

SUPPLEMENTARY MATERIAL

Supplementary S1



Figure S1 – Map of the Italian administrative Regions.

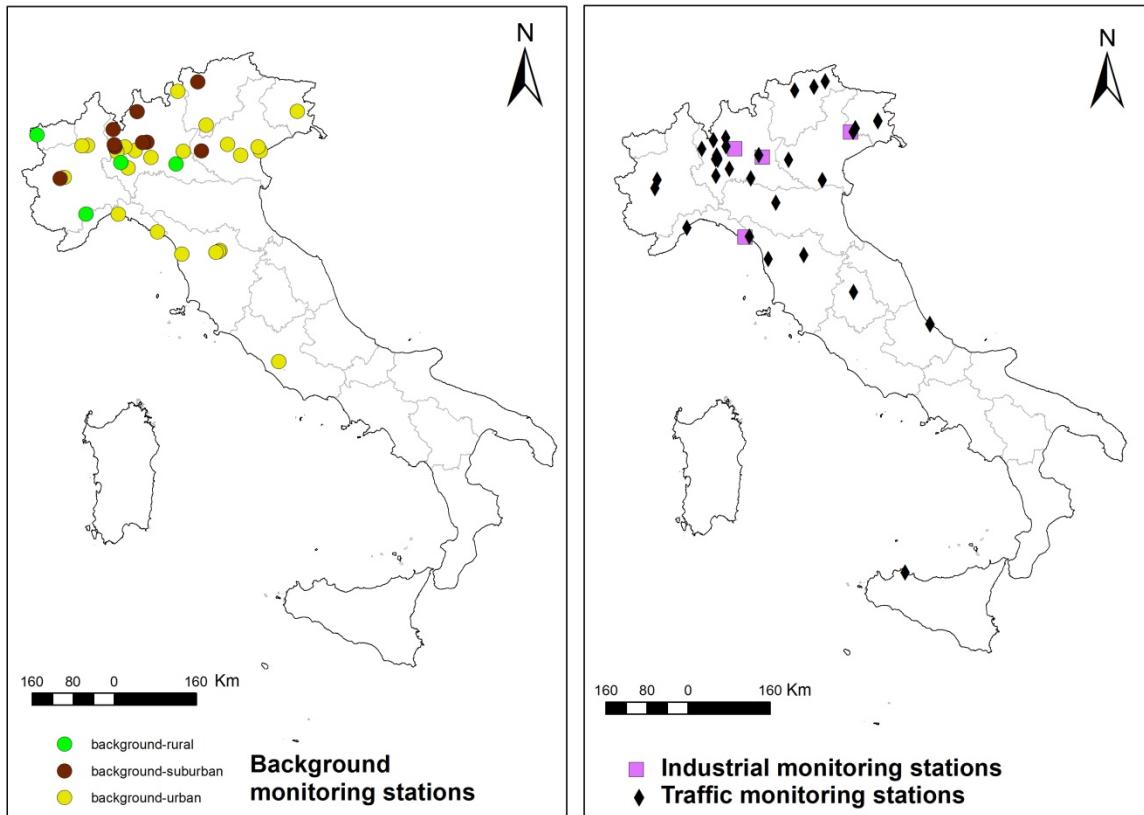


Figure S2 – Map of the Italian sites monitoring NO₂ concentrations considered for the study.

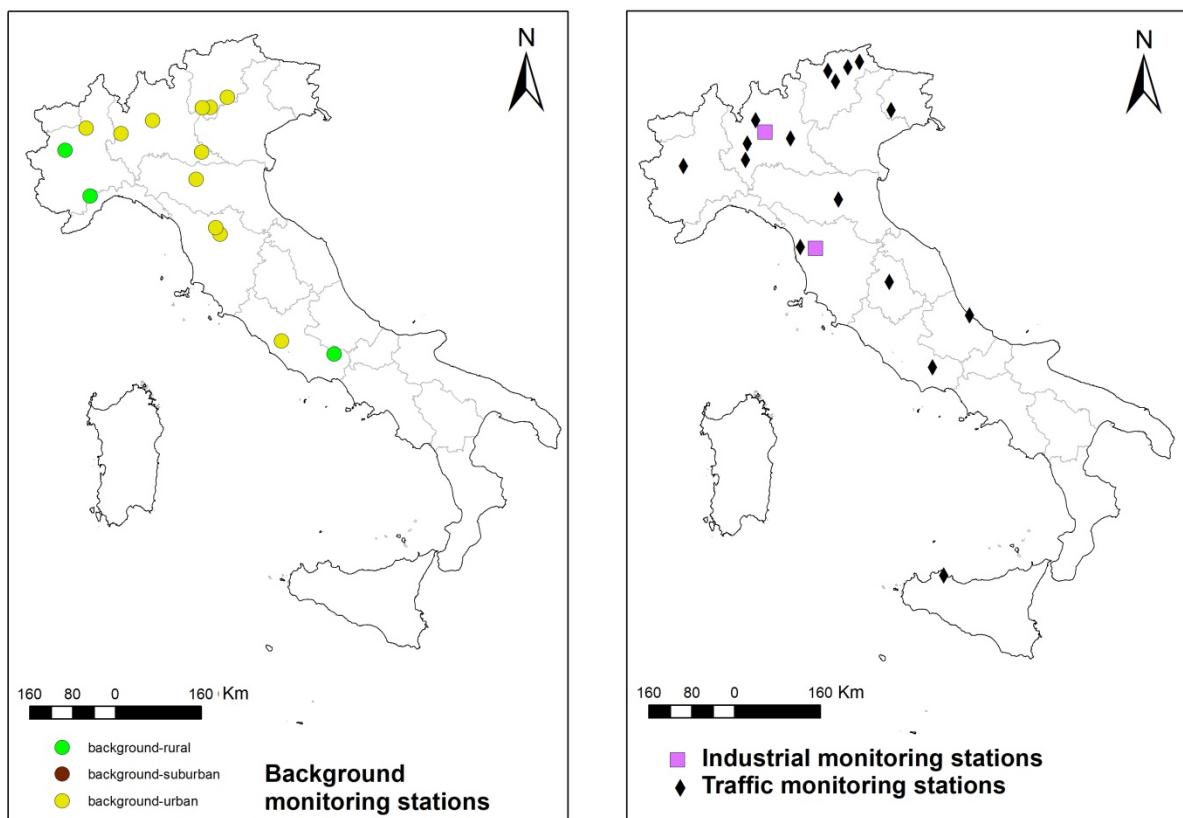


Figure S3 – Map of the Italian sites monitoring PM10 concentrations considered for the study.

