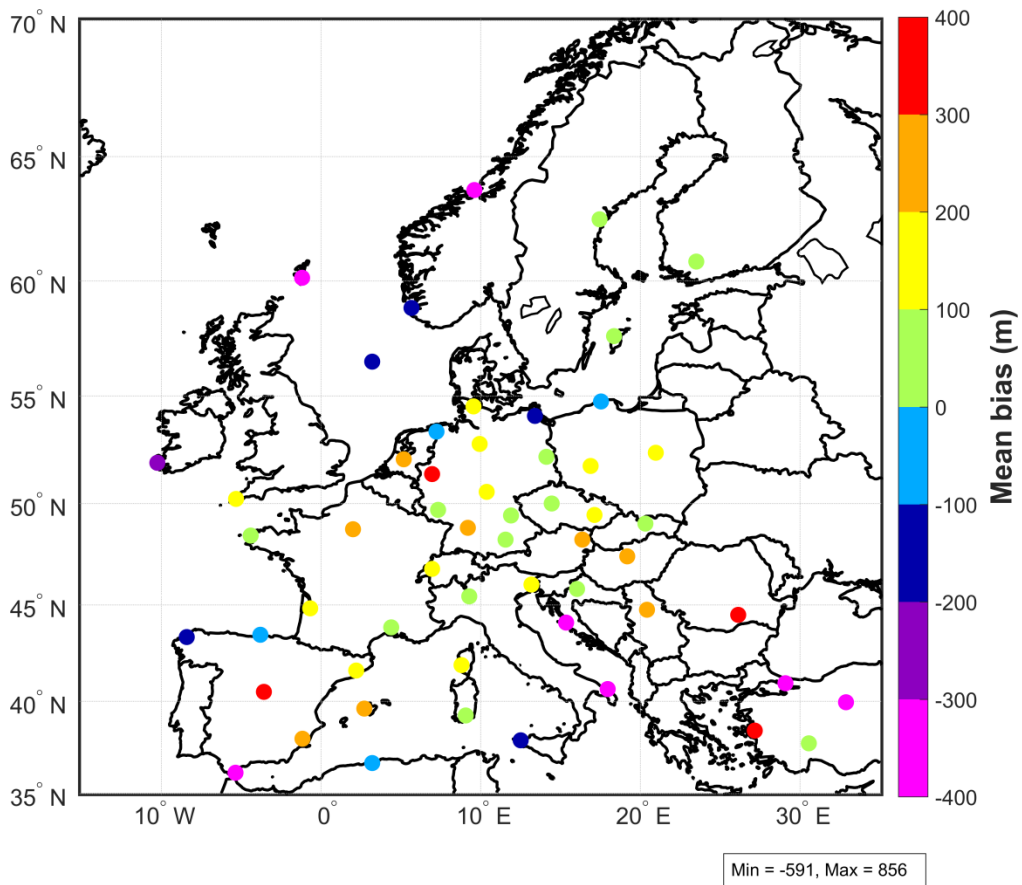
Figure S7. PBLH mean bias at 12 UTC for each station in Europe for the summer of 2010. There are 5 stations outside the color scale.
