

Supplementary materials for "Determination of weather-driven trends of atmospheric emissions, concentrations and deposition over Europe during the last decades"

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S1 Biogenic emission maps over Europe

Figures S1 to S4 provides maps of summertime (June to August) biogenic emissions for TVOC (sum of all emitted VOC), isoprene, monoterpenes and sesquiterpenes. Emissions are calculated with the Big-Leaf approach of CHIMERE 2020 (Menut et al., 2021) or with the canopy approach using either MEGAN 2.1 Emissions Factors (EF) (Guenther et al., 2012), MEGAN 5 3.2 EF (Guenther, 2024) or ORCHIDEE EF (Messina et al., 2016). In general, the spatial distribution of biogenic emissions are similar between the different approaches but the level of emissions differ strongly for specific VOC compounds.

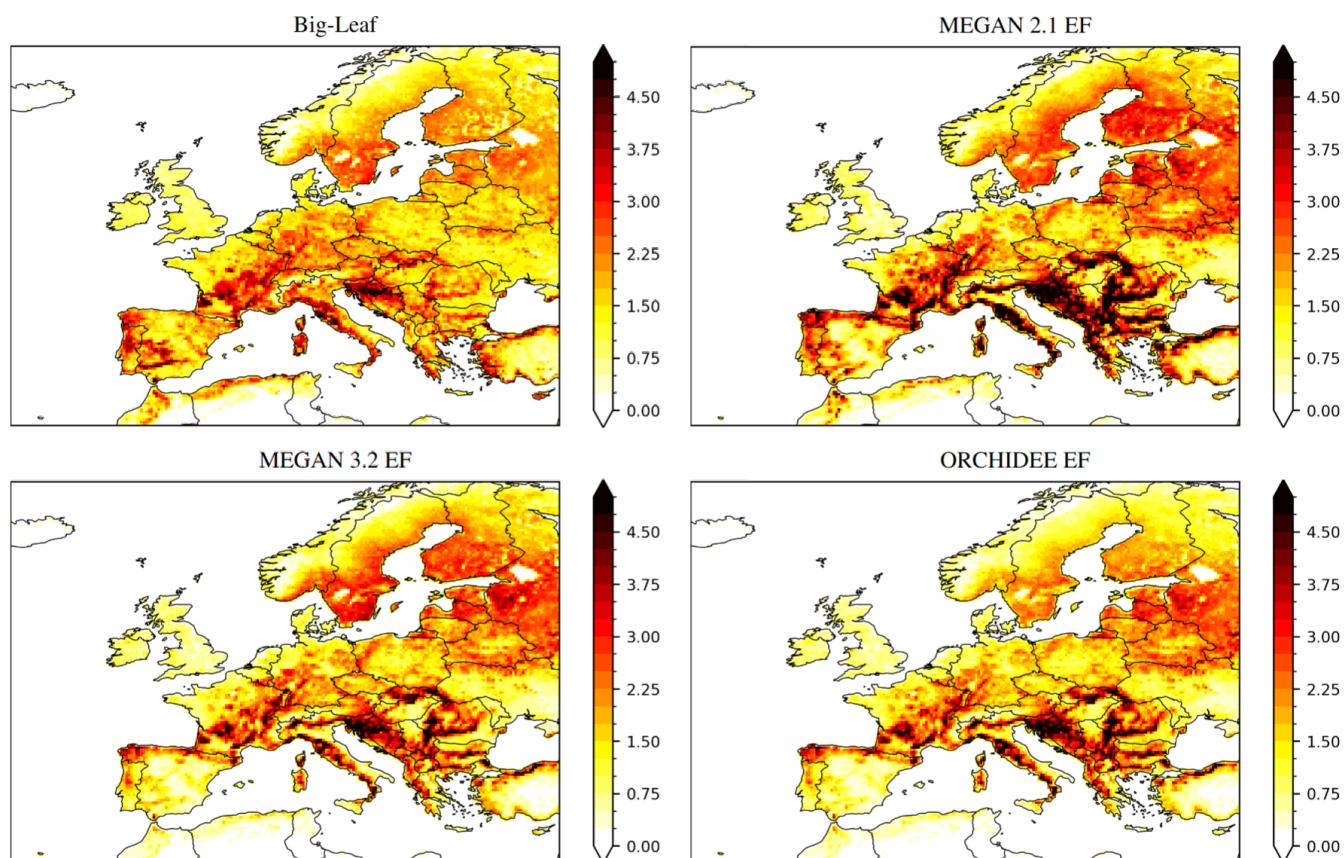


Figure S1. Maps of the sum of summertime total biogenic VOC emissions (in kt/cell) for year 2018.

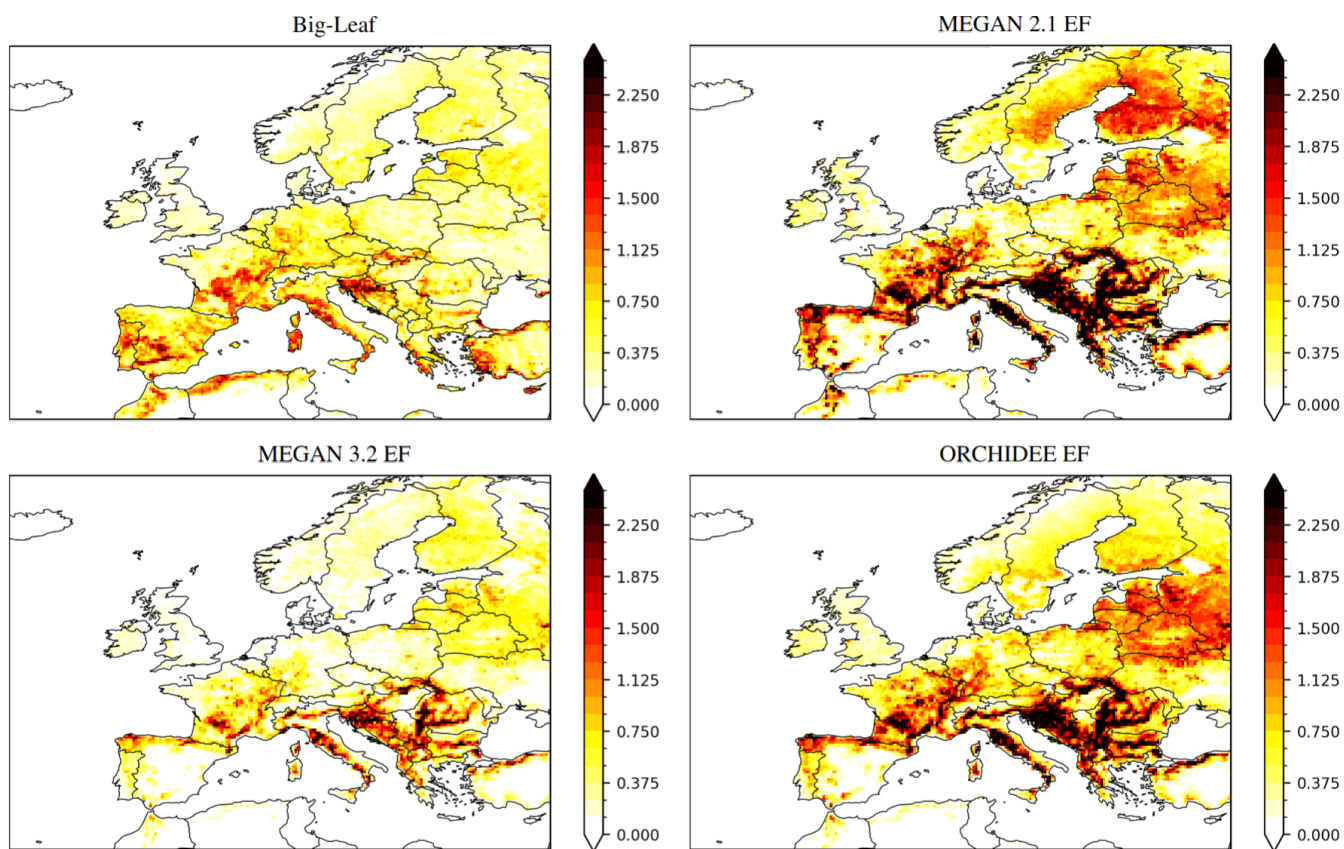


Figure S2. Maps of isoprene summertime biogenic emissions (in kT/cell) for year 2018.

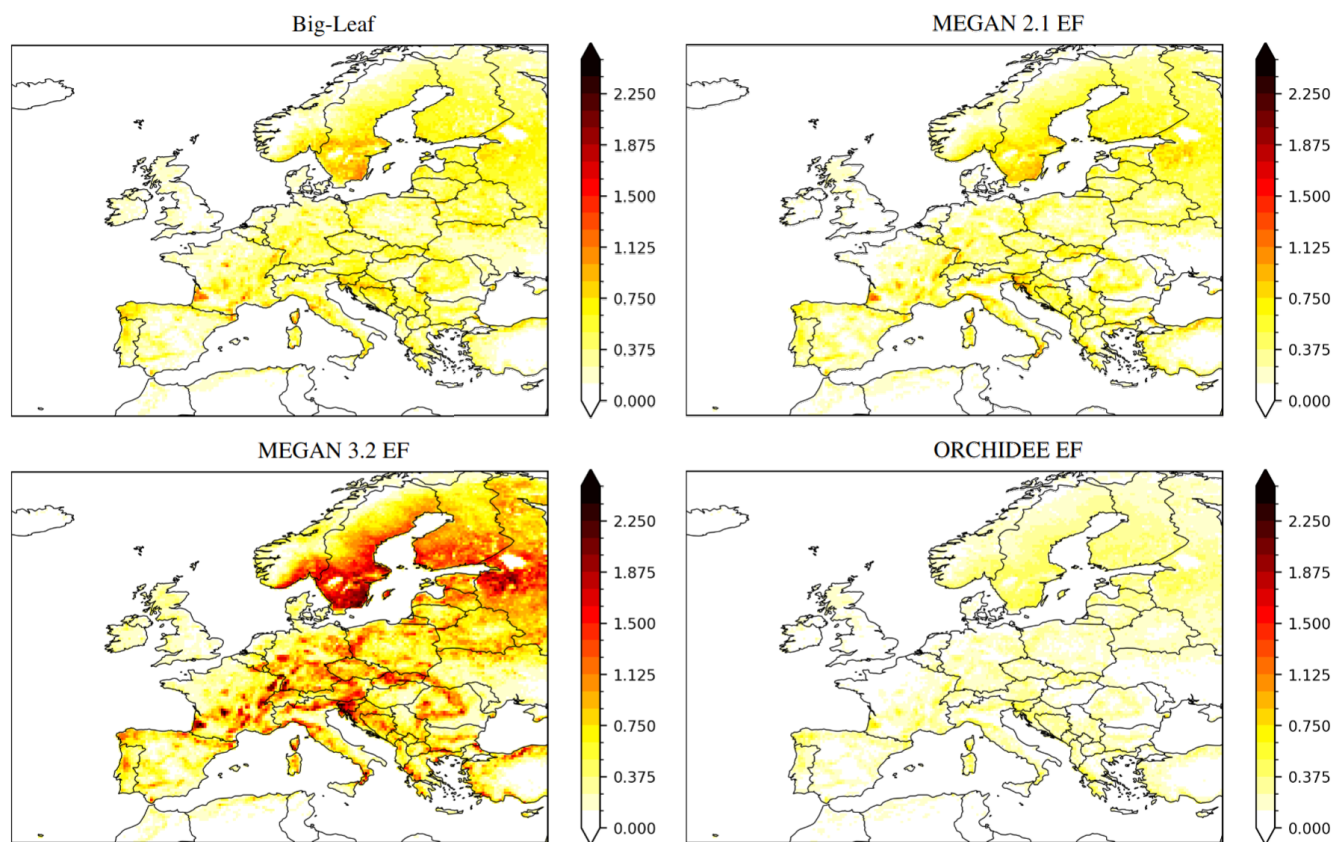


Figure S3. Maps of monoterpene summertime biogenic emissions (in kT/cell) for year 2018.

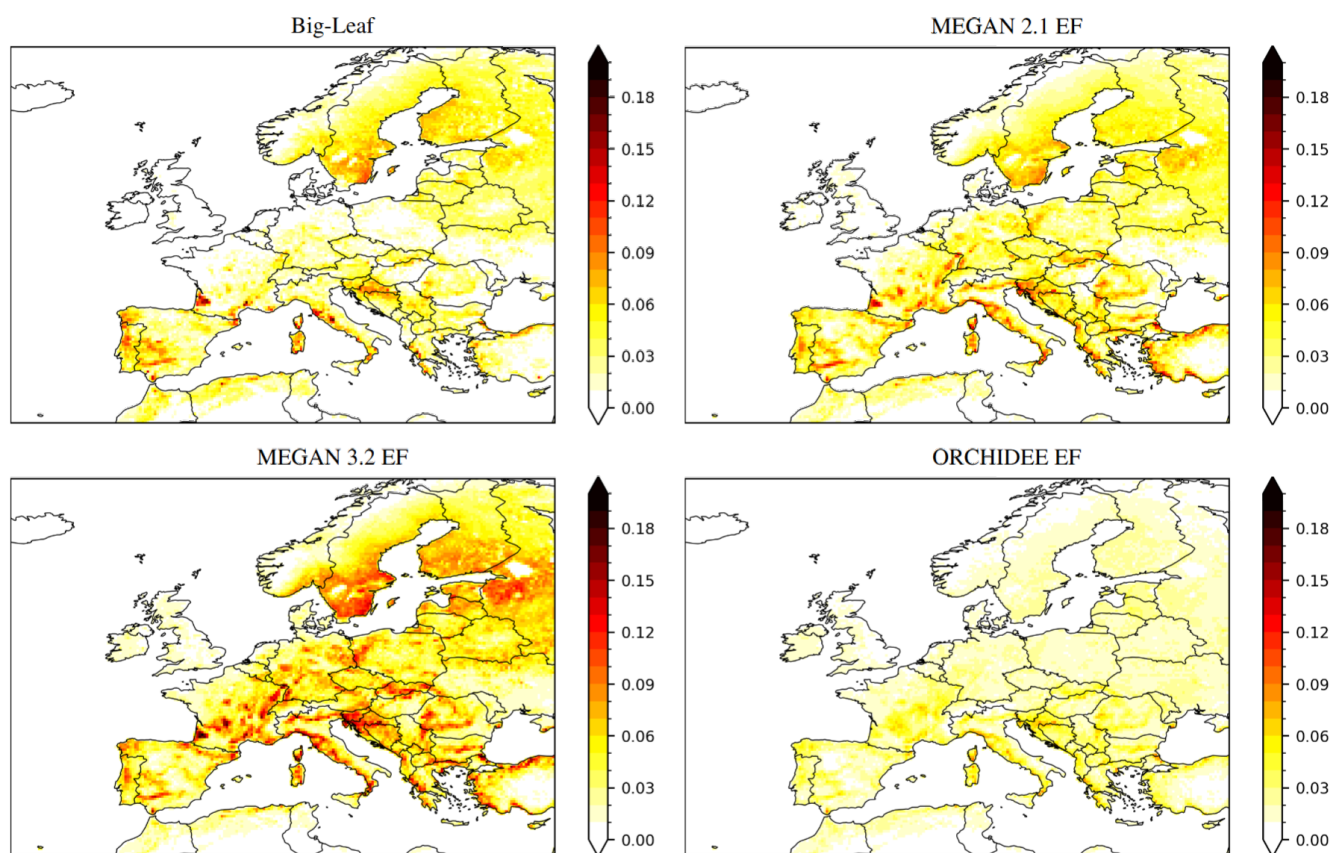


Figure S4. Maps of sesquiterpene summertime biogenic emissions (in kT/cell) for year 2018.