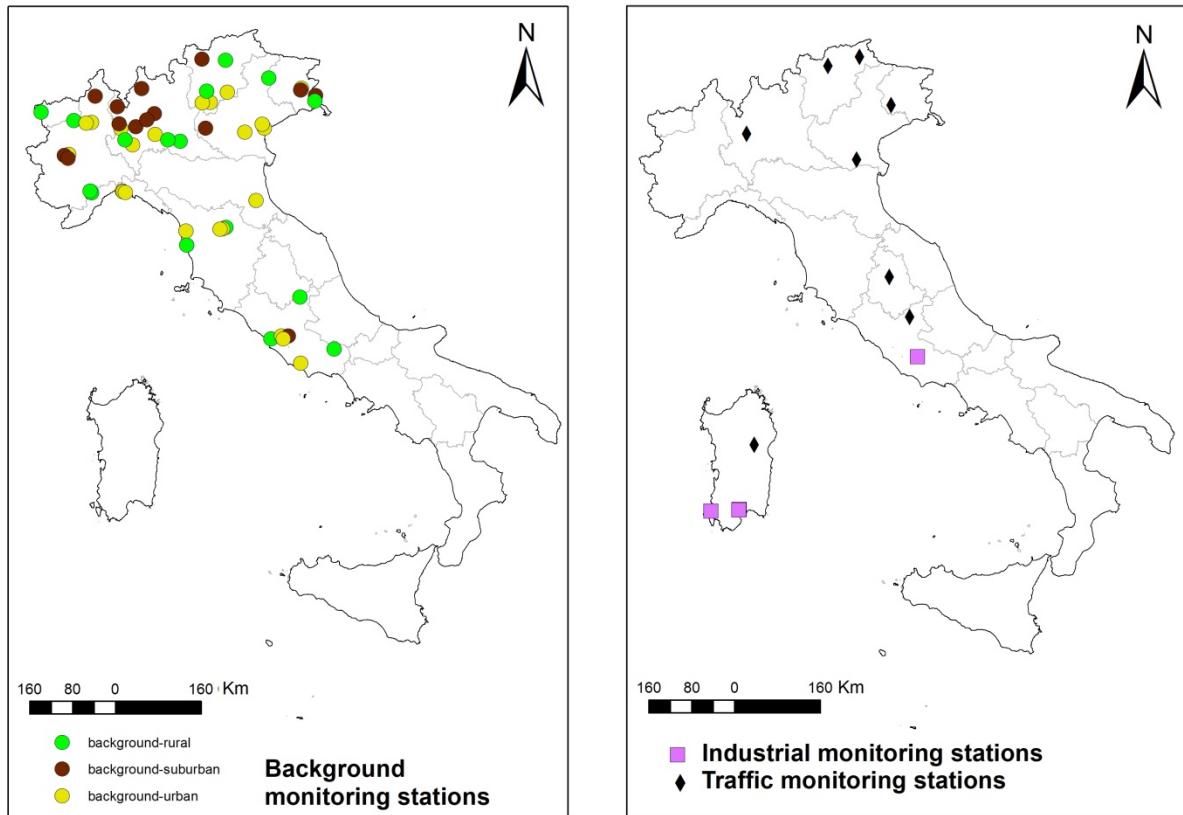


Figure S4 – Map of the Italian sites monitoring O₃ concentrations considered for the study.

Supplementary S2 – Emissions Trend

Table S1 – SNAP Classification

SNAP - macrosector	
SNAP code	Description
01	Combustion in energy and transformation industries
02	Non-industrial combustion plants
03	Combustion in manufacturing industry
04	Production processes
05	Extraction and distribution of fossil fuels and geothermal energy
06	Solvent use and other product use
07	Road transport
08	Other mobile sources and machinery
09	Waste treatment and disposal
10	Agriculture

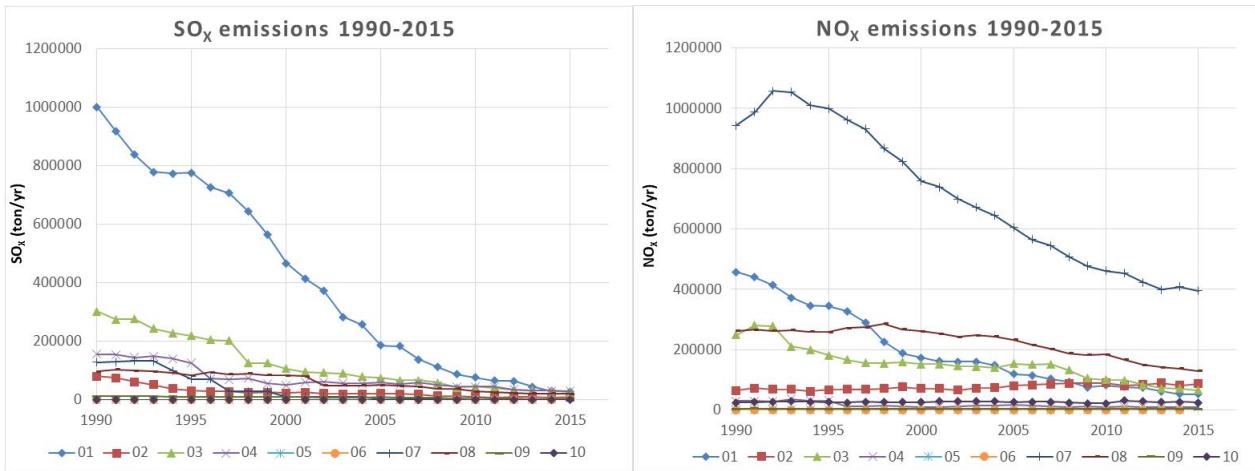


Figure S5 – Emission trend for SO_x (on the left) and NO_x (on the right) by SNAP sector for the period 1990-2015 elaborated for the national official submission 2017 used for the simulations of the MINNI model.