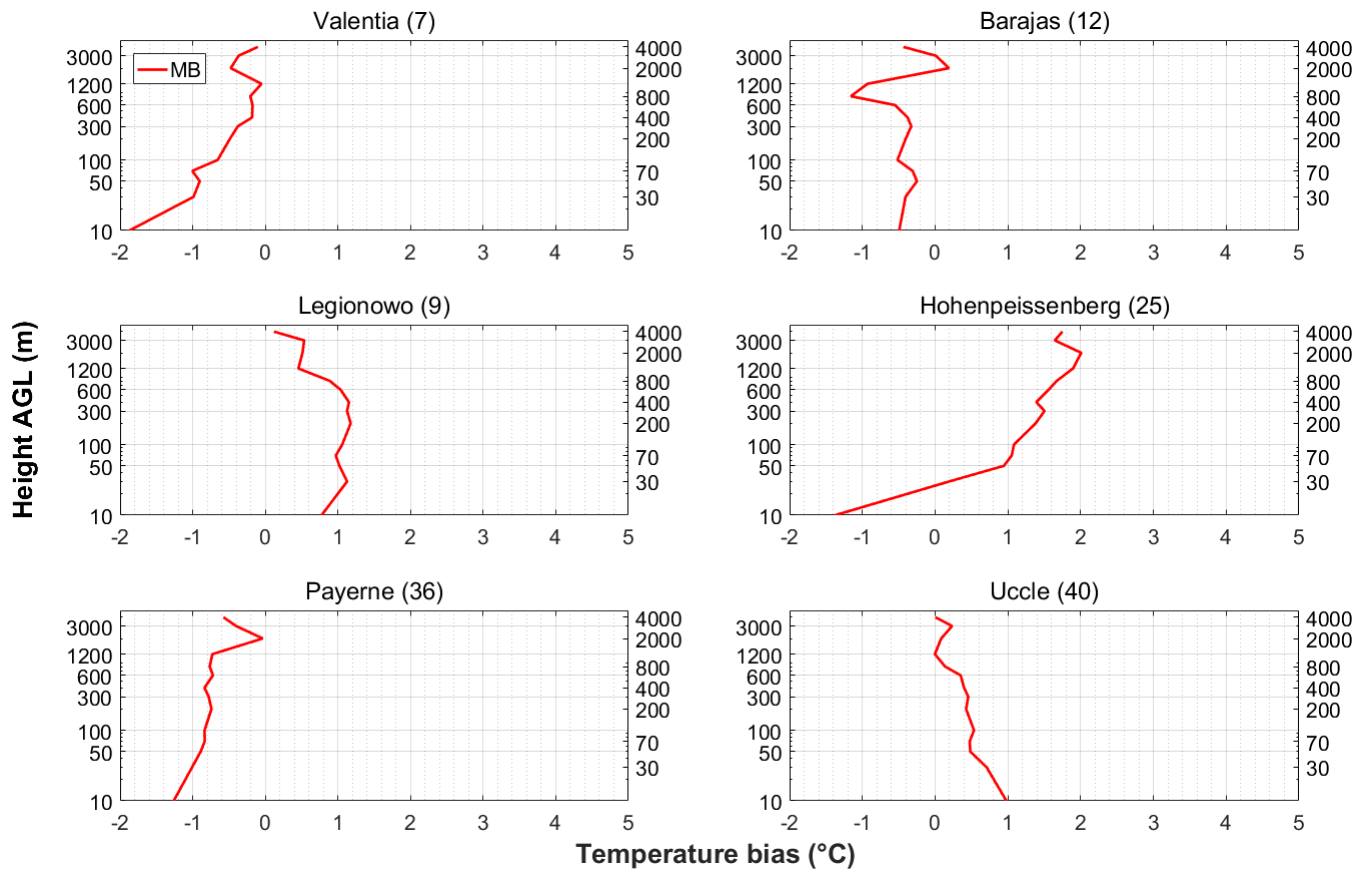
Figure S8. Vertical profiles of temperature mean bias for 6 stations for the summer of 2010. The number of ozonesondes available for each station is reported in parentheses at the top of each panel. Heights of 14 model layers are shown on both y-axes which are in logarithmic scale.