S2 Evaluation of CHIMERE simulations

Several simulations are performed over year 2018 to estimate biogenic emissions and the model performance with several configurations:

- 10 – 1 simulation with the original CHIMERE v2020r1 with meteorologically independent emissions.
- 3 simulations with the new CHIMERE version using meteorologically independent emissions, with the default LAI and using MEGAN 2.1, MEGAN 3.2 or ORCHIDEE biogenic EF.
- 1 simulation with MEGAN 3.2 EF and meteorologically independent emissions but LAI from HiQLAI.

Simulations are performed for the 1990-2022 period over a $0.25^{\circ} \times 0.25^{\circ}$ domain covering Europe. Emission calculations
15 and CHIMERE simulations are performed using meteorological data from the ERA5 reanalysis (Hersbach et al., 2020).

Simulations performed with the CHIMERE version developed in this study are carried out with the H²O SOA scheme including SOA formation from monoterpene oxidation by autoxidation (Chrit et al., 2017; Sartelet et al., 2020). Following Couvidat et al. (2018), anthropogenic primary organic aerosols are assumed to be semivolatile. Simulations performed with
the original CHIMERE 2020 use the default configuration of CHIMERE with the SOA scheme of Bessagnet et al. (2008) and
20 assume nonvolatile POA.

S2.1 Evaluation of biogenic emissions

The biogenic emissions simulated over Europe under the different configurations are illustrated in Fig. S5, which shows total biogenic VOC emissions (TVOC; sum of all emitted VOC) as well as the emissions of the most important SOA precursors (isoprene, monoterpenes and sesquiterpenes). Maps of annual biogenic emissions are shown in the Supplementary Materials.

25 Simulated annual TVOC emissions over EUR are similar between the different configurations with the highest estimate obtained with the canopy approach using MEGAN2.1 EF (9600 kT). Other estimates range between 7500 kT and 8400 kT. While TVOC estimates are similar between the different approaches, the emission speciation differs widely. Isoprene emissions range from 1750 kT (canopy approach with MEGAN 3.2) to 4050 kT (canopy approach with MEGAN 2.1). On the contrary, monoterpenes and sesquiterpenes are much higher with MEGAN 3.2 EFs (2990 kT and 215 kT, respectively) than with the
30 other EF databases (between 920 and 1670 kT for monoterpenes and between 96 and 160 kT for sesquiterpenes).

The higher terpene emissions and lower isoprene emissions obtained with MEGAN 3.2 EF are consistent with other studies over Europe that indicated a strong underestimation (by a factor 3) of terpene emissions and overestimation of isoprene emissions (by a similar factor) (Jiang et al., 2019; Ciccioli et al., 2023). Based on these studies, Wang et al. (2024) increased the emissions of monoterpenes and sesquiterpenes simulated by CHIMERE v2020r1 by a factor 3 and decreased isoprene emissions by the same factor. The authors obtained significantly improved simulation results for summertime PM_{2.5} and organic
35 aerosols, as evidenced by comparisons with measurements. According to our results, MEGAN 3.2 could be considered the most up-to-date and realistic EF database.

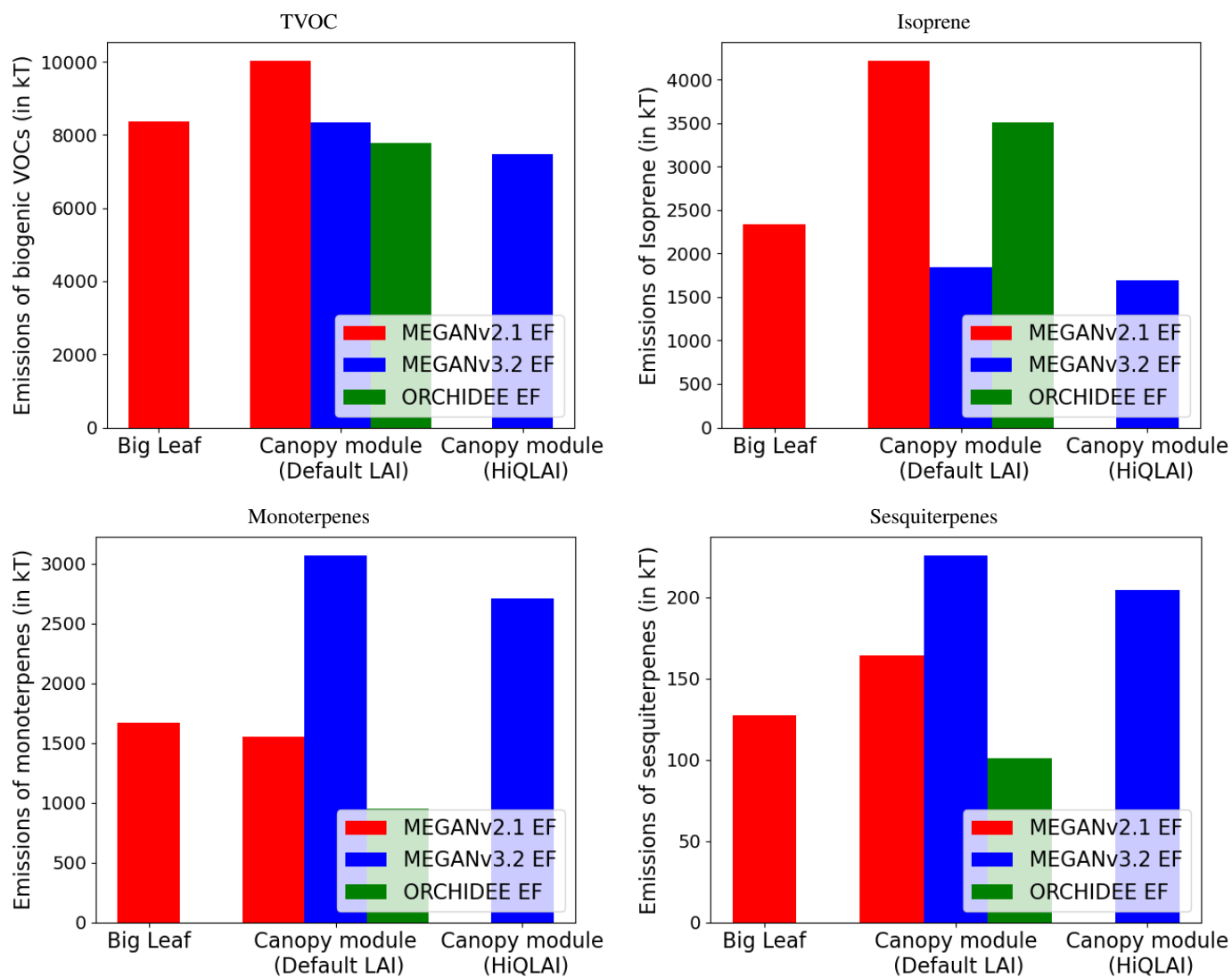


Figure S5. Simulated biogenic emissions cumulated over EUR for year 2018 (in kT) with the "big-leaf" and the canopy approaches.

Changing the LAI database also affects emissions. As the average LAI from the HiQLAI database is lower by around 8% over EUR than the default LAI, biogenic TVOC emissions estimated when using the HiQLAI database are lower by 10% over EUR. However, the HiQLAI combined to GlobMAP database is the only database covering the whole simulation period.

S2.2 Model performance evaluation

The effect of the different model improvements on performance is assessed by comparisons to in-situ measurements of surface concentrations reported by the European air quality observation network AQ e-Reporting (EEA, 2023). This database gathers observations provided by the different member countries of the European Environment Agency (EEA). Only the background rural stations are selected because of the resolution of simulations which is not sufficient to represent appropriately urban areas. The performance of simulations are evaluated based on the Root Mean Square Error (RMSE), the averaged bias, and on the correlation.

Fig. S6 illustrates, for daily O_3 and $PM_{2.5}$ concentrations, the comparison of the model results with observations for the original CHIMERE version with non-dynamic emissions (referred to as "CHIMERE 2020") and the modified version with dynamic emissions computed as a function of meteorology (referred to as "final setup"). Fig. S7 shows the simulated temporal evolution of daily maximum O_3 concentrations. Fig. S8 shows the simulated temporal evolution of daily PM_{10} and NO_2 concentrations. Scores (Mean bias, RMSE, correlation) are shown in Figures S9 to S13 for daily maximum O_3 , daily averaged O_3 , $PM_{2.5}$, PM_{10} and NO_2 . In general, "final setup" has better results than "CHIMERE 2020".

The seasonal cycle of daily averaged O_3 concentrations is more accurately reproduced with "final setup". While "CHIMERE 2020" tends to overestimate daily averaged O_3 concentrations by around $7.0 \mu g/m^3$ from July to November, the averaged bias is only of $1.1 \mu g/m^3$ with "final setup" for the same period. However, all simulations show a large underestimation of daily averaged O_3 concentrations during April and May (mean bias equal to $-3.5 \mu g/m^3$ with "CHIMERE 2020" and to $-9.1 \mu g/m^3$ with "final setup"). This underestimation could be potentially due to a lack of NO_x emissions during this period. This underestimation of concentrations during this specific period leads to a compensation effect for "CHIMERE 2020". The averaged bias is equal to $1.8 \mu g/m^3$ and $-2.1 \mu g/m^3$ for "CHIMERE 2020" and "final setup", respectively. The RMSE and correlation are similar between the two CHIMERE versions (RMSE equal to 16.4 and $16.2 \mu g/m^3$ for "CHIMERE 2020" and "final setup", respectively; correlations are equal to 0.79 for both simulations).

Similar results are obtained for daily maximum O_3 concentrations with the two CHIMERE versions (RMSE and correlation around 16.8 and 0.82 for both versions, respectively). Slightly better results are obtained during summer with "final setup" (mean biases around 2.4 and $2.1 \mu g/m^3$ with "CHIMERE 2020" and "final setup", respectively). However, due to an underestimation of concentrations during April and May (mean biases around -8.5 and $-10.0 \mu g/m^3$ with "CHIMERE 2020" and "final setup", respectively), slightly better biases are found on annual averages with "CHIMERE 2020" (mean biases around -1.1 and $-2.6 \mu g/m^3$ with "CHIMERE 2020" and "final setup", respectively).

No significant impact on RMSE or correlation is found for O_3 concentrations due to the choice of biogenic emission factors or the use of dynamic anthropogenic emissions (with changes in RMSE and correlation around $0.3 \mu g/m^3$ and 0.01 , respectively). However, small differences in the simulated bias are found. Using the default LAI, annual biases are lower using

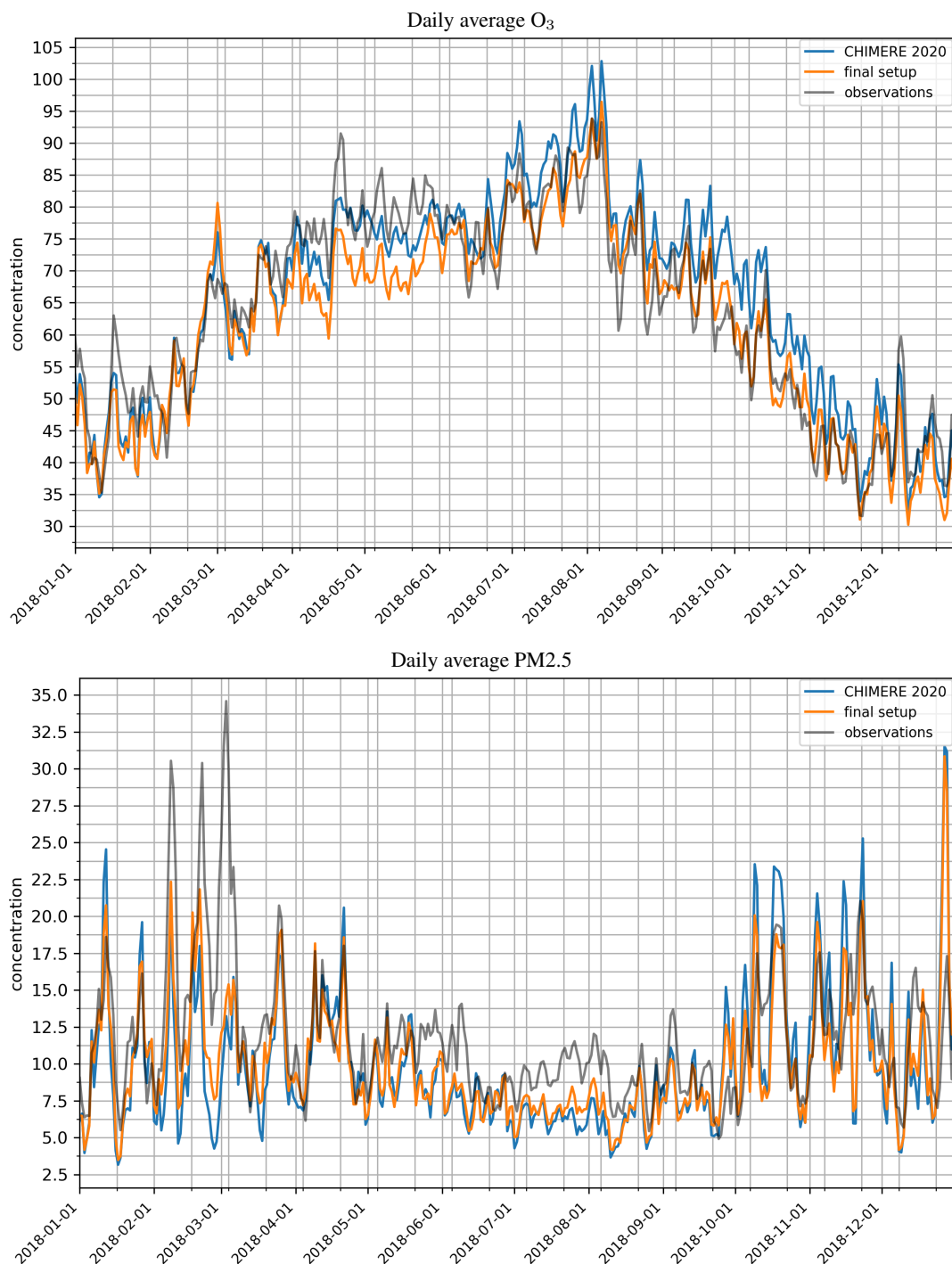


Figure S6. Temporal evolution of O₃ and PM_{2.5} daily concentrations (in $\mu\text{g}/\text{m}^3$) averaged over stations for measurements, the original CHIMERE version with non-dynamic emissions (referred to as "CHIMERE 2020") and the modified version with dynamic emissions computed as a function of meteorology (referred to as "final setup").

MEGAN 2.1 EFs ($-1.5 \mu\text{g}/\text{m}^3$ for daily averages and $-1.6 \mu\text{g}/\text{m}^3$ for daily maximum) and higher with MEGAN 3.2 EFs ($-2.1 \mu\text{g}/\text{m}^3$ for daily averages and $-2.8 \mu\text{g}/\text{m}^3$ for daily maximum). This feature is due to higher biogenic TVOC emissions using MEGAN 2.1 EFs. Even though TVOC emissions using HiQLAI are lower, simulated concentrations are slightly higher when
75 using HiQLAI (average increase by 0.6 and $0.4 \mu\text{g}/\text{m}^3$ on daily averages and daily maximum, respectively). This is due to lower O_3 deposition velocities for lower LAI values.