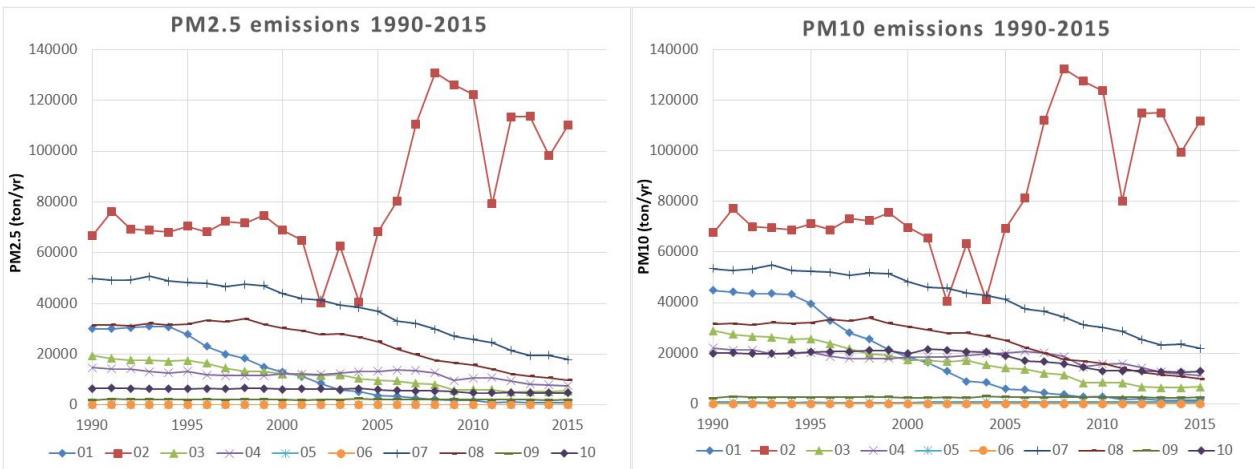


Figure S6 – Emission trend for PM2.5 (on the left) and PM10 (on the right) by SNAP sector for the period 1990-2015 elaborated for the national official submission 2017 used for the simulations of the MINNI model.

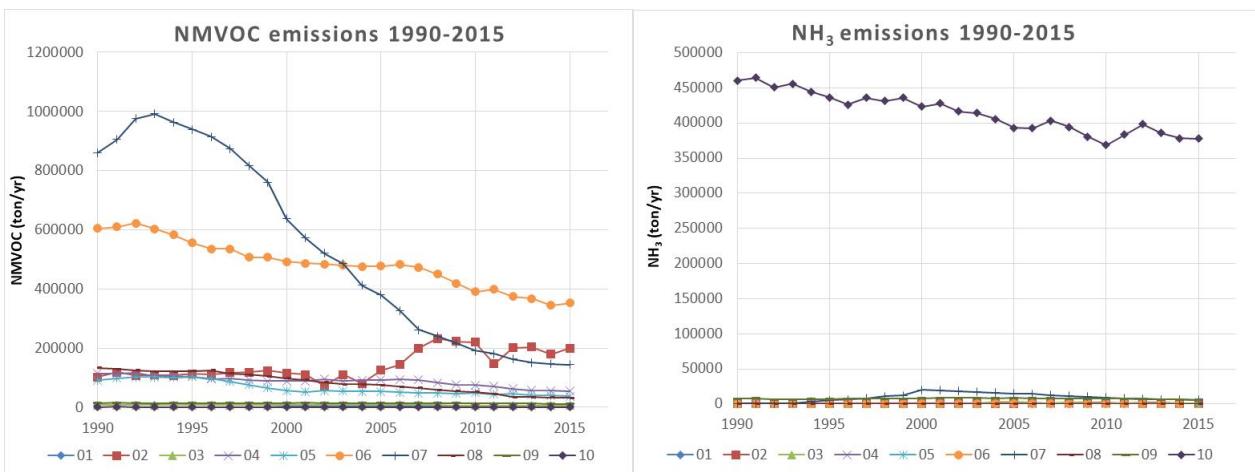


Figure S7 – Emission trend for NMVOC (on the left) and NH₃ (on the right) by SNAP sector for the period 1990-2015 elaborated for the national official submission 2017 used for the simulations of the MINNI model.

Supplementary S3 – Meteorological anomalies (2003-2010)