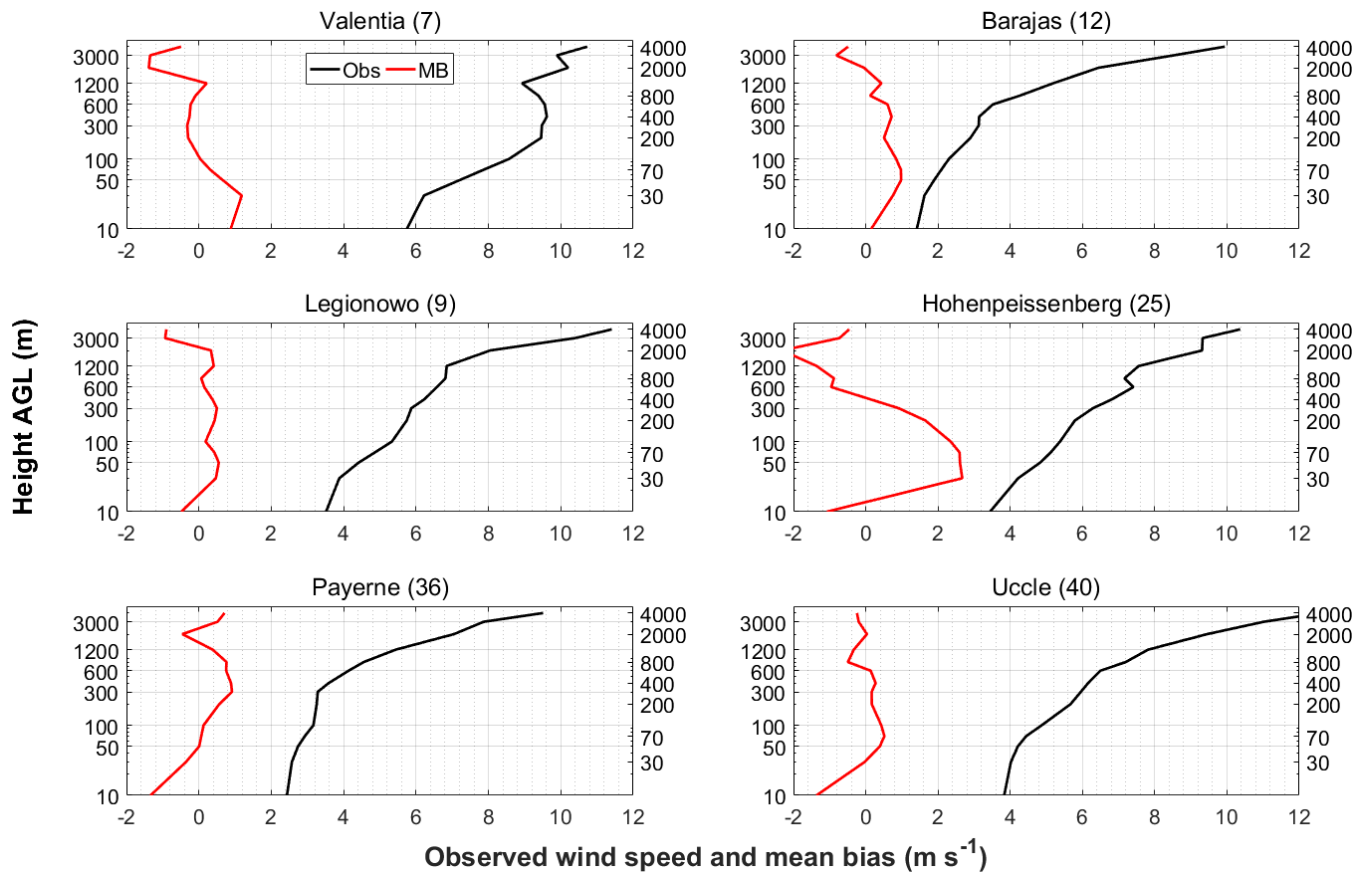
Figure S9. Vertical profiles of observed wind speed and mean bias for 6 stations for the summer of 2010. The number of ozonesondes available for each station is reported in parentheses at the top of each panel. Heights of 14 model layers are shown on both y-axes which are in logarithmic scale.