Particle concentrations are generally better simulated with the “final setup”, as indicated by improved performance metrics (RMSE of $8.1 \mu\text{g}/\text{m}^3$ and $10.6 \mu\text{g}/\text{m}^3$ for PM_{2.5} and PM₁₀, and correlations of 0.62 and 0.57, respectively) compared to “CHIMERE 2020” (RMSE of $9.0 \mu\text{g}/\text{m}^3$ and $11.8 \mu\text{g}/\text{m}^3$, and correlations of 0.54 and 0.50, respectively). However, “final setup”
80 still underestimates PM₁₀ and PM_{2.5} concentrations, especially during summer and in February. This underestimation corresponds to the current state of knowledge on PM modeling. The mean bias in PM_{2.5} summertime concentrations is slightly reduced by the use of MEGAN 3.2 EFs ($-1.7 \mu\text{g}/\text{m}^3$ compared to $-2.2 \mu\text{g}/\text{m}^3$ with MEGAN 2.1 EFs and $-2.4 \mu\text{g}/\text{m}^3$ ORCHIDEE EFs) due to higher monoterpene and sesquiterpene emissions (which are more efficient SOA precursor than isoprene for which emissions are significantly lower with MEGAN 3.2). Summertime RMSE ($5.1 \mu\text{g}/\text{m}^3$ with MEGAN 3.2 compared
85 to $5.5 \mu\text{g}/\text{m}^3$ with MEGAN 2.1 EF and $5.3 \mu\text{g}/\text{m}^3$ ORCHIDEE EF) and correlation (0.52 with MEGAN 3.2 compared to 0.48 $\mu\text{g}/\text{m}^3$ with MEGAN 2.1 EF and $0.45 \mu\text{g}/\text{m}^3$ ORCHIDEE EF) are also improved using MEGAN 3.2. A better performance on summertime PM₁₀ concentrations is obtained with “final setup” (bias, RMSE and correlation are respectively equal to $-5.7 \mu\text{g}/\text{m}^3$, $8.5 \mu\text{g}/\text{m}^3$ and 0.54 with “final setup” and to $-7.8 \mu\text{g}/\text{m}^3$, $10.4 \mu\text{g}/\text{m}^3$ and 0.41 with “CHIMERE 2020”), largely due to the implementation of fungal spores. Moreover, the modeling of higher PM_{2.5} peaks with “final setup” (compared to
90 “CHIMERE 2020”) which are closer to the observations during the months of February and March 2018 is also explained by the integration of HDDs which represents the higher residential heating during the cold spells in early 2018. Conversely, PM_{2.5} peaks in November and December 2018 are reduced due to relatively mild temperatures above seasonal normals. In general, using dynamic parameterizations for emissions also leads to a slight improvement of scores for both PM_{2.5} and PM₁₀ with a decrease in RMSE and an increase in correlation.

95 Higher NO_2 concentrations are simulated with “final setup” (mean bias equal to $-1.8 \mu\text{g}/\text{m}^3$ against $-2.8 \mu\text{g}/\text{m}^3$ with “CHIMERE 2020”) due to the calculation of soil NO_x emissions and the effect of temperature on traffic emissions. RMSE is slightly reduced from 5.11 to $4.9 \mu\text{g}/\text{m}^3$. On the contrary, the correlation slightly decreases from 0.61 to 0.60.

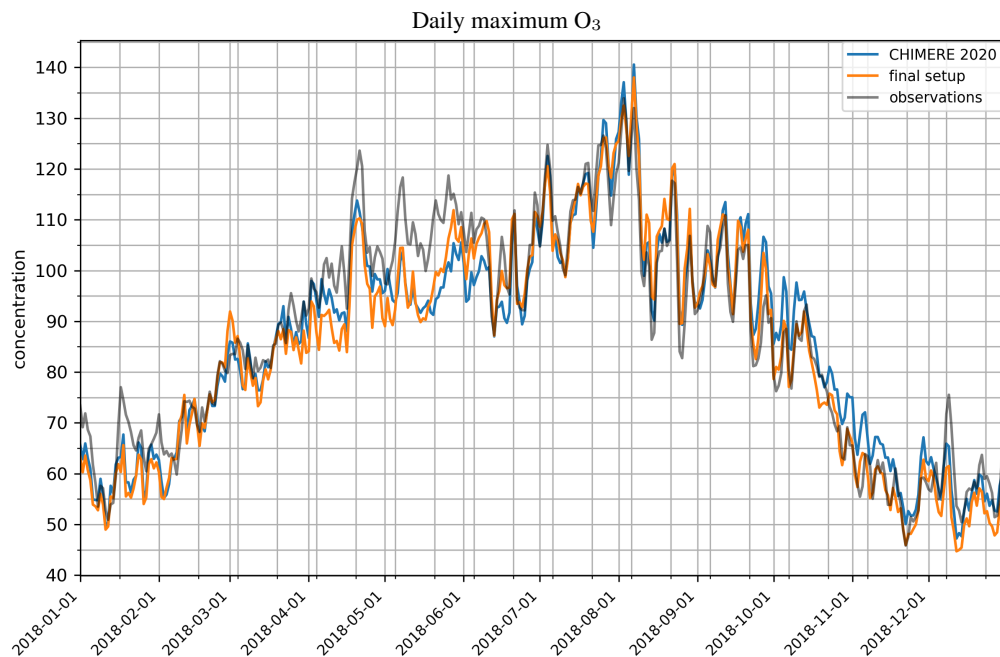


Figure S7. Temporal evolution of daily maximum O₃ concentrations (in $\mu\text{g}/\text{m}^3$) averaged over all stations of the domain, the original CHIMERE version with nondynamic emissions (referred to as "CHIMERE 2020") and the modified version with dynamic emissions computed as a function of meteorology (referred to as "final setup").

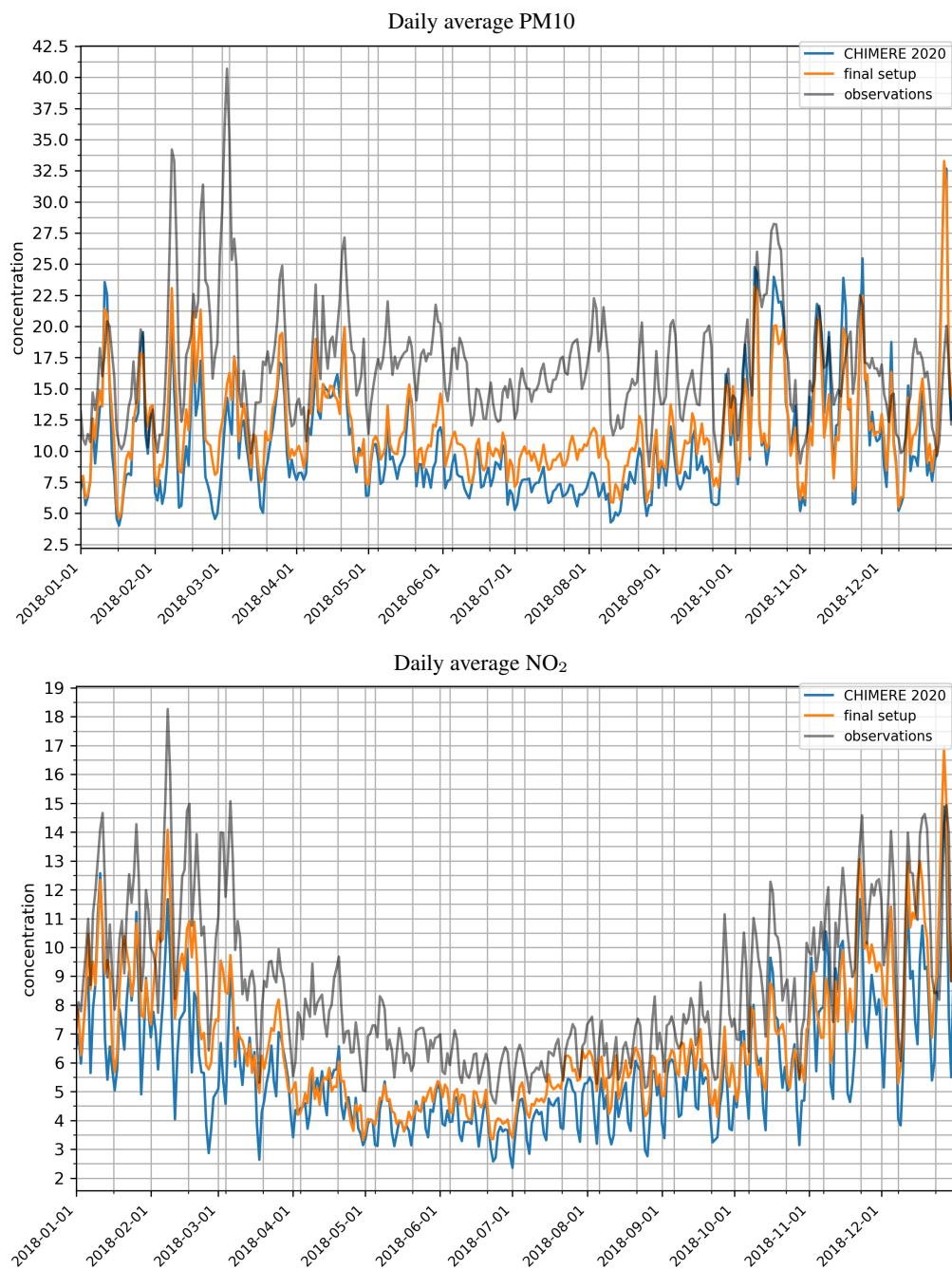


Figure S8. Temporal evolution of daily averaged PM10 and NO₂ concentrations (in $\mu\text{g}/\text{m}^3$) averaged over all stations of the domain, the original CHIMERE version with nondynamic emissions (referred to as "CHIMERE 2020") and the modified version with dynamic emissions computed as a function of meteorology (referred to as "final setup").

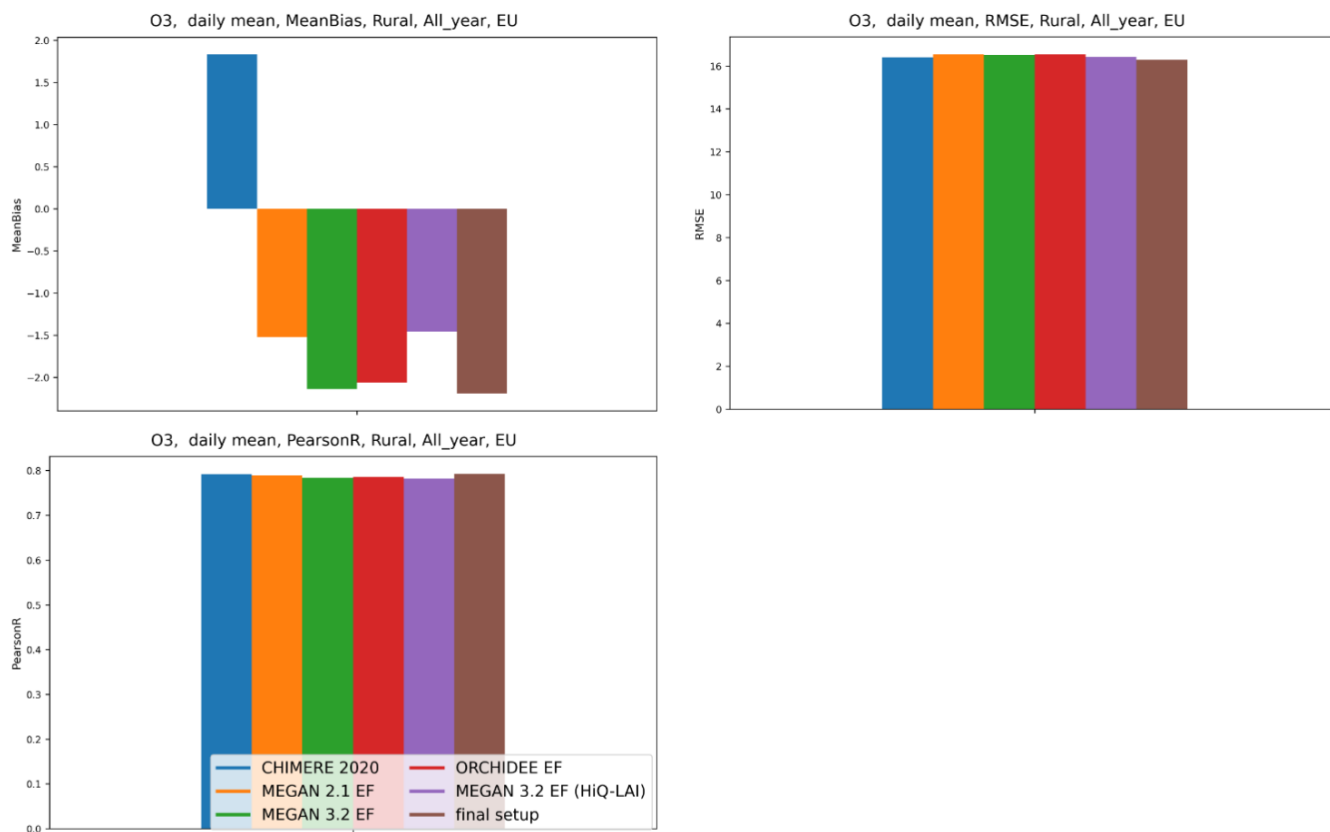


Figure S9. Scores of the various CHIMERE simulations for daily averaged O₃ concentrations. Mean biases and RMSE are in μg/m³.