S3.1 Temperature

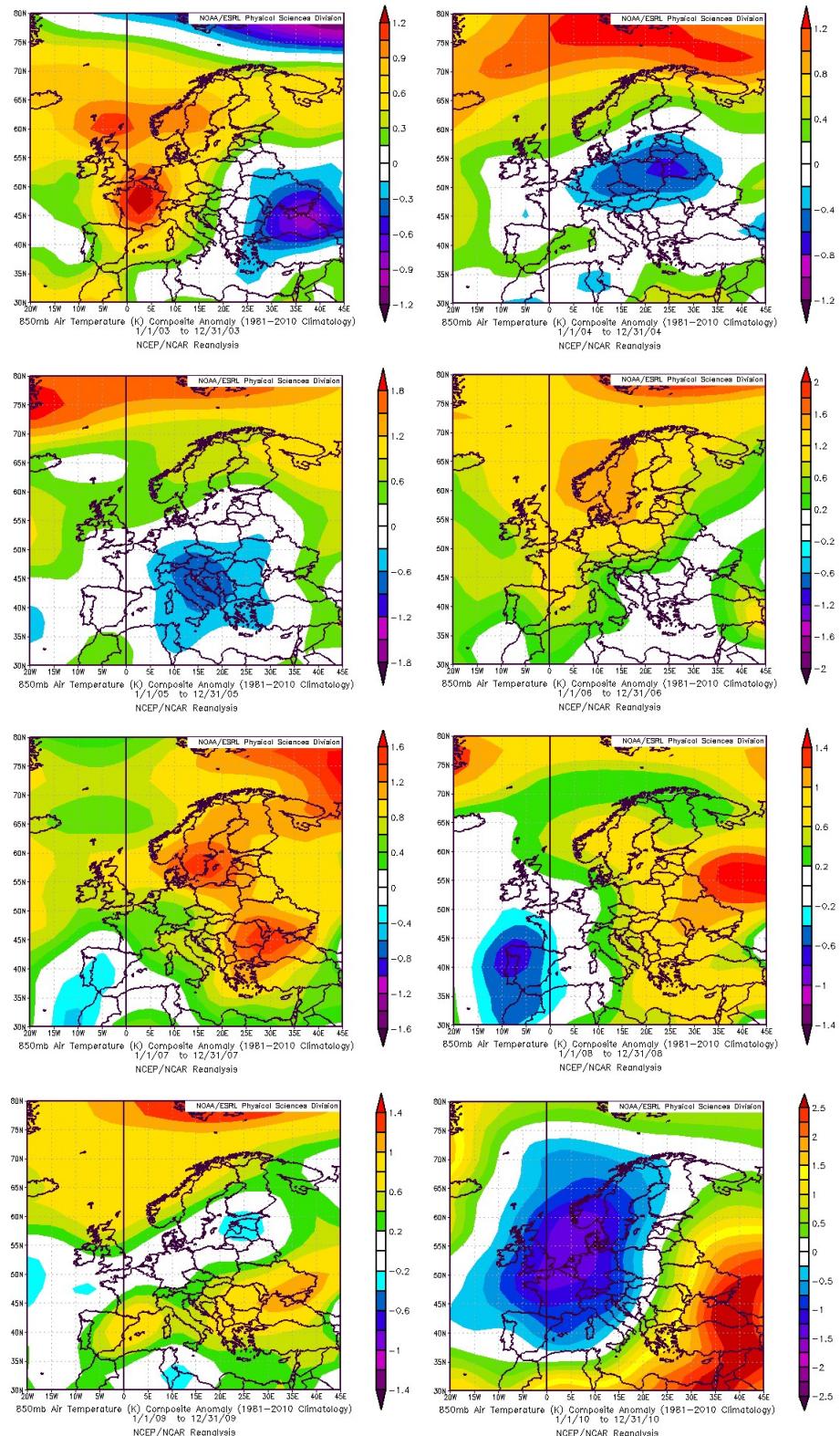


Figure S8 – Temperature (850-hPa) anomalies elaborated from the NCEP/NCAR Reanalysis. Image provided by the NOAA/ESRL Physical Sciences Laboratory, Boulder Colorado from their Web site at <http://psl.noaa.gov/>.

S3.2 500-hPa geopotential height

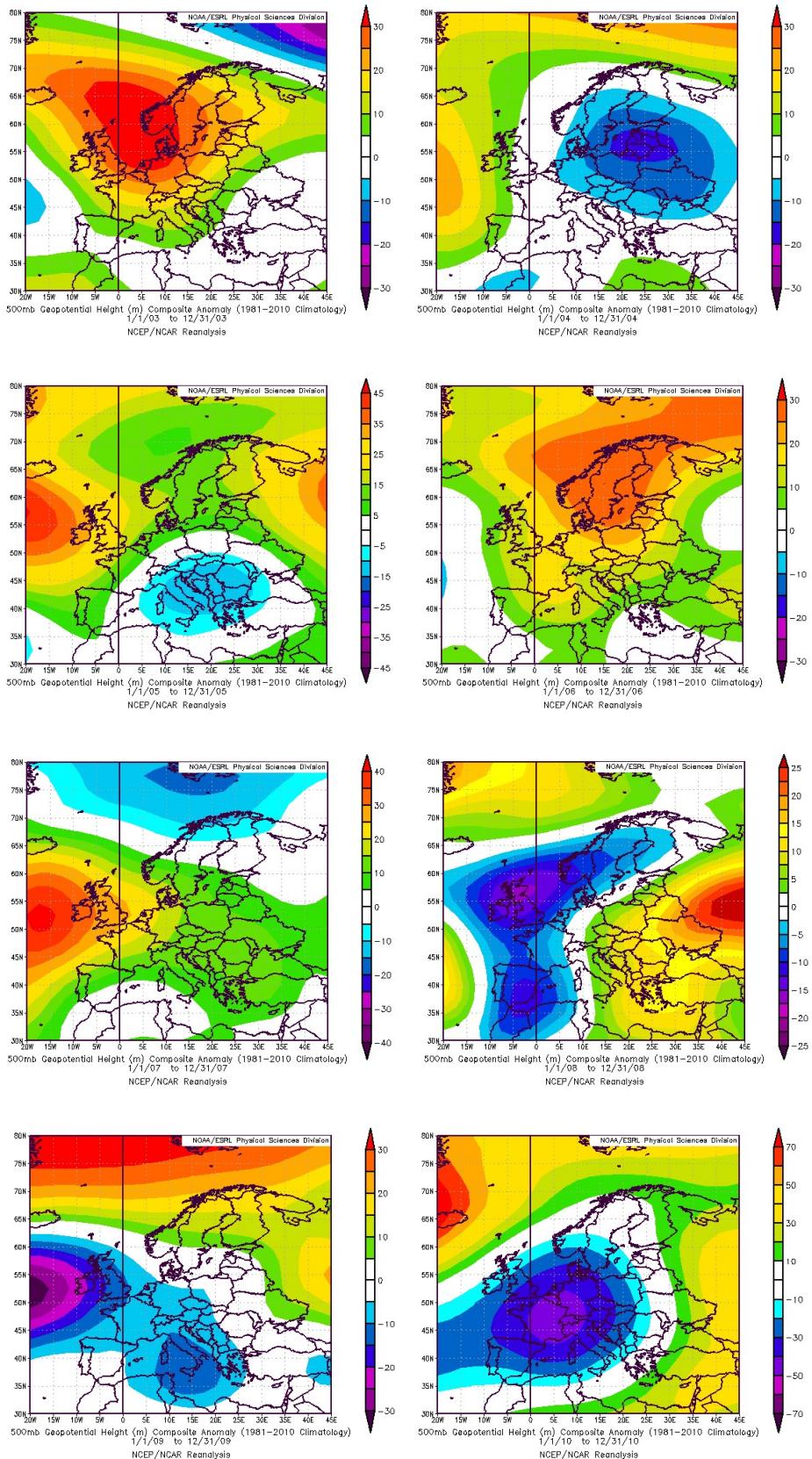


Figure S9 – 500-hPa geopotential height anomalies elaborated from the NCEP/NCAR Reanalysis. Image provided by the NOAA/ESRL Physical Sciences Laboratory, Boulder Colorado from their Web site at <http://psl.noaa.gov/>.