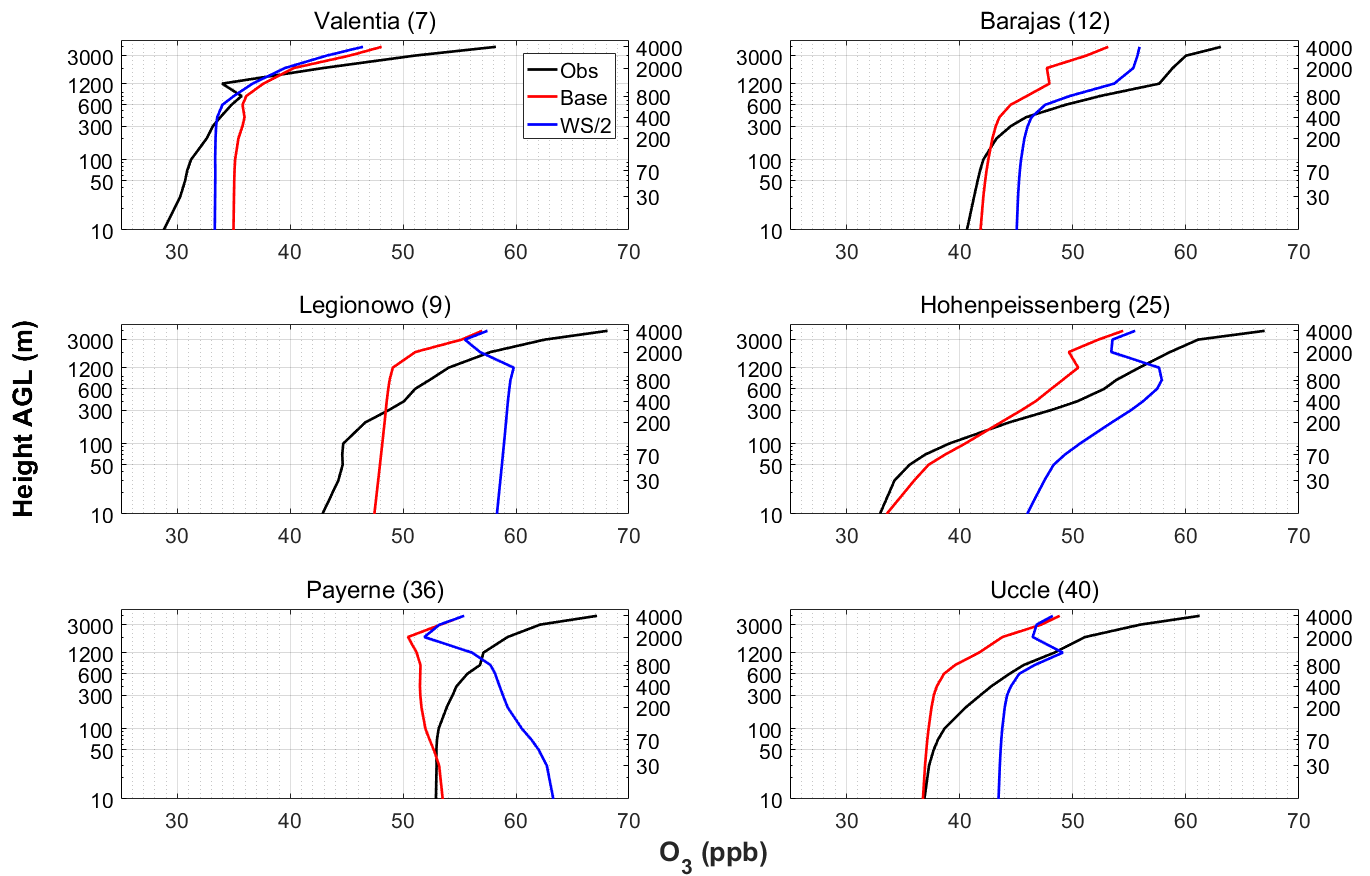
Figure S10. Observed and modeled O_3 mixing ratio vertical profiles for 6 stations for the summer of 2010. The number of ozonesondes available for each station is reported in parentheses at the top of each panel. Heights of 14 model layers are shown on both y-axes which are in logarithmic scale.