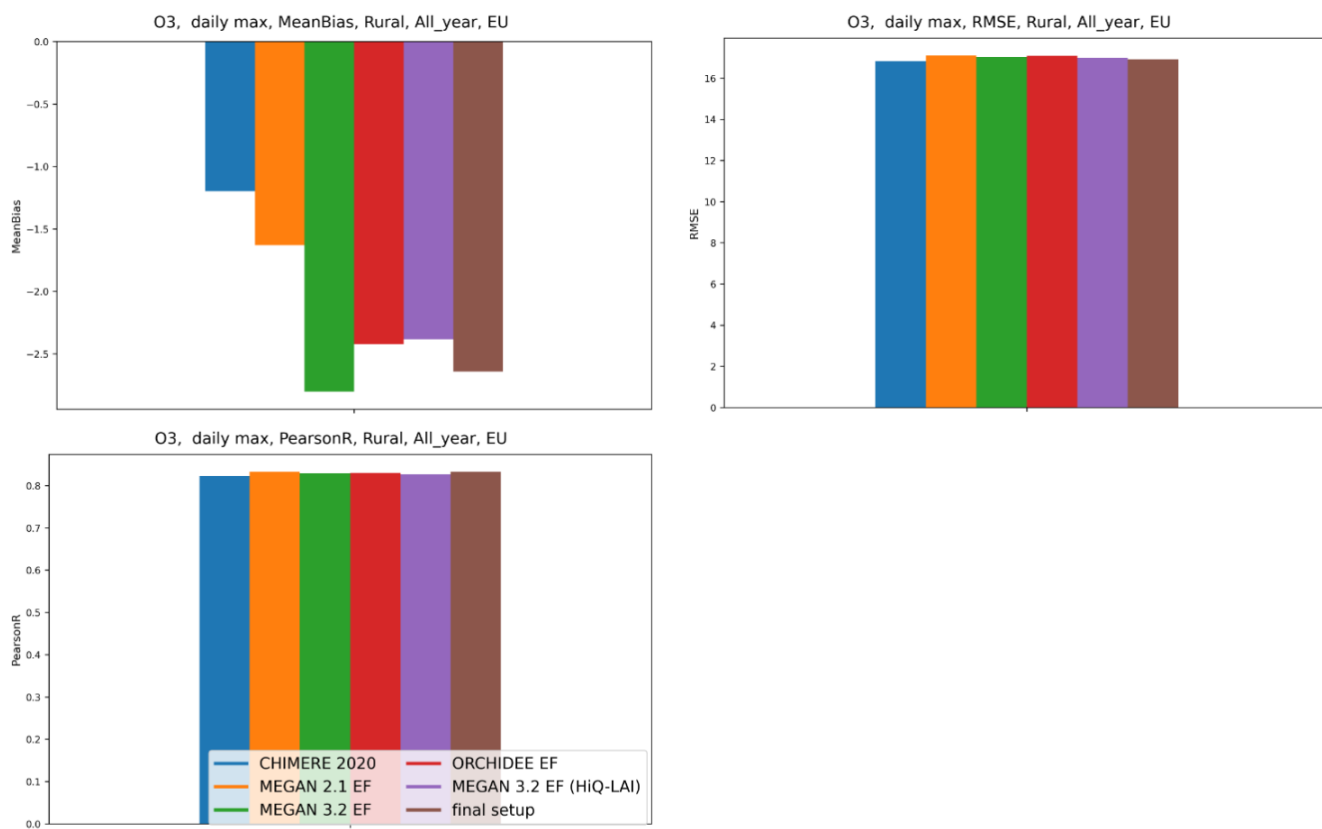


Figure S10. Scores of the various CHIMERE simulations for daily maximum O₃ concentrations. Mean biases and RMSE are in $\mu\text{g}/\text{m}^3$.

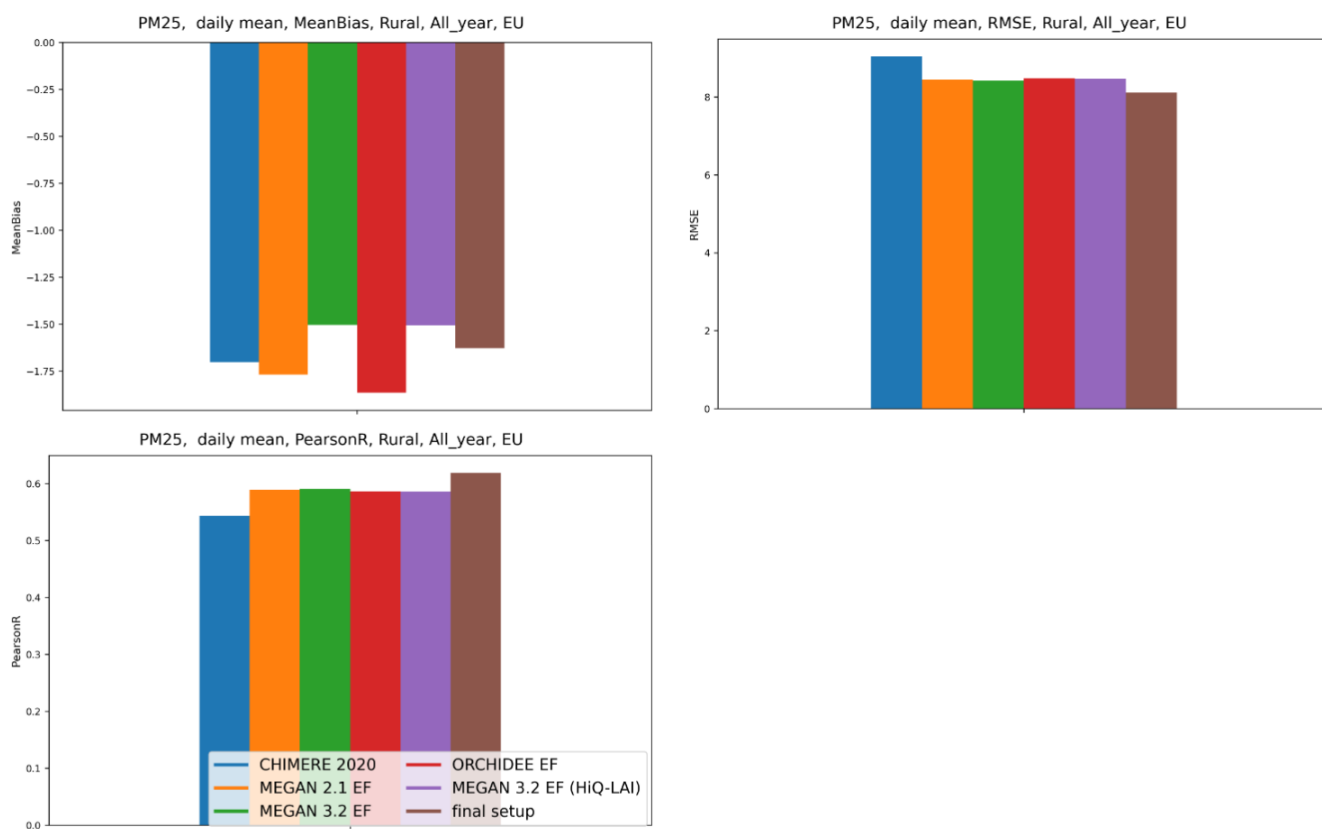


Figure S11. Scores of the various CHIMERE simulations for daily averaged PM2.5 concentrations. Mean biases and RMSE are in $\mu\text{g}/\text{m}^3$.

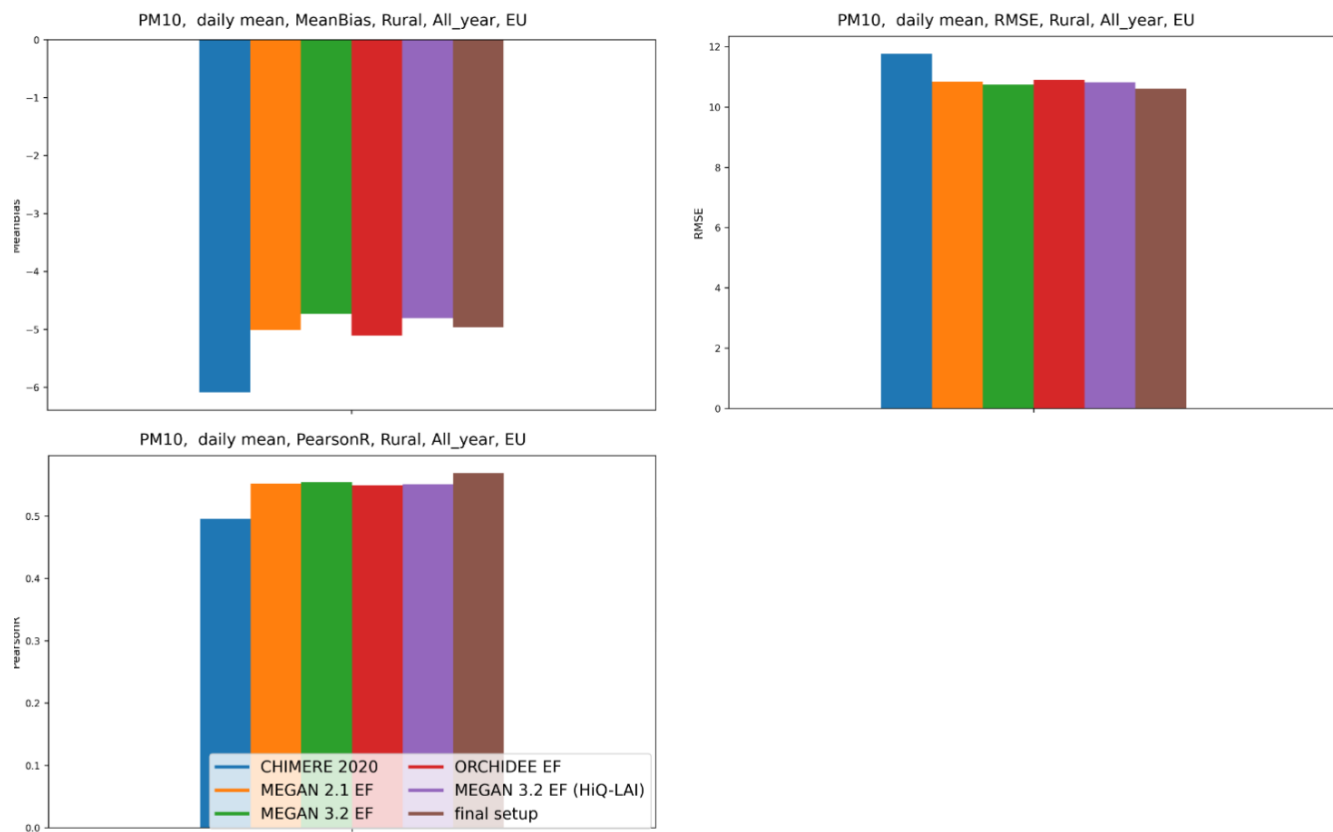


Figure S12. Scores of the various CHIMERE simulations for daily averaged PM10 concentrations. Mean biases and RMSE are in $\mu\text{g}/\text{m}^3$.

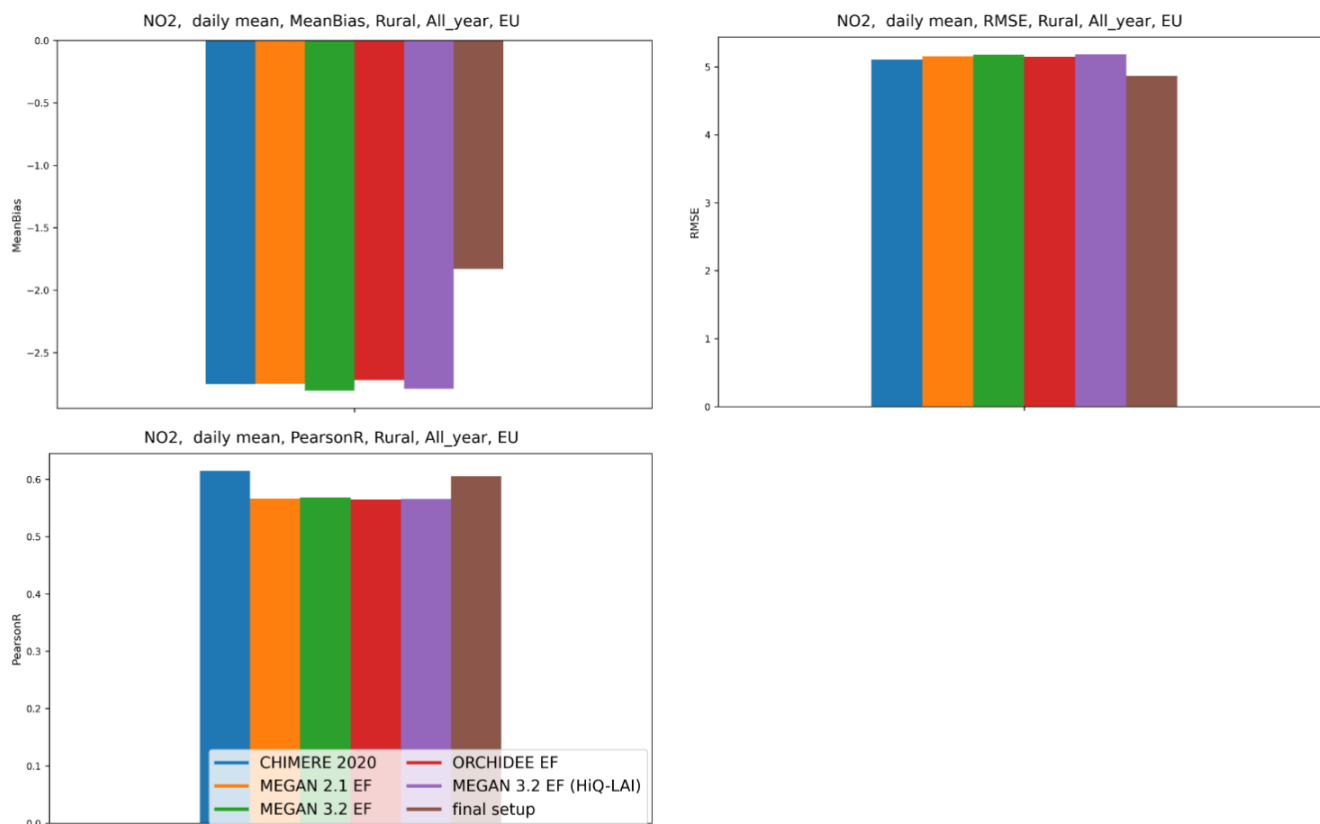


Figure S13. Scores of the various CHIMERE simulations for daily averaged NO₂ concentrations. Mean biases and RMSE are in μg/m³.

S3 Evolution of meteorological parameters during the 1990-2022 period

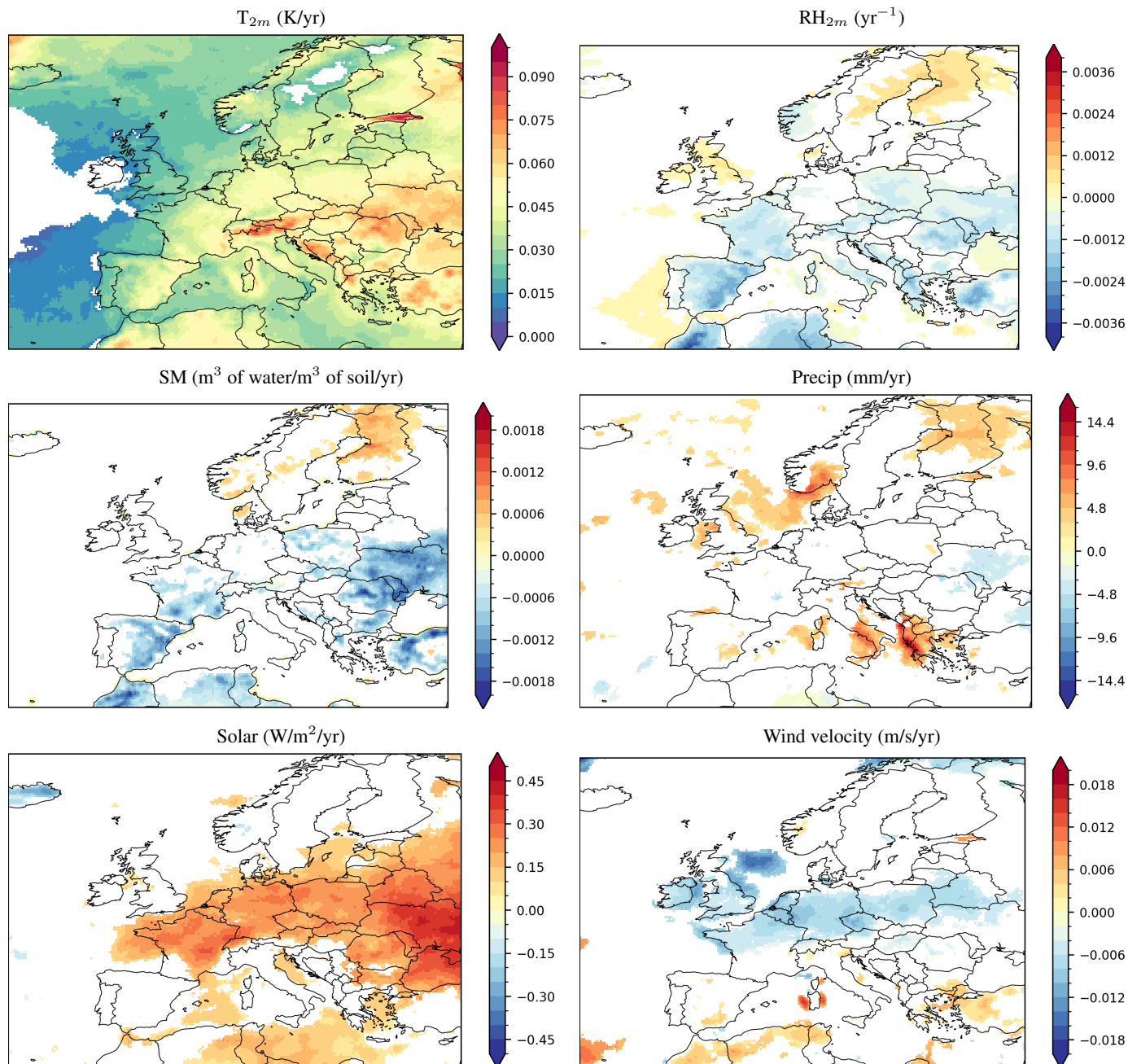


Figure S14. Maps of interannual trends over Europe for several meteorological drivers during the 1990-2022 period. Only significant trends are shown.