S3.3 Precipitation rate

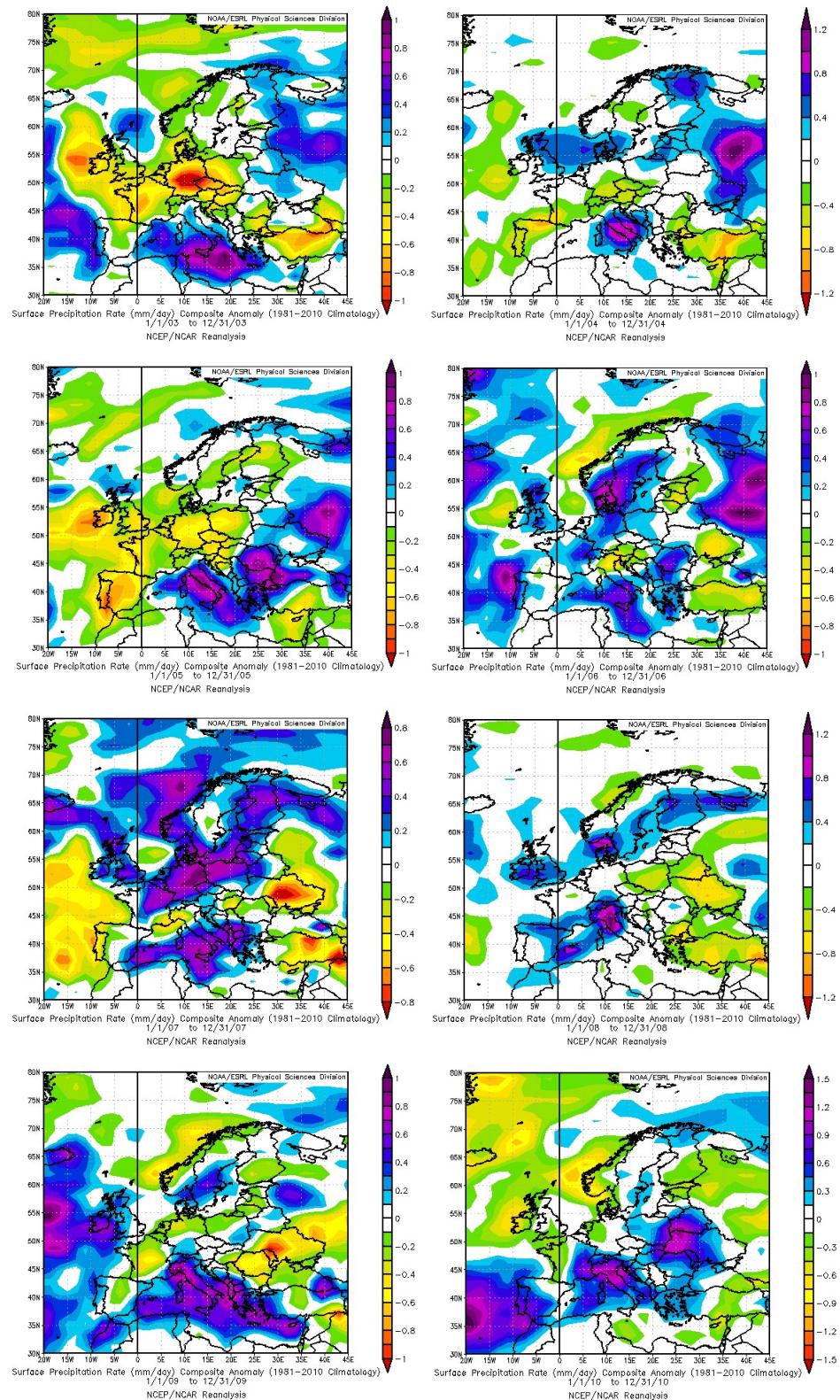


Figure S10 – Precipitation rate anomalies elaborated from the NCEP/NCAR Reanalysis. Image provided by the NOAA/ESRL Physical Sciences Laboratory, Boulder Colorado from their Web site at <http://psl.noaa.gov/>.

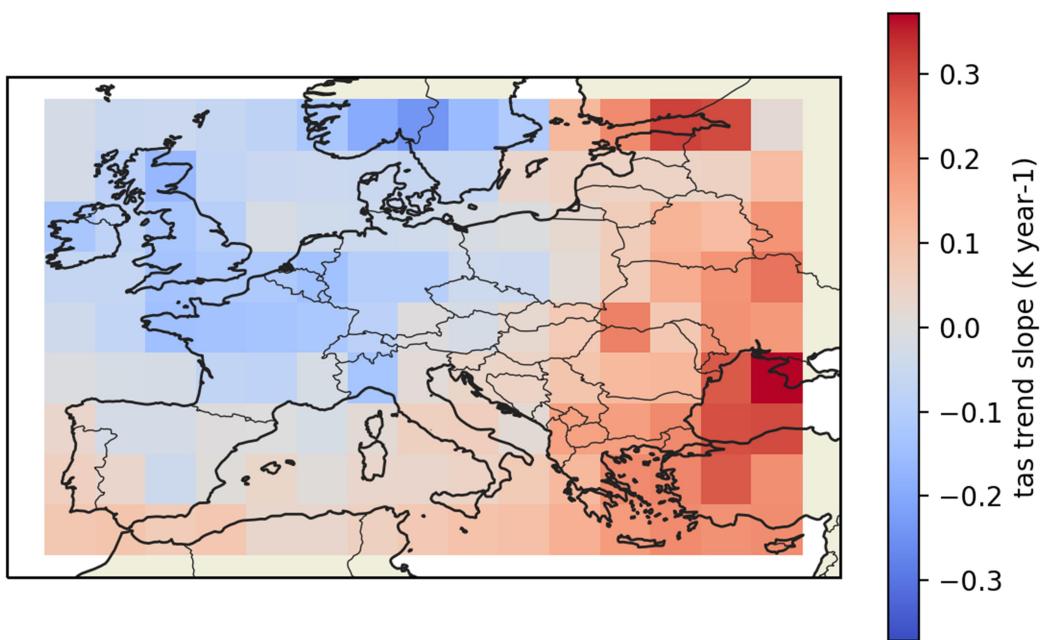


Figure S11 – Near surface temperature trend (K yr^{-1}) for the period 2003-2010 computed from Copernicus Climate Data Service.

Supplementary S4 – Model simulation validation

Definition of model evaluation statistics

The performance statistical indexes used to compare modelled and observed values are reported in the following definitions

MB (Mean bias)

$$MB = \frac{1}{n} \sum_{i=1}^n M_i - O_i$$

RMSE (Root mean squared error)

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

corr (correlation coefficient)

$$corr = \frac{1}{(n-1)} \sum_{i=1}^n \left(\frac{M_i - \bar{M}}{\sigma_M} \right) \left(\frac{O_i - \bar{O}}{\sigma_O} \right)$$

where the letters O and M stand for observations and model results, respectively, the subscript i indicates the time step, the overbars indicate the time average over n time intervals, while the symbol σ indicates the standard deviation.

Supplementary S5 – Trend results

S5.1 NO₂

The following table shows the results for NO₂ slopes for all the stations with a significant trend ($p<0.05$) per Region, type and zone all over the Italian territory. The rows in red show different behaviour between observed and modelled data (increasing trend in observations and decreasing one in simulations).