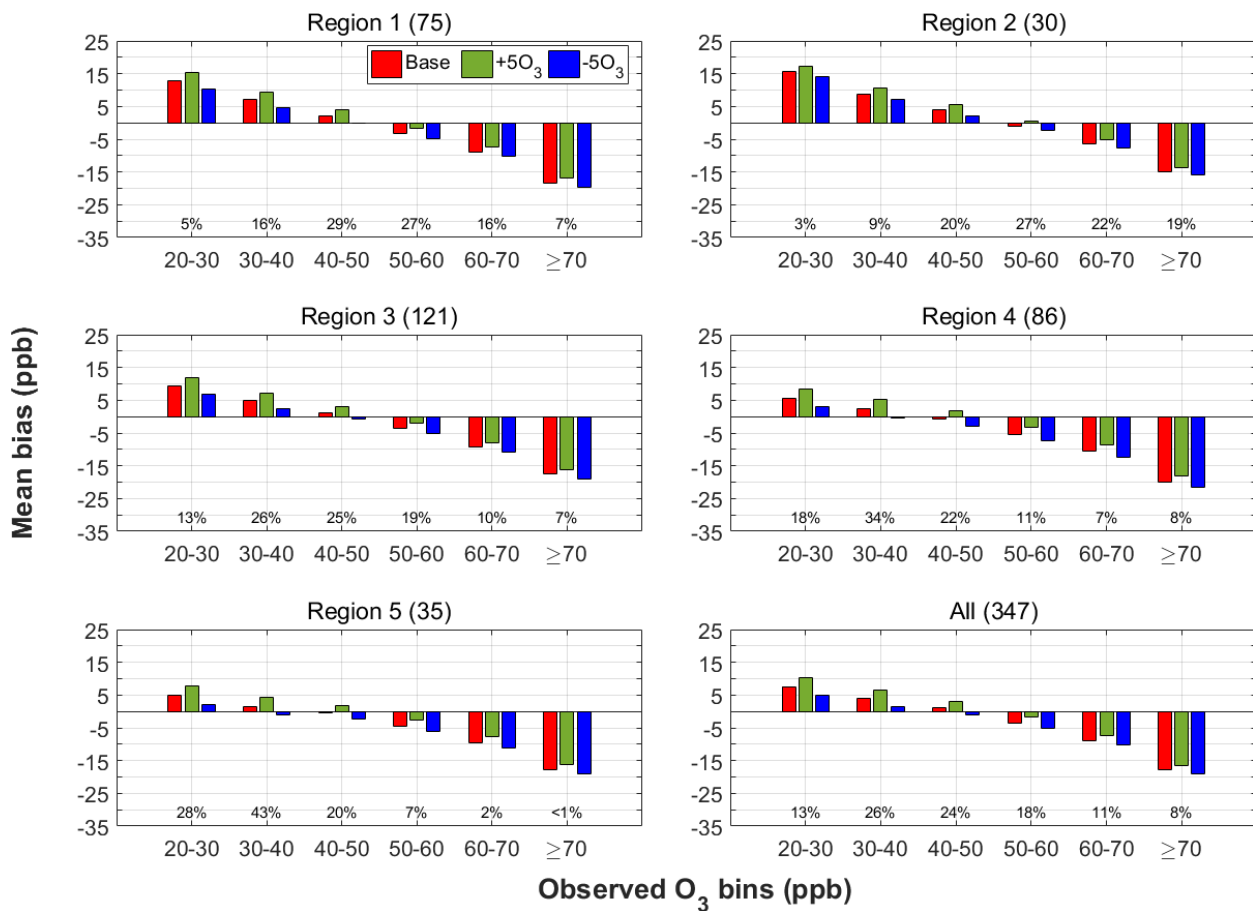
Figure S11. Mean bias of the afternoon surface O_3 mixing ratio for each bin of observed surface O_3 mixing ratios for various O_3 ICBC scenarios in summer 2010. Percentage values below the bars indicate the fraction of the values assigned to each bin for each region. The number of stations available for each region is reported in parentheses at the top of each panel.