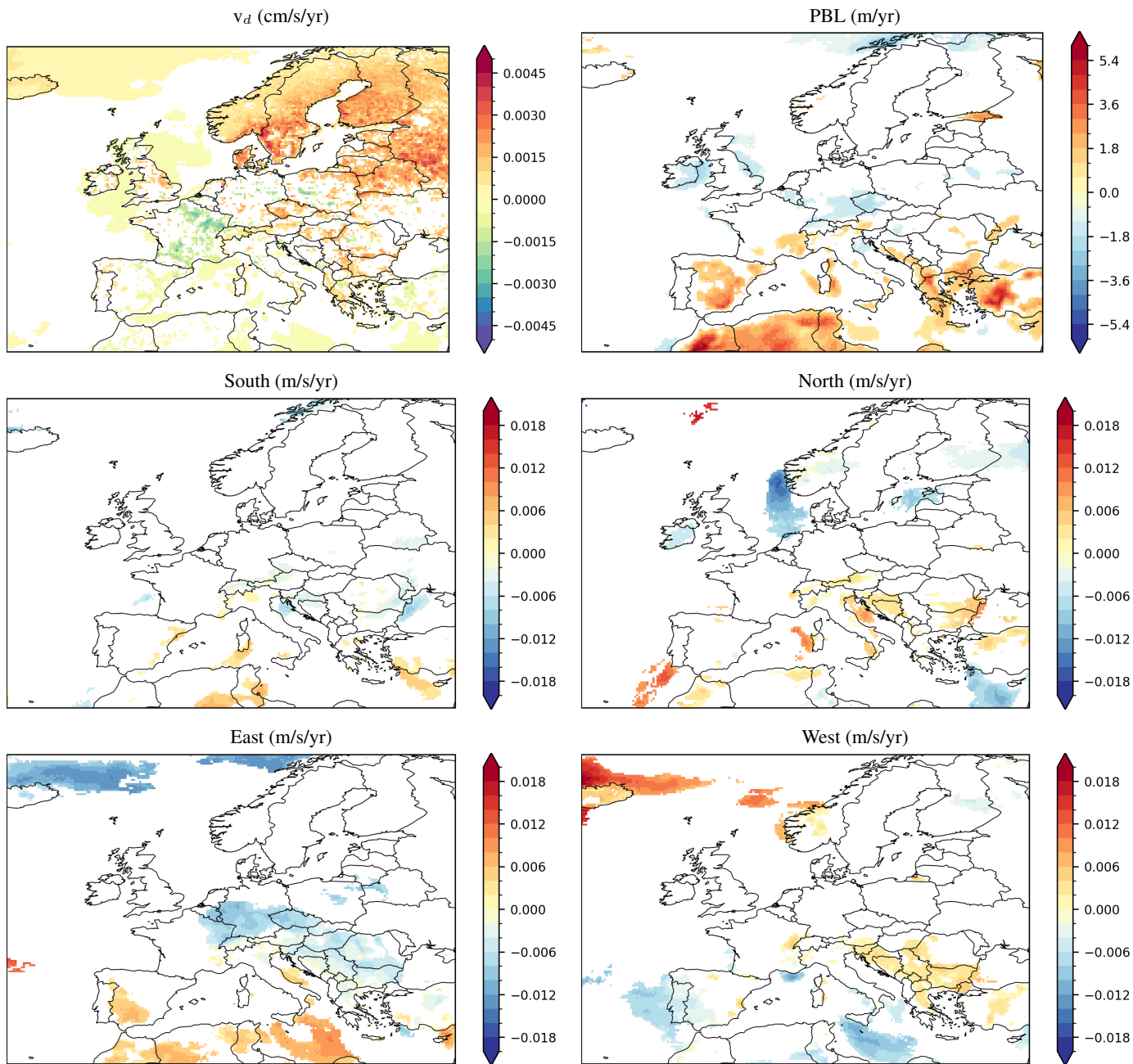


Figure S15. Maps of interannual trends over Europe for several meteorological drivers during the 1990-2022 period. Only significant trends are shown.

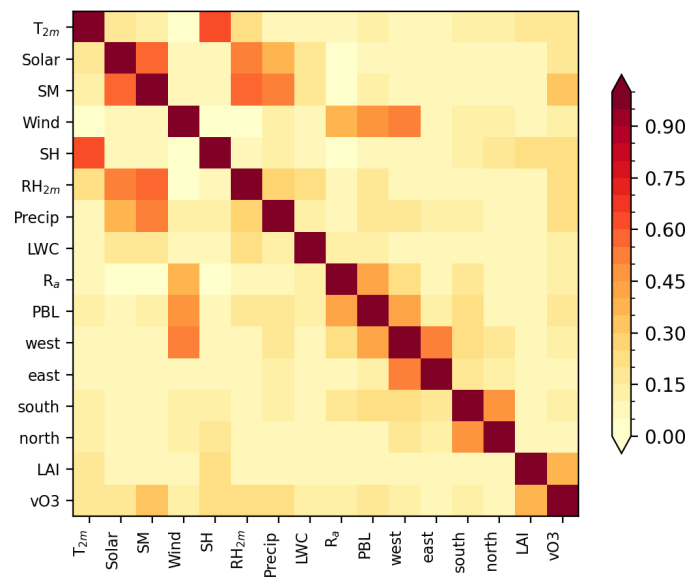


Figure S16. Autocorrelation matrix between the annual values of the different meteorological drivers considered in this study.

S4 Evolution of emissions during the 1990-2022 period

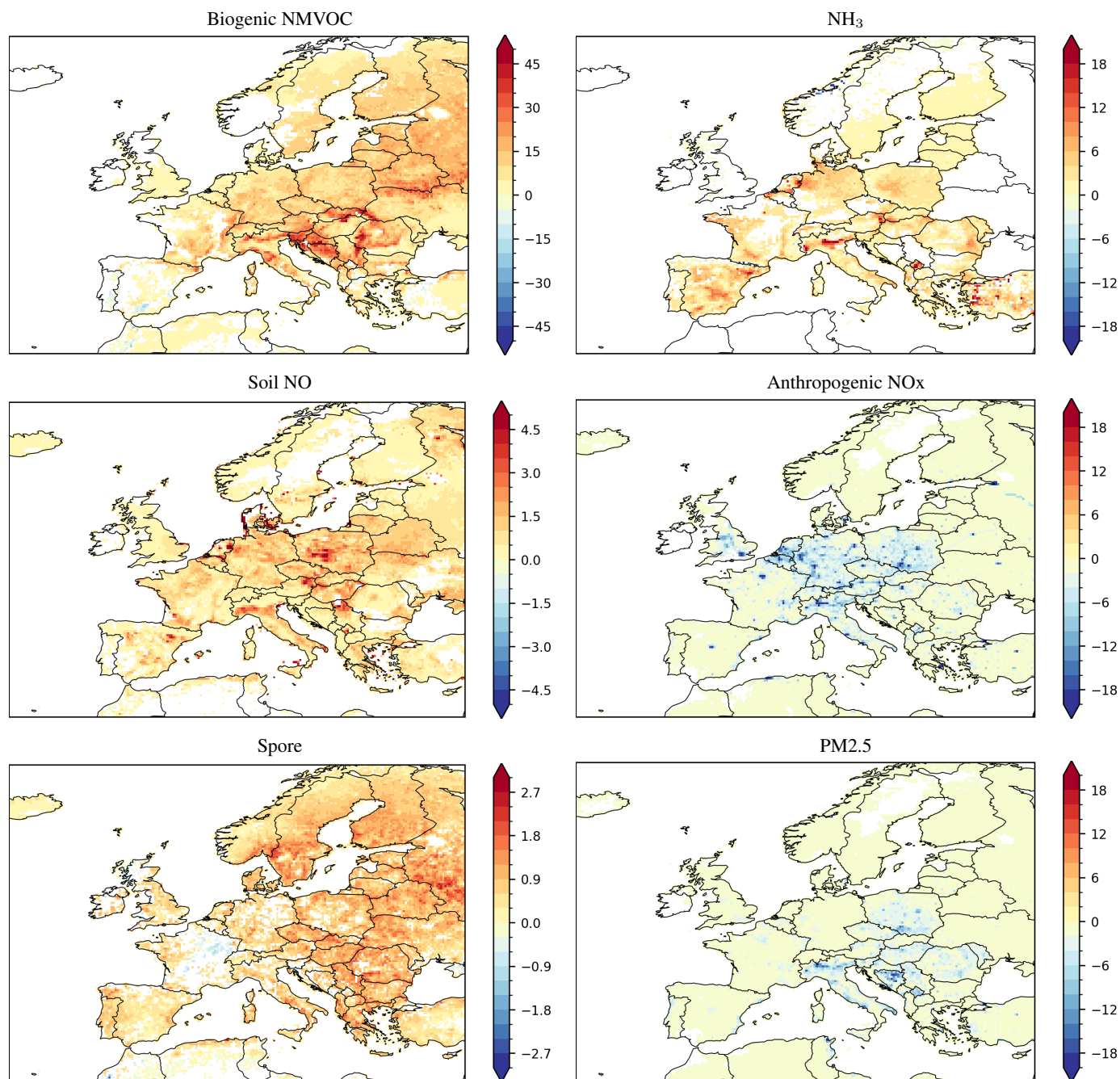


Figure S17. Maps of emissions interannual trends (in $\text{mg}/\text{m}^2/\text{yr}$) over Europe during the 1990-2022 period. Only significant trends are shown.

	temperature sensitivities		Weather-driven trends	
	Value	R ²	Value	R ²
Population-weighted Ozone metrics				
daily O ₃	1.14 µg/m ³ (2.11%) per °C	0.37	0.0361 µg/m ³ (0.07%) per year	0.1
max O ₃	1.82 µg/m ³ (2.41%) per °C	0.43	0.0639 µg/m ³ (0.08%) per year	0.14
4MDA8	3.86 µg/m ³ (2.71%) per °C	0.14	Nonsignificant	
SOMO35	477.71 µg/m ³ .h (12.08%) per °C	0.38	19.93 µg/m ³ .h (0.5%) per year	0.18
AOT40c	1741 µg/m ³ .h (15.35%) per °C	0.26	78.32 µg/m ³ .h (0.69%) per year	0.15
Population-weighted annual NO ₂ concentrations				
NO ₂	-0.37 (-4.2%) per °C	0.54	-0.0093 (-0.11%) per year	0.1
Population-weighted annual PM concentrations				
PM10	-0.45 µg/m ³ (-3.46%) per °C	0.14	-0.0296 µg/m ³ (-0.23%) per year	0.17
PM2.5	-0.53 µg/m ³ (-4.73%) per °C	0.21	-0.0325 µg/m ³ (-0.29%) per year	0.22
SIA	-0.28 µg/m ³ (-4.55%) per °C	0.21	-0.0155 µg/m ³ (-0.25%) per year	0.17
SOA	0.032 µg/m ³ (9.41%) per °C	0.3	0.0016 µg/m ³ (0.48%) per year	0.22
Primary PM	-0.31 µg/m ³ (-6.83%) per °C	0.27	-0.0189 µg/m ³ (-0.41%) per year	0.28
Fungal spore	0.036 µg/m ³ (7.51%) per °C	0.36	0.0019 µg/m ³ (0.4%) per year	0.29
Dust	0.045 µg/m ³ (7.86%) per °C	0.19	Nonsignificant	
Sea salt	0.012 µg/m ³ (3.18%) per °C	0.11	Nonsignificant	
Total deposition over EUR				
Total S _{oxidized}	-26.89 kT (-2.42%) per °C	0.30	Nonsignificant	
Total N _{reduced}	74.21 kT (3.2%) per °C	0.49	3.45 kT (0.15%) per year	0.33
Total N _{oxidized}	-20.42 kT (-0.9%) per °C	0.1	Nonsignificant	
Wet S _{oxidized}	-37.94 kT (-4.54%) per °C	0.36	Nonsignificant	
Wet N _{reduced}	Nonsignificant		Nonsignificant	
Wet N _{oxidized}	-59.97 kT(-4.65%) per °C	0.35	Nonsignificant	
Dry S _{oxidized}	11.06 kT (3.99%) per °C	0.39	Nonsignificant	
Dry N _{reduced}	63.7 kT (6.74%) per °C	0.7	2.51 kT (0.27%) per year	0.34
Dry N _{oxidized}	39.54 kT (4.05%) per °C	0.62	1.21 kT (0.12%) per year	0.18

Table S1. Temperature sensitivities and weather-driven trends of concentrations over EUR due to climate change between 1990 and 2010. Relative temperature sensitivities and weather-driven trends are provided under parenthesis. R²: square of the correlation coefficient. Trends and sensitivities appear in black, orange and red if they are classified as "highly significant" (p-values<0.01), "significant" (0.01≤p-values<0.05) and "slightly significant" (0.05≤p-values<0.1) respectively. Trends and sensitivities with p-values>0.1 are shown as "Non-significant".