Table S2 – Results of observed and modelled trends for NO₂ concentrations per stations with $p<0.05$.

airbase_code	Region	Type	Zone	range_obs ($\mu\text{g m}^{-3}\text{yr}^{-1}$)	range_sim ($\mu\text{g m}^{-3}\text{yr}^{-1}$)	range%_obs (% yr^{-1})	range%_sim (% yr^{-1})
IT0499A	TRENTINO	background	suburban	0.56 (0.40 - 0.72)	-0.17 (-0.24 - -0.10)	4.04 (2.82 - 5.52)	-4.01 (-5.29 - -2.64)
IT0507A	TRENTINO	traffic	urban	-1.00 (-1.30 - -0.73)	-1.21 (-1.42 - -1.01)	-2.81 (-3.56 - -2.15)	-6.27 (-7.05 - -5.57)
IT0508A	TRENTINO	traffic	urban	-1.05 (-1.45 - -0.70)	-0.44 (-0.52 - -0.38)	-4.10 (-5.39 - -2.89)	-5.19 (-5.86 - -4.64)
IT0554A	PIEMONTE	background	urban	-1.17 (-1.89 - -0.44)	-2.09 (-2.63 - -1.64)	-2.13 (-3.25 - -0.82)	-3.98 (-4.79 - -3.20)
IT0591A	TRENTINO	background	urban	-0.84 (-1.16 - -0.53)	-0.75 (-0.95 - -0.60)	-2.40 (-3.12 - -1.59)	-3.43 (-4.16 - -2.84)
IT0702A	EMILIA ROMAGNA	traffic	urban	-2.71 (-3.20 - -2.23)	-0.40 (-0.64 - -0.20)	-4.64 (-5.20 - -3.93)	-1.29 (-2.03 - -0.67)
IT0705A	LOMBARDIA	traffic	urban	-0.92 (-1.59 - -0.47)	-1.14 (-1.66 - -0.69)	-1.55 (-2.56 - -0.81)	-1.79 (-2.52 - -1.12)
IT0770A	LOMBARDIA	background	urban	-1.22 (-1.99 - -0.40)	-0.91 (-1.29 - -0.56)	-2.04 (-3.08 - -0.71)	-1.68 (-2.35 - -1.07)
IT0771A	LOMBARDIA	traffic	urban	-1.86 (-2.51 - -1.09)	-0.91 (-1.18 - -0.69)	-2.51 (-3.30 - -1.52)	-2.74 (-3.42 - -2.16)
IT0775A	LOMBARDIA	background	suburban	-0.85 (-1.32 - -0.39)	-0.11 (-0.22 - -0.03)	-2.80 (-4.12 - -1.37)	-1.32 (-2.44 - -0.38)
IT0777A	LOMBARDIA	traffic	urban	-1.61 (-2.30 - -0.79)	-0.77 (-1.08 - -0.53)	-2.61 (-3.47 - -1.35)	-2.31 (-3.12 - -1.64)
IT0839A	LOMBARDIA	background	urban	-0.54 (-0.90 - -0.17)	-0.38 (-0.62 - -0.22)	-1.50 (-2.37 - -0.48)	-1.27 (-2.00 - -0.74)
IT0854A	LIGURIA	background	urban	-1.37 (-2.17 - -0.52)	-1.64 (-1.87 - -1.40)	-2.83 (-4.11 - -1.17)	-4.31 (-4.79 - -3.78)
IT0861A	TOSCANA	traffic	urban	4.26 (3.29 - 5.22)	-0.61 (-0.97 - -0.32)	6.57 (4.82 - 8.82)	-2.23 (-3.32 - -1.22)
IT0912A	LOMBARDIA	background	urban	1.19 (0.67 - 1.75)	-0.88 (-1.09 - -0.68)	3.27 (1.77 - 5.09)	-2.38 (-2.88 - -1.90)
IT0963A	VENETO	background	urban	-1.17 (-1.64 - -0.61)	-1.24 (-1.46 - -0.99)	-2.99 (-4.11 - -1.67)	-2.98 (-3.38 - -2.43)
IT0966A	FRIULI VENEZIA GIULIA	background	urban	-0.54 (-0.91 - -0.09)	-1.27 (-1.47 - -1.08)	-1.93 (-3.05 - -0.32)	-4.76 (-5.32 - -4.12)
IT0977A	VALLE D'AOSTA	background	rural	-0.68 (-0.96 - -0.39)	-0.07 (-0.12 - -0.04)	-10.04 (-12.29 - -6.68)	-2.33 (-3.56 - -1.18)
IT1005A	LIGURIA	traffic	urban	-1.01 (-1.84 - -0.11)	-0.97 (-1.11 - -0.84)	-2.07 (-3.57 - -0.23)	-4.96 (-5.42 - -4.39)
IT1010A	LOMBARDIA	background	urban	-1.65 (-2.55 - -0.87)	-0.75 (-1.03 - -0.42)	-3.26 (-4.61 - -1.83)	-1.83 (-2.41 - -1.05)
IT1098A	FRIULI VENEZIA GIULIA	traffic	suburban	-1.06 (-1.83 - -0.20)	-0.81 (-0.96 - -0.68)	-2.49 (-3.86 - -0.49)	-3.76 (-4.30 - -3.27)
IT1104A	LOMBARDIA	traffic	urban	-4.92 (-6.20 - -3.77)	-1.00 (-1.23 - -0.76)	-6.95 (-8.13 - -5.69)	-2.64 (-3.20 - -2.07)
IT1120A	PIEMONTE	background	suburban	-0.91 (-1.41 - -0.51)	-1.98 (-2.31 - -1.63)	-2.07 (-3.02 - -1.23)	-4.73 (-5.26 - -4.02)
IT1125A	PIEMONTE	traffic	urban	-1.06 (-1.54 - -0.48)	-1.57 (-1.93 - -1.22)	-2.50 (-3.46 - -1.20)	-4.36 (-5.16 - -3.51)
IT1143A	LIGURIA	traffic	urban	-0.76 (-1.13 - -0.34)	-1.48 (-1.67 - -1.32)	-2.14 (-3.09 - -1.01)	-5.35 (-5.76 - -4.88)
IT1153A	LOMBARDIA	traffic	urban	-0.99 (-1.35 - -0.68)	-0.54 (-0.72 - -0.44)	-2.48 (-3.27 - -1.75)	-1.99 (-2.55 - -1.65)
IT1174A	LOMBARDIA	background	rural	-1.01 (-1.34 - -0.68)	-0.50 (-0.73 - -0.30)	-2.91 (-3.74 - -2.04)	-1.70 (-2.41 - -1.04)
IT1176A	LAZIO	background	urban	-0.93 (-1.47 - -0.40)	-1.55 (-1.90 - -1.20)	-1.71 (-2.63 - -0.76)	-2.95 (-3.47 - -2.38)
IT1177A	VENETO	background	urban	-1.37 (-1.66 - -1.06)	-0.67 (-0.96 - -0.39)	-3.13 (-3.69 - -2.52)	-2.11 (-2.92 - -1.28)
IT1203A	LOMBARDIA	background	suburban	-0.95 (-1.32 - -0.59)	-1.02 (-1.39 - -0.70)	-2.66 (-3.56 - -1.72)	-2.51 (-3.29 - -1.77)
IT1207A	SICILIA	traffic	urban	-2.85 (-3.60 - -1.73)	-1.48 (-1.75 - -1.22)	-4.66 (-5.60 - -3.08)	-6.57 (-7.47 - -5.65)
IT1246A	PIEMONTE	background	urban	-0.83 (-1.18 - -0.50)	-0.59 (-0.78 - -0.46)	-2.66 (-3.57 - -1.68)	-4.15 (-5.21 - -3.39)
IT1247A	PIEMONTE	background	urban	-0.55 (-1.03 - -0.12)	-0.52 (-0.68 - -0.37)	-1.61 (-2.79 - -0.37)	-3.63 (-4.50 - -2.65)
IT1286A	LOMBARDIA	traffic	urban	-1.44 (-2.25 - -0.71)	-0.48 (-0.79 - -0.26)	-2.88 (-4.14 - -1.52)	-1.49 (-2.34 - -0.83)
IT1335A	TRENTINO	traffic	urban	-0.52 (-0.88 - -0.20)	-0.78 (-0.95 - -0.60)	-1.41 (-2.27 - -0.56)	-5.38 (-6.25 - -4.41)
IT1336A	VENETO	traffic	urban	-3.27 (-3.87 - -2.74)	-0.59 (-0.81 - -0.37)	-5.28 (-5.95 - -4.68)	-1.91 (-2.54 - -1.25)
IT1409A	TOSCANA	traffic	urban	-0.81 (-1.15 - -0.47)	-0.57 (-0.68 - -0.45)	-1.82 (-2.49 - -1.10)	-2.59 (-3.04 - -2.13)
IT1422A	ABRUZZO	traffic	urban	-2.01 (-3.24 - -0.43)	-1.26 (-1.43 - -1.10)	-2.51 (-3.72 - -0.58)	-6.08 (-6.69 - -5.57)
IT1453A	VENETO	background	urban	-1.36 (-1.81 - -0.84)	-0.89 (-1.11 - -0.71)	-2.78 (-3.45 - -1.82)	-2.45 (-2.99 - -2.01)
IT1459A	LOMBARDIA	background	suburban	-0.95 (-1.46 - -0.37)	-1.32 (-1.70 - -0.97)	-2.31 (-3.35 - -0.96)	-3.16 (-3.92 - -2.43)
IT1463A	LOMBARDIA	background	suburban	0.57 (0.10 - 1.16)	-0.33 (-0.72 - -0.01)	2.01 (0.34 - 4.42)	-0.79 (-1.69 - -0.02)

IT1544A	LIGURIA	industrial	urban	0.96 (0.55 - 1.54)	-1.24 (-1.42 - -1.06)	2.77 (1.50 - 4.86)	-4.72 (-5.21 - -4.20)
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S5.2 PM10

The following table shows the results for PM10 slopes for all the stations with a significant trend ($p<0.05$) per Region, type and zone all over the Italian territory.

Table S3 – Results of observed and modelled trends for PM10 concentrations per stations with $p<0.05$.

airbase_code	Region	Type	Zone	range_obs ($\mu\text{g m}^{-3}\text{yr}^{-1}$)	range_sim ($\mu\text{g m}^{-3}\text{yr}^{-1}$)	range%_obs (% yr^{-1})	range%_sim (% yr^{-1})
IT0559A	LOMBARDIA	industrial	suburban	-2.59 (-3.14 - -2.17)	-0.81 (-1.37 - -0.20)	-5.08 (-5.88 - -4.43)	-2.11 (-3.38 - -0.55)
IT0705A	LOMBARDIA	traffic	urban	-1.85 (-2.30 - -1.44)	-1.26 (-2.04 - -0.48)	-3.42 (-4.16 - -2.71)	-2.36 (-3.58 - -0.94)
IT0707A	LOMBARDIA	background	urban	-2.25 (-2.80 - -1.68)	-0.98 (-1.53 - -0.42)	-4.53 (-5.38 - -3.53)	-2.61 (-3.92 - -1.19)
IT0709A	LOMBARDIA	background	urban	-1.99 (-2.83 - -1.09)	-0.94 (-1.41 - -0.37)	-3.69 (-4.97 - -2.18)	-2.75 (-3.87 - -1.16)
IT0776A	LOMBARDIA	traffic	urban	-1.86 (-2.23 - -1.44)	-0.79 (-1.09 - -0.47)	-4.56 (-5.23 - -3.71)	-3.26 (-4.28 - -2.08)
IT0940A	EMILIA ROMAGNA	background	urban	-0.72 (-1.21 - -0.05)	-1.19 (-1.64 - -0.67)	-1.99 (-3.24 - -0.14)	-3.36 (-4.45 - -2.05)
IT0953A	LAZIO	background	urban	-0.60 (-0.98 - -0.11)	-0.55 (-0.86 - -0.09)	-1.98 (-3.04 - -0.37)	-1.90 (-2.85 - -0.31)
IT1079A	SICILIA	traffic	urban	-0.88 (-1.26 - -0.45)	-0.93 (-1.17 - -0.66)	-2.22 (-3.06 - -1.17)	-3.91 (-4.68 - -2.96)
IT1104A	LOMBARDIA	traffic	urban	-1.56 (-2.11 - -1.00)	-1.31 (-1.78 - -0.84)	-3.24 (-4.22 - -2.14)	-3.55 (-4.59 - -2.41)
IT1159A	EMILIA ROMAGNA	traffic	urban	-1.21 (-1.70 - -0.82)	-0.65 (-1.18 - -0.16)	-2.74 (-3.63 - -1.94)	-2.08 (-3.48 - -0.55)
IT1247A	PIEMONTE	background	urban	-1.51 (-1.98 - -1.02)	-0.68 (-1.09 - -0.21)	-4.15 (-5.09 - -3.00)	-3.48 (-5.20 - -1.20)

S5.3 O₃

The following table shows the results for O₃-MDA8 (Apr/Sep) slopes for all the stations with a significant trend ($p<0.05$) per Region, type and zone all over the Italian territory. The rows in red show different behaviour between observed and modelled data (increasing trend in observations and decreasing one in simulations).