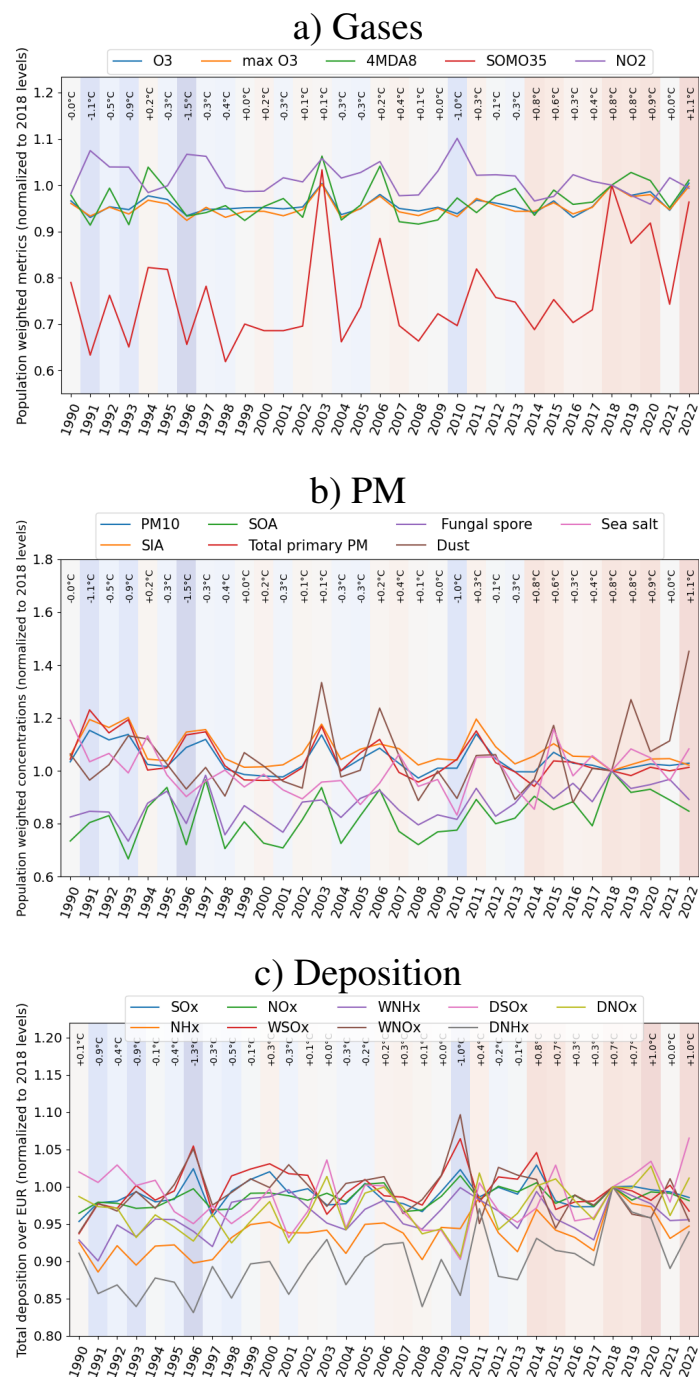


Figure S18. Interannual evolution of the population-weighted metrics for gaseous (a) and PM (b) compounds and total deposition of S_{oxidized} , N_{reduced} and N_{oxidized} over EUR. Results are normalized relative to 2018 levels. The bar colors on the background illustrate the variations of the average annual temperature over EUR compared the average of the 1990-2022 period.

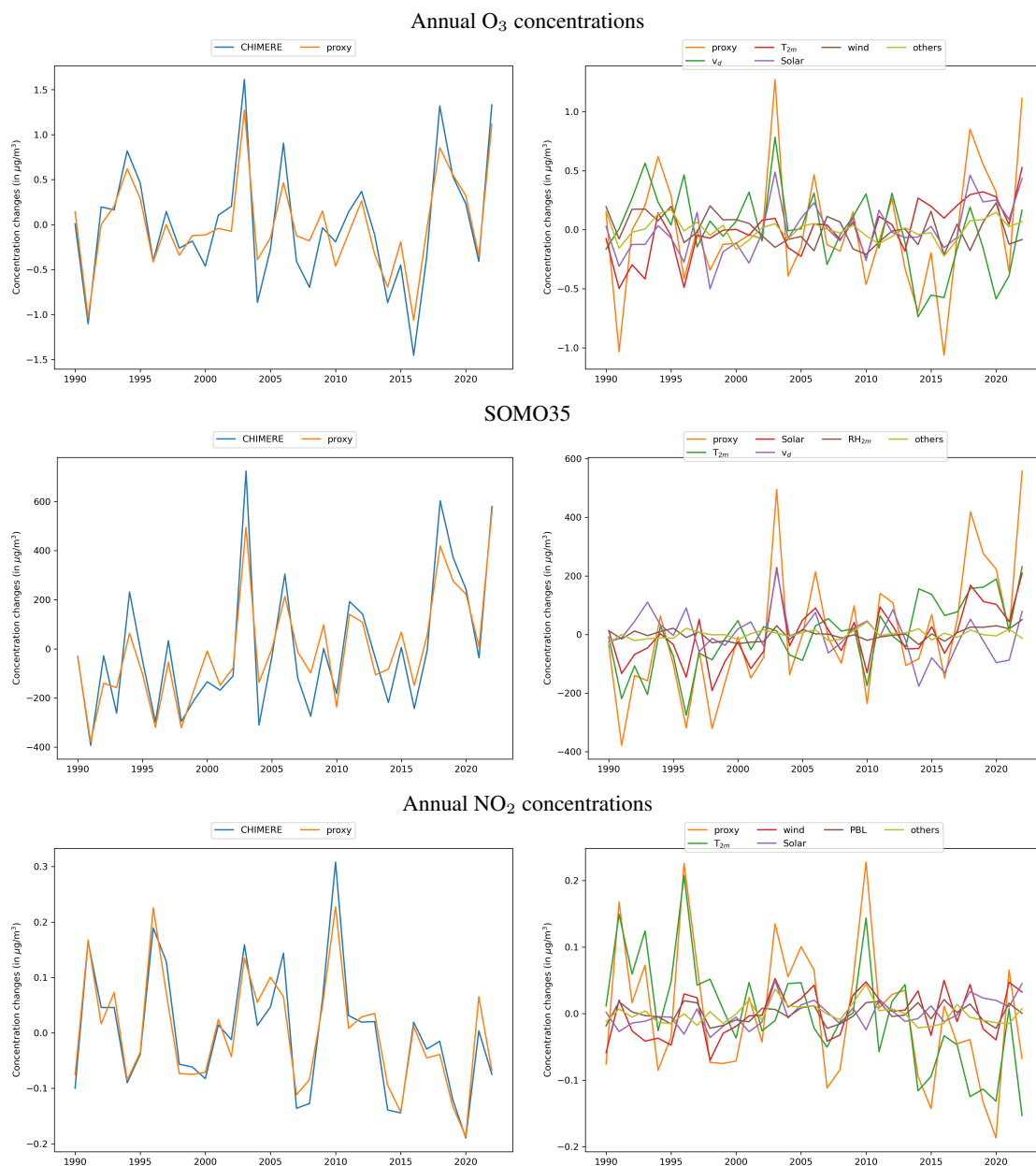


Figure S19. Evolution of several metrics (annual O₃ concentrations, 4DMA8 and annual NO₂ concentrations) simulated by CHIMERE and the meteorological proxy (left) and estimated contribution of the meteorological drivers. Only the contribution of the first 5 dominant drivers are shown, the sum of the contributions of the other drivers correspond to "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

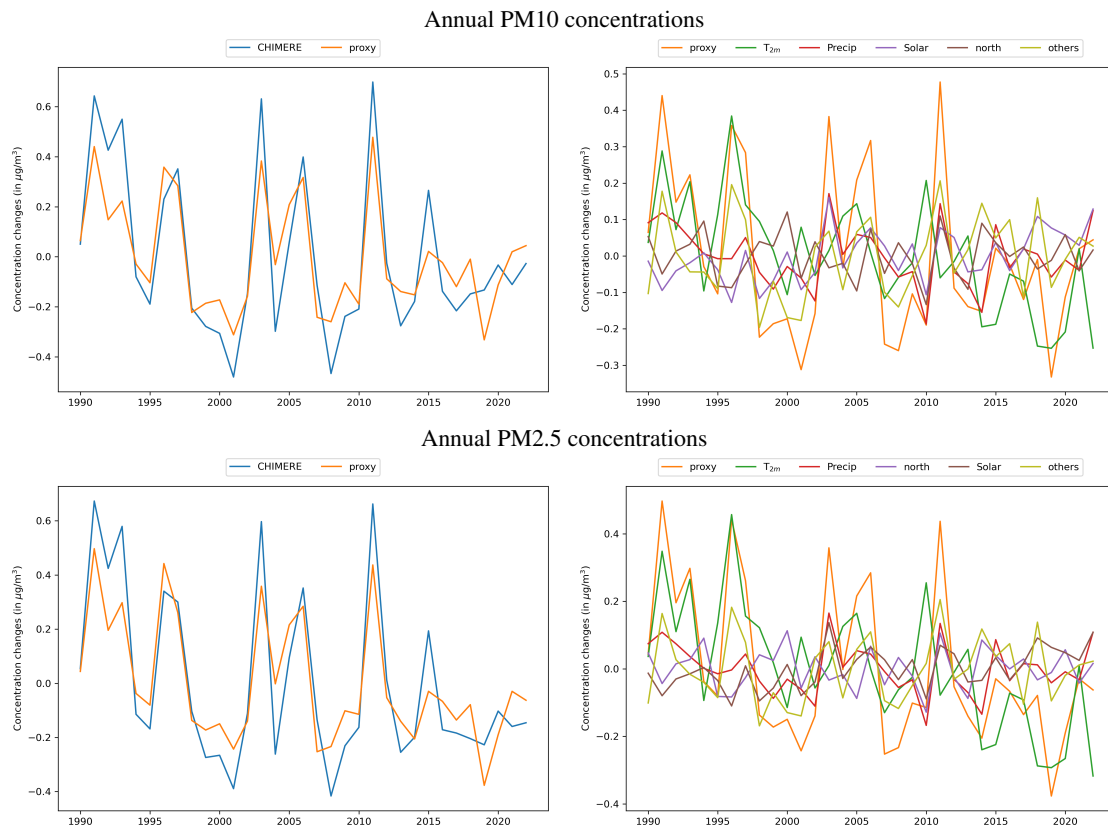


Figure S20. Evolution of annual PM10 and PM2.5 concentrations simulated by CHIMERE and the meteorological proxy (left) and estimated contribution of the meteorological drivers. Only the contribution of the first 5 five dominant drivers are show, the sum of the contributions of the other drivers correspond to "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

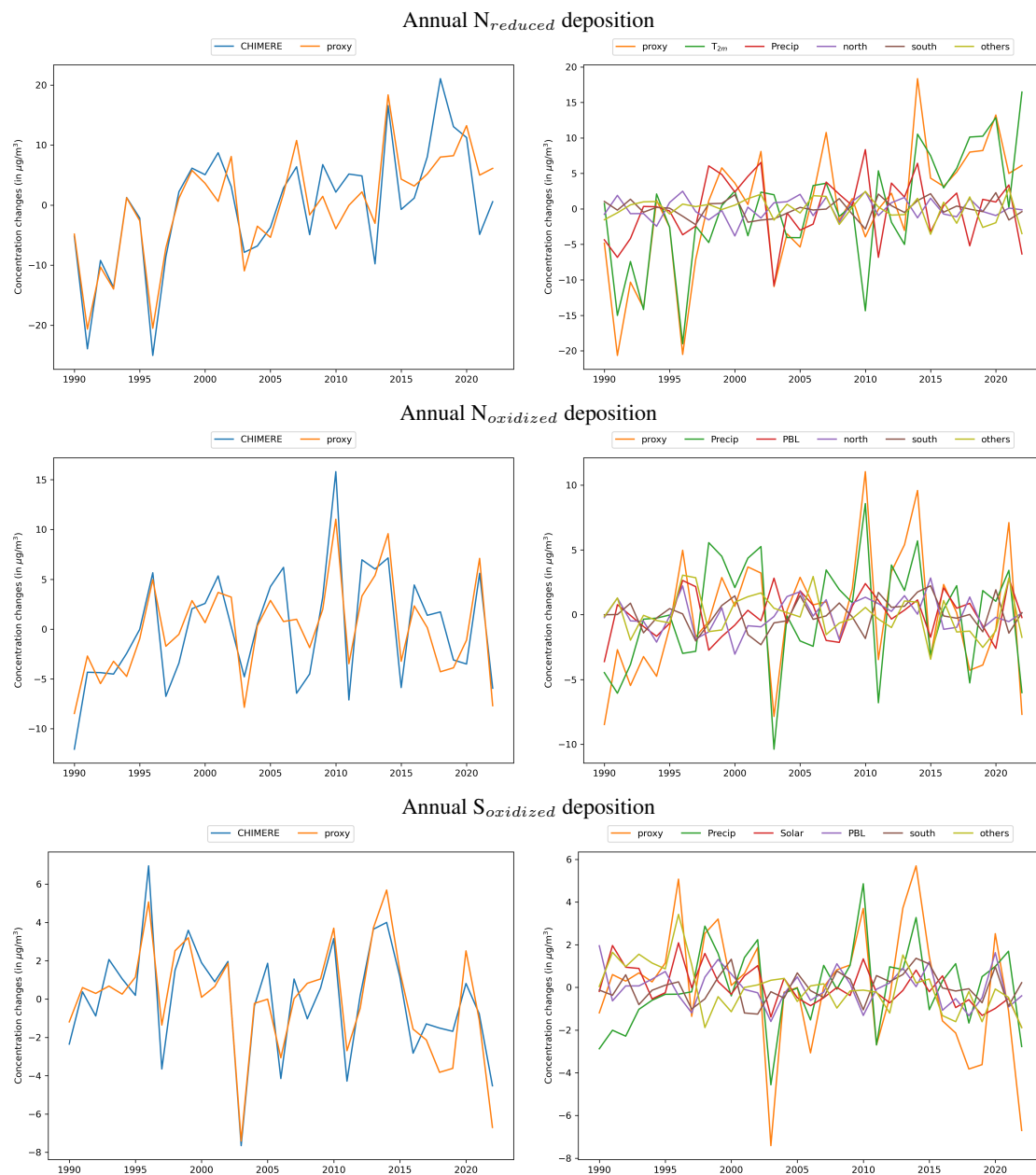


Figure S21. Evolution of annual $N_{reduced}$, $N_{oxidized}$ and $S_{oxidized}$ deposition simulated by CHIMERE and the meteorological proxy (left) and estimated contribution of the meteorological drivers . Only the contribution of the first 5 five dominant drivers are show, the sum of the contributions of the other drivers correspond to "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

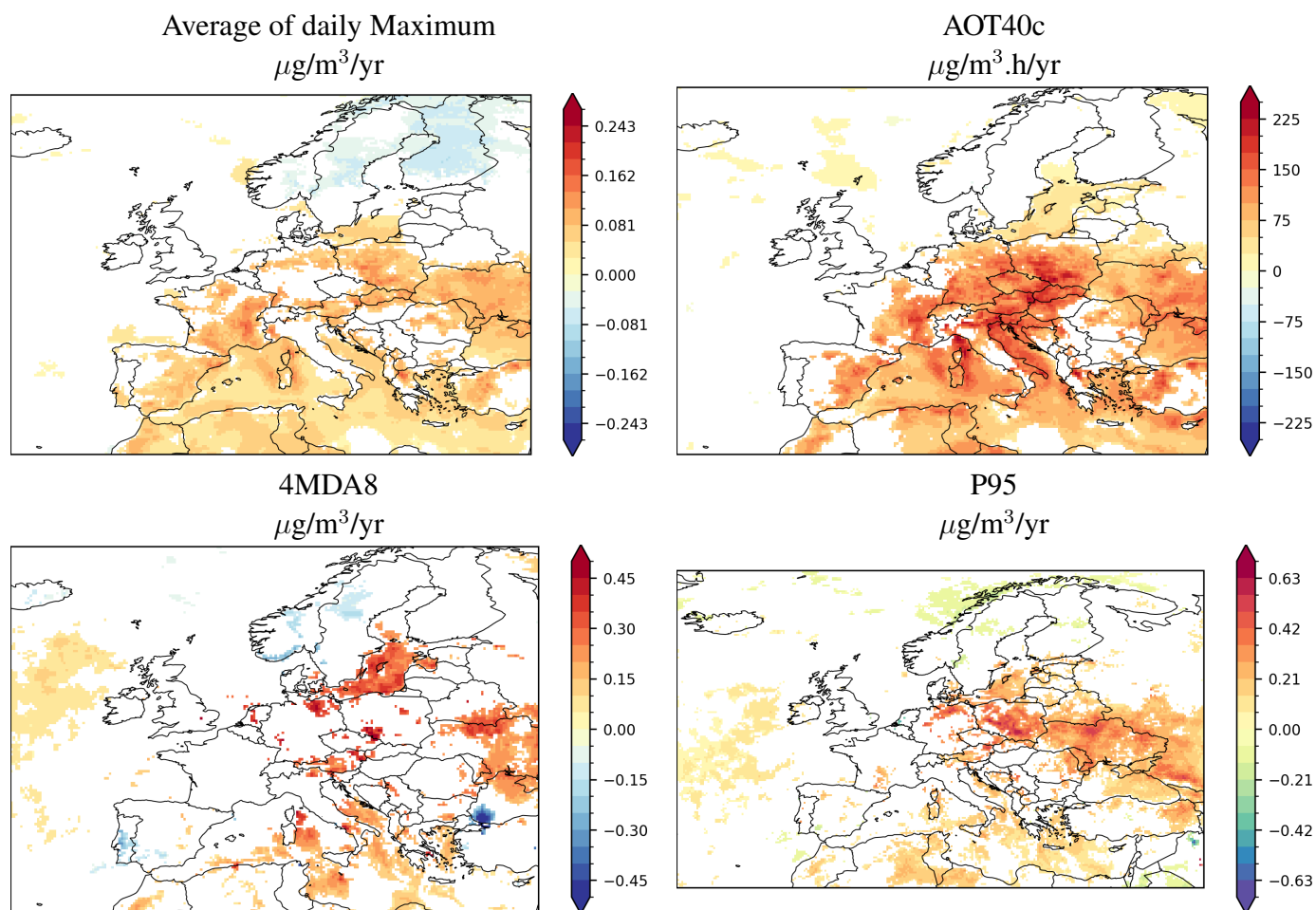


Figure S22. Maps of interannual trends per °C due to climate change over Europe for the annual average of daily maximum ozone concentrations, AOT40c, 4MDA8 and P95 (95th percentile of daily ozone concentrations).

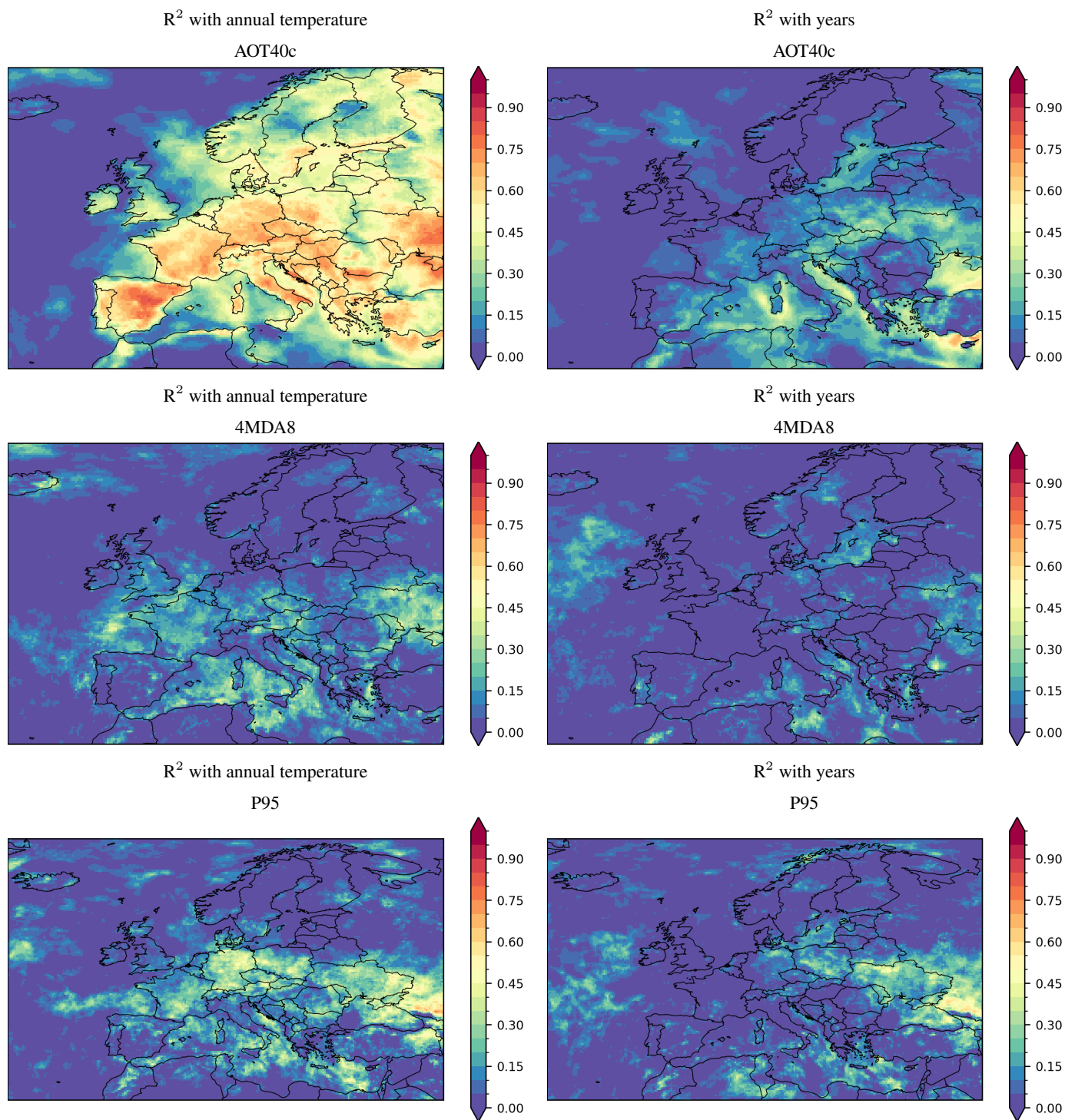


Figure S23. Determination coefficient with annual temperature, years and of the multilinear model for AOT40c, 4MDA8 and P95 (95th percentile of daily ozone concentrations).

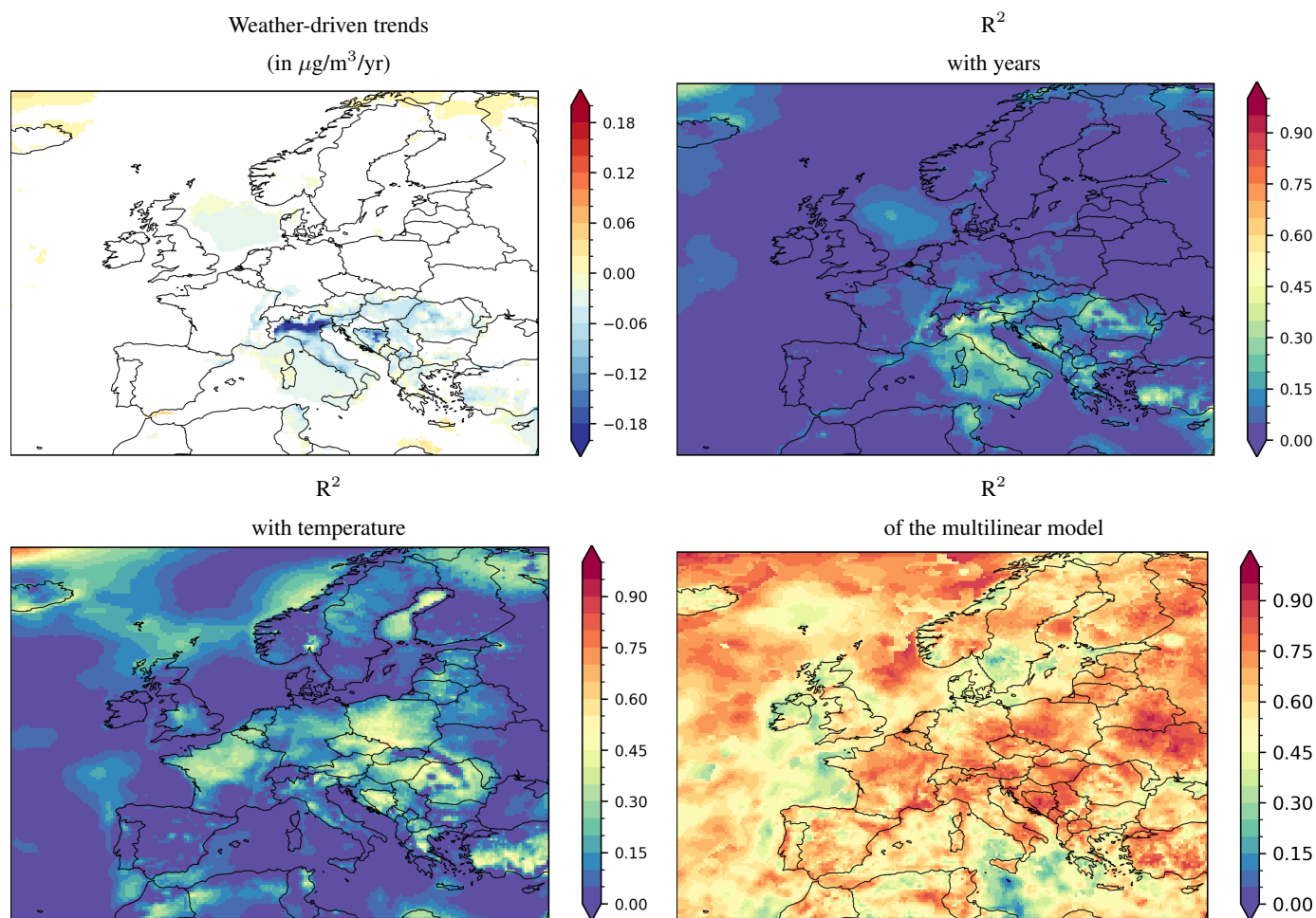


Figure S24. Maps of weather-driven trends of PM_{2.5} annual concentrations and R^2 of concentrations with years, temperature and of the multilinear model.

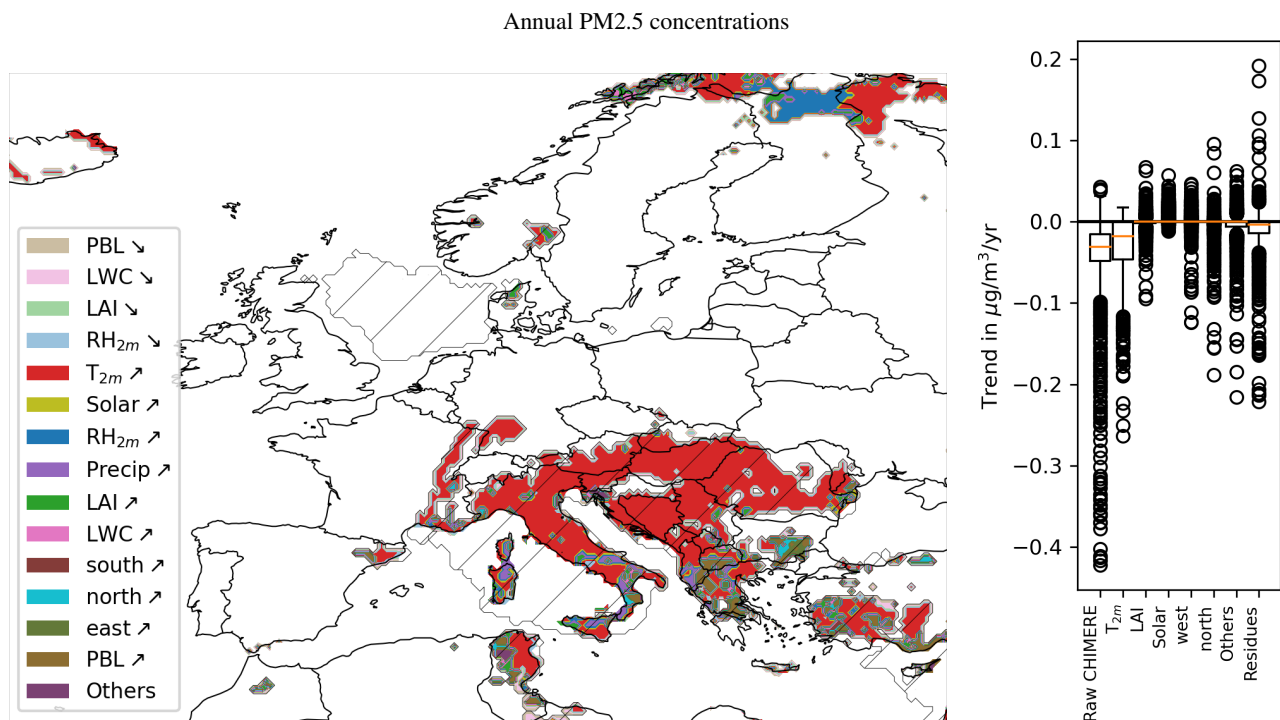


Figure S25. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for PM2.5 annual concentrations. The 14 first dominant drivers (with a positive or negative trend) are shown in the map of the dominant drivers, cells where the evolution of PM2.5 concentrations is dominated by other drivers are gathered in "Others". Similarly, only the first 5 dominant drivers in the boxplot, the sum of the contributions of the other drivers are shown in "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

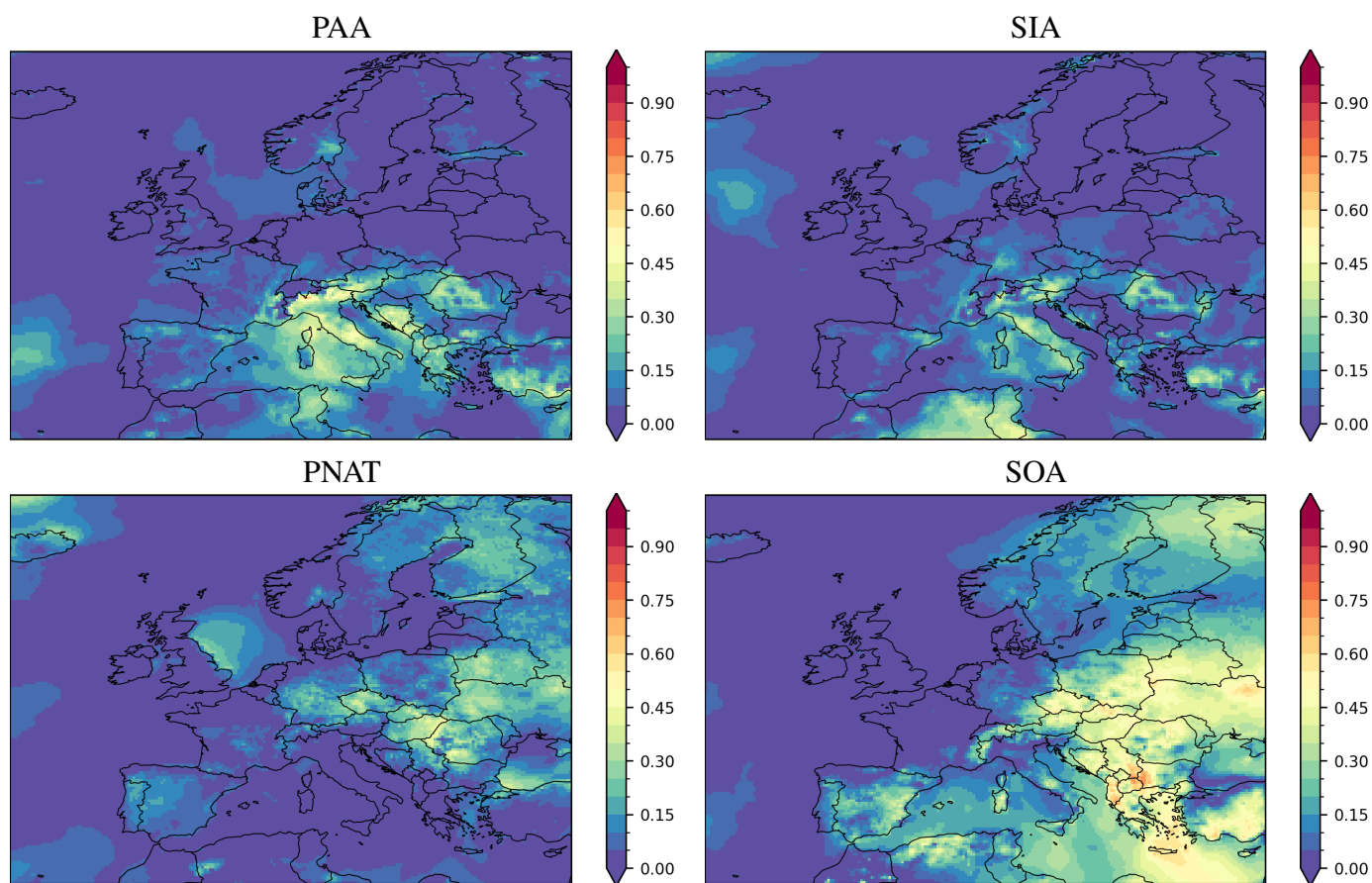


Figure S26. Maps of R^2 with years for PM component concentrations.

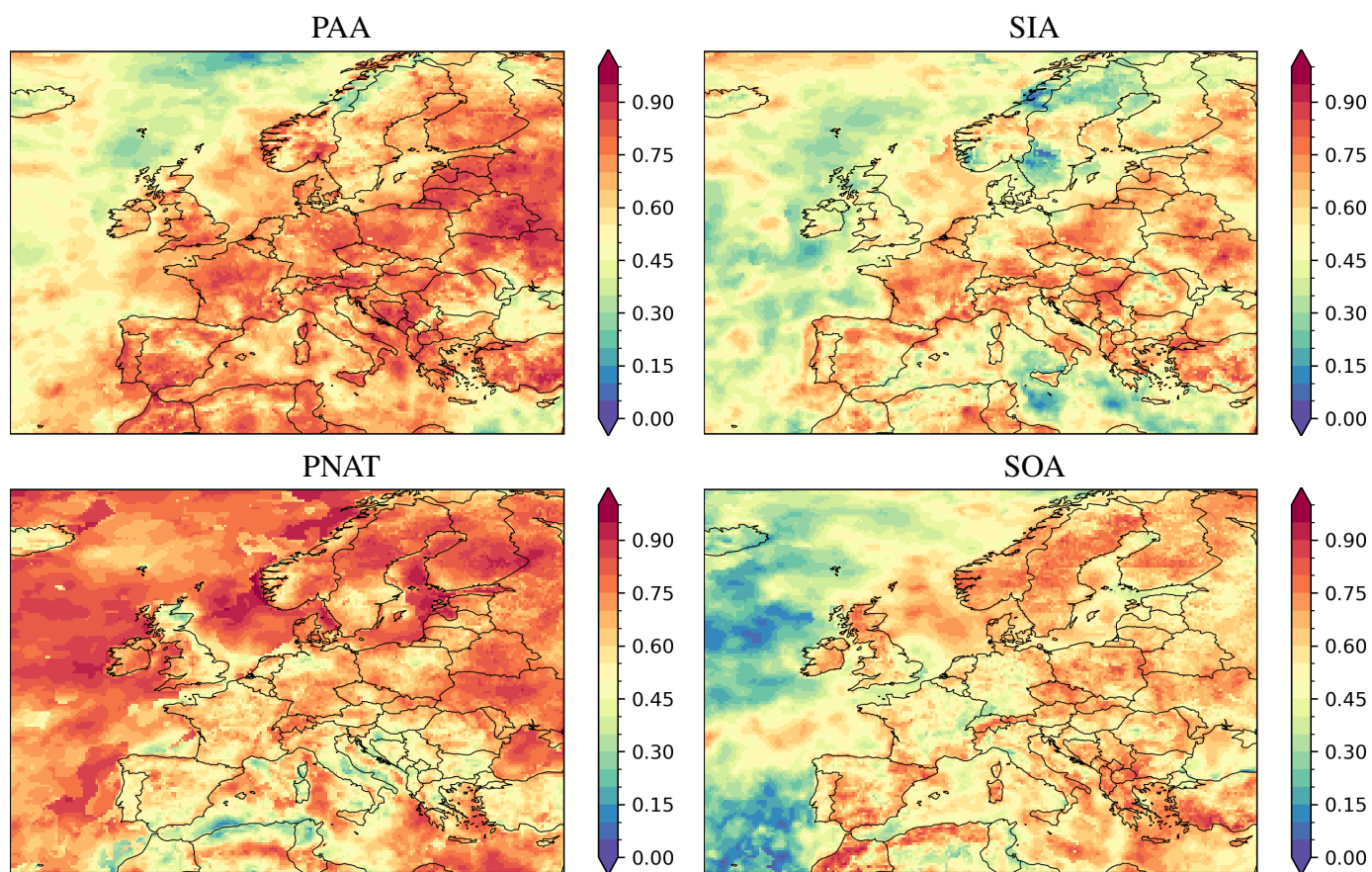


Figure S27. Maps of R^2 of the multilinear model for PM component concentrations.

Annual PAA concentrations

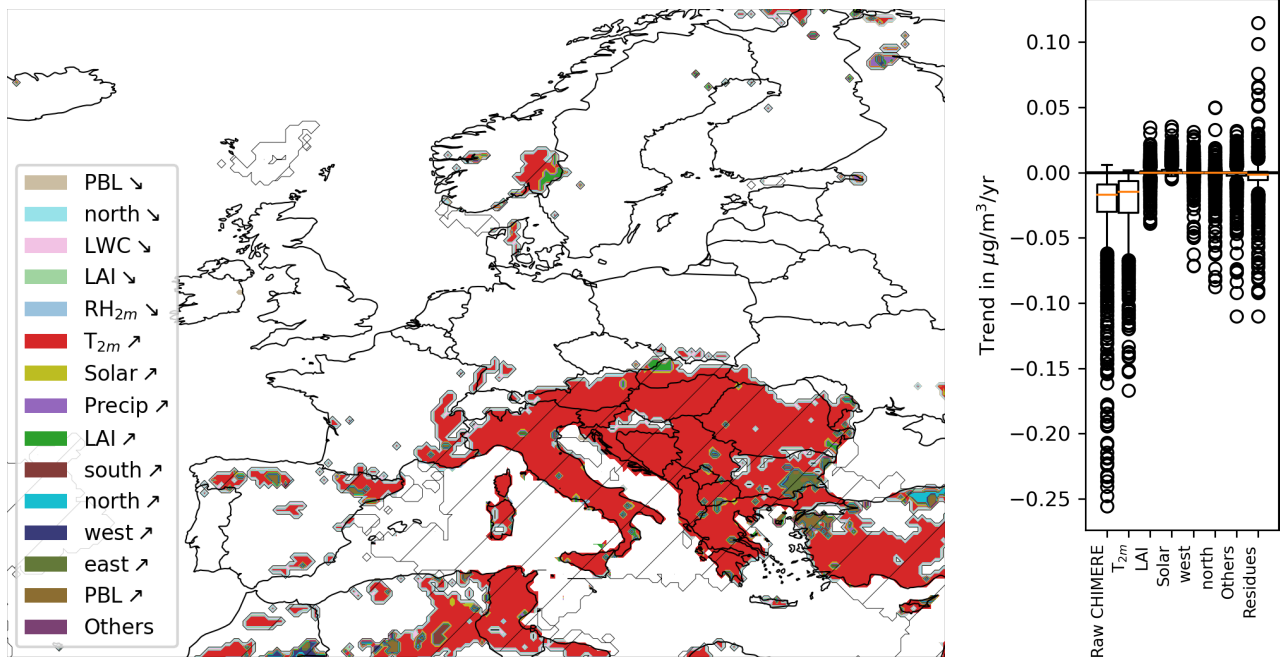


Figure S28. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for PAA annual concentrations. The 14 first dominant drivers (with a positive or negative trend) are shown in the map of the dominant drivers, cells where the evolution of PAA concentrations is dominated by other drivers are gathered in "Others". Similarly, only the first 5 five dominant drivers in the boxplot, the sum of the contributions of the other drivers are shown in "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

Annual SIA concentrations

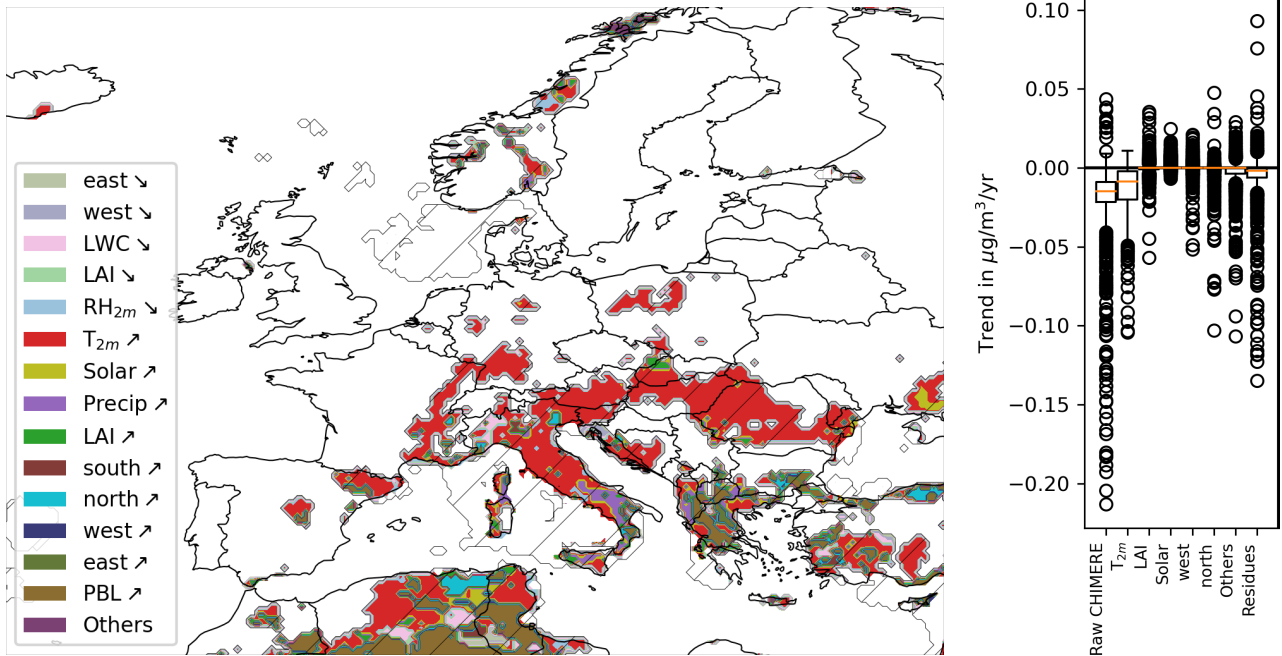


Figure S29. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for SIA annual concentrations. The 14 first dominant drivers (with a positive or negative trend) are shown in the map of the dominant drivers, cells where the evolution of SIA concentrations is dominated by other drivers are gathered in "Others". Similarly, only the first 5 five dominant drivers in the boxplot, the sum of the contributions of the other drivers are shown in "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

Annual SOA concentrations

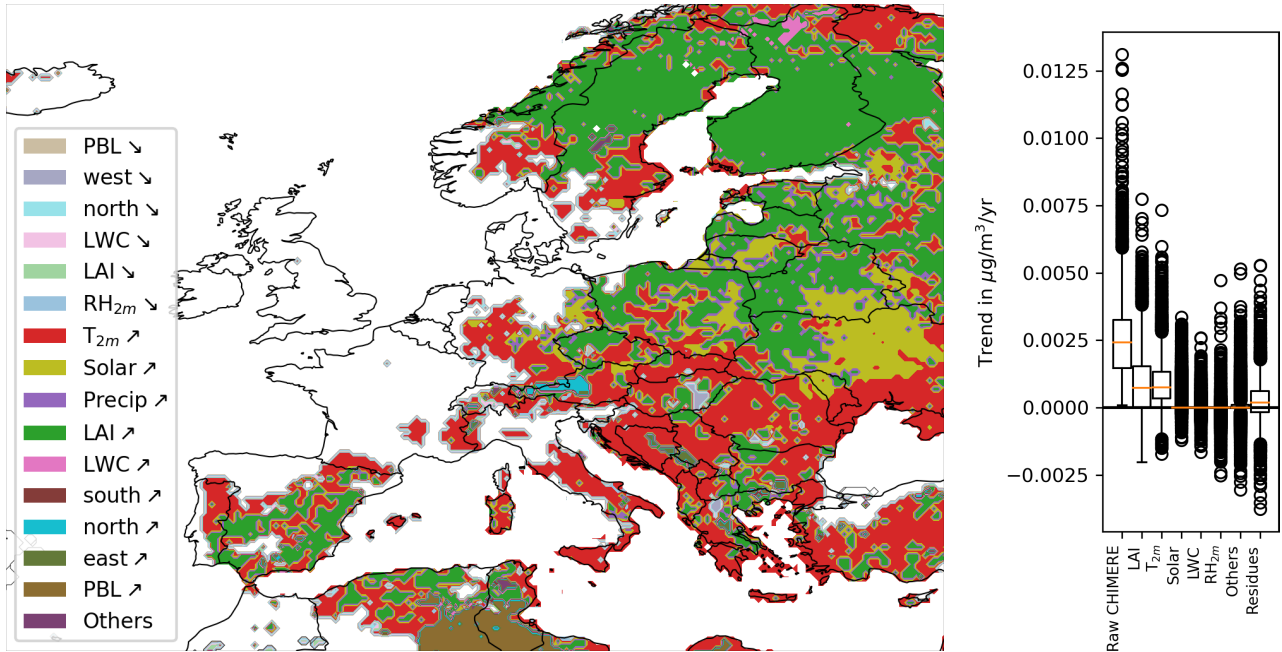


Figure S30. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for SOA annual concentrations. The 14 first dominant drivers (with a positive or negative trend) are shown in the map of the dominant drivers, cells where the evolution of SOA concentrations is dominated by other drivers are gathered in "Others". Similarly, only the first 5 five dominant drivers in the boxplot, the sum of the contributions of the other drivers are shown in "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

Annual PNAT concentrations