Table S4 – Results of observed and modelled trends for O₃-MDA8 (Apr/Sep) concentrations per stations with $p<0.05$.

airbase_code	Region	Type	Zone	range_obs ($\mu\text{g m}^{-3}\text{yr}^{-1}$)	range_sim ($\mu\text{g m}^{-3}\text{yr}^{-1}$)	range%_obs (% yr^{-1})	range%_sim (% yr^{-1})
IT0499A	TRENTINO	background	suburban	1.10 (0.21 - 2.10)	-0.67 (-1.01 - -0.56)	1.33 (0.24 - 2.68)	-0.79 (-1.18 - -0.66)
IT0505A	TRENTINO	background	rural	-1.45 (-1.91 - -0.98)	-0.30 (-0.71 - -0.17)	-1.13 (-1.47 - -0.78)	-0.33 (-0.77 - -0.19)
IT0508A	TRENTINO	traffic	urban	1.54 (1.06 - 2.01)	-0.56 (-0.84 - -0.38)	1.98 (1.34 - 2.63)	-0.63 (-0.94 - -0.43)
IT0858A	LIGURIA	background	urban	2.07 (1.05 - 3.59)	-0.49 (-0.97 - -0.03)	2.12 (1.03 - 3.92)	-0.40 (-0.78 - -0.02)
IT0867A	LAZIO	traffic	urban	3.67 (2.20 - 5.88)	-0.79 (-1.23 - -0.50)	4.82 (2.58 - 9.30)	-0.75 (-1.14 - -0.48)
IT0883A	TOSCANA	background	rural	-1.08 (-1.70 - -0.46)	-0.79 (-1.22 - -0.40)	-0.95 (-1.46 - -0.42)	-0.69 (-1.04 - -0.35)
IT0888A	LAZIO	industrial	suburban	-3.27 (-3.98 - -2.36)	-1.06 (-1.70 - -0.52)	-3.26 (-3.87 - -2.41)	-0.94 (-1.48 - -0.47)
IT0948A	TOSCANA	background	urban	1.19 (0.27 - 2.05)	-0.70 (-1.22 - -0.24)	1.18 (0.26 - 2.14)	-0.60 (-1.02 - -0.21)
IT0953A	LAZIO	background	urban	-3.39 (-4.54 - -2.44)	-1.40 (-2.34 - -0.73)	-3.00 (-3.87 - -2.24)	-1.23 (-1.99 - -0.66)
IT0988A	VALLE D'AOSTA	background	rural	-4.64 (-6.23 - -2.40)	-0.62 (-1.17 - -0.19)	-3.80 (-4.82 - -2.17)	-0.65 (-1.21 - -0.20)
IT0989A	LAZIO	background	rural	-6.92 (-7.64 - -6.45)	-0.58 (-0.71 - -0.47)	-4.31 (-4.69 - -4.11)	-0.60 (-0.74 - -0.49)
IT1017A	LOMBARDIA	background	suburban	-2.51 (-3.41 - -1.58)	-1.74 (-2.84 - -0.93)	-2.05 (-2.74 - -1.34)	-1.42 (-2.25 - -0.78)
IT1048A	EMILIA ROMAGNA	background	urban	-2.60 (-3.57 - -1.55)	-1.43 (-2.10 - -0.85)	-2.28 (-3.05 - -1.40)	-1.20 (-1.72 - -0.73)
IT1110A	TOSCANA	background	urban	-0.86 (-1.36 - -0.21)	-1.15 (-1.65 - -0.68)	-0.86 (-1.34 - -0.22)	-0.94 (-1.32 - -0.57)
IT1174A	LOMBARDIA	background	rural	-3.52 (-5.04 - -2.32)	-0.66 (-1.63 - -0.09)	-2.74 (-3.74 - -1.87)	-0.54 (-1.31 - -0.08)
IT1182A	UMBRIA	traffic	urban	1.42 (0.68 - 2.23)	-0.79 (-1.23 - -0.36)	1.53 (0.71 - 2.51)	-0.73 (-1.11 - -0.34)
IT1191A	TRENTINO	background	rural	-1.62 (-2.89 - -0.20)	-0.70 (-1.18 - -0.30)	-1.24 (-2.17 - -0.16)	-0.68 (-1.12 - -0.30)
IT1203A	LOMBARDIA	background	suburban	-4.81 (-5.80 - -4.14)	-0.94 (-1.90 - -0.16)	-3.33 (-3.90 - -2.91)	-0.78 (-1.52 - -0.14)
IT1216A	FRIULI VENEZIA GIULIA	background	suburban	2.40 (1.71 - 3.72)	-0.89 (-1.65 - -0.29)	2.94 (2.01 - 4.86)	-0.76 (-1.38 - -0.25)
IT1236A	TOSCANA	background	rural	-5.29 (-5.98 - -4.32)	-0.79 (-1.41 - -0.24)	-4.13 (-4.62 - -3.50)	-0.65 (-1.14 - -0.20)
IT1246A	PIEMONTE	background	urban	-2.03 (-3.16 - -0.89)	-0.67 (-1.27 - -0.21)	-1.63 (-2.46 - -0.74)	-0.60 (-1.13 - -0.19)
IT1247A	PIEMONTE	background	urban	-4.38 (-5.38 - -3.40)	-0.94 (-1.50 - -0.50)	-3.36 (-3.95 - -2.68)	-0.87 (-1.36 - -0.47)
IT1279A	SARDEGNA	industrial	suburban	-2.45 (-3.66 - -1.31)	-0.84 (-1.32 - -0.43)	-2.32 (-3.34 - -1.29)	-0.73 (-1.14 - -0.38)
IT1397A	SARDEGNA	industrial	suburban	-1.80 (-2.99 - -0.47)	-0.87 (-1.31 - -0.52)	-1.81 (-2.90 - -0.49)	-0.77 (-1.13 - -0.46)
IT1427A	SARDEGNA	traffic	urban	-1.51 (-2.72 - -0.66)	-0.38 (-0.50 - -0.35)	-1.60 (-2.76 - -0.74)	-0.40 (-0.51 - -0.36)
IT1474A	FRIULI VENEZIA GIULIA	background	rural	2.61 (0.78 - 4.60)	-0.79 (-1.49 - -0.19)	2.55 (0.71 - 4.93)	-0.67 (-1.24 - -0.17)
IT1478A	FRIULI VENEZIA GIULIA	background	rural	-3.83 (-5.57 - -2.17)	-0.56 (-1.21 - -0.13)	-3.17 (-4.39 - -1.93)	-0.55 (-1.16 - -0.13)
IT1515A	PIEMONTE	background	suburban	-3.23 (-4.45 - -1.82)	-0.66 (-0.81 - -0.58)	-2.61 (-3.50 - -1.54)	-0.68 (-0.83 - -0.60)
IT1519A	PIEMONTE	background	rural	-2.84 (-3.34 - -2.41)	-1.14 (-1.59 - -0.86)	-2.17 (-2.53 - -1.89)	-1.00 (-1.39 - -0.76)
IT1576A	SARDEGNA	industrial	suburban	2.29 (0.89 - 3.37)	-1.04 (-1.46 - -0.57)	2.96 (1.08 - 4.80)	-0.86 (-1.19 - -0.48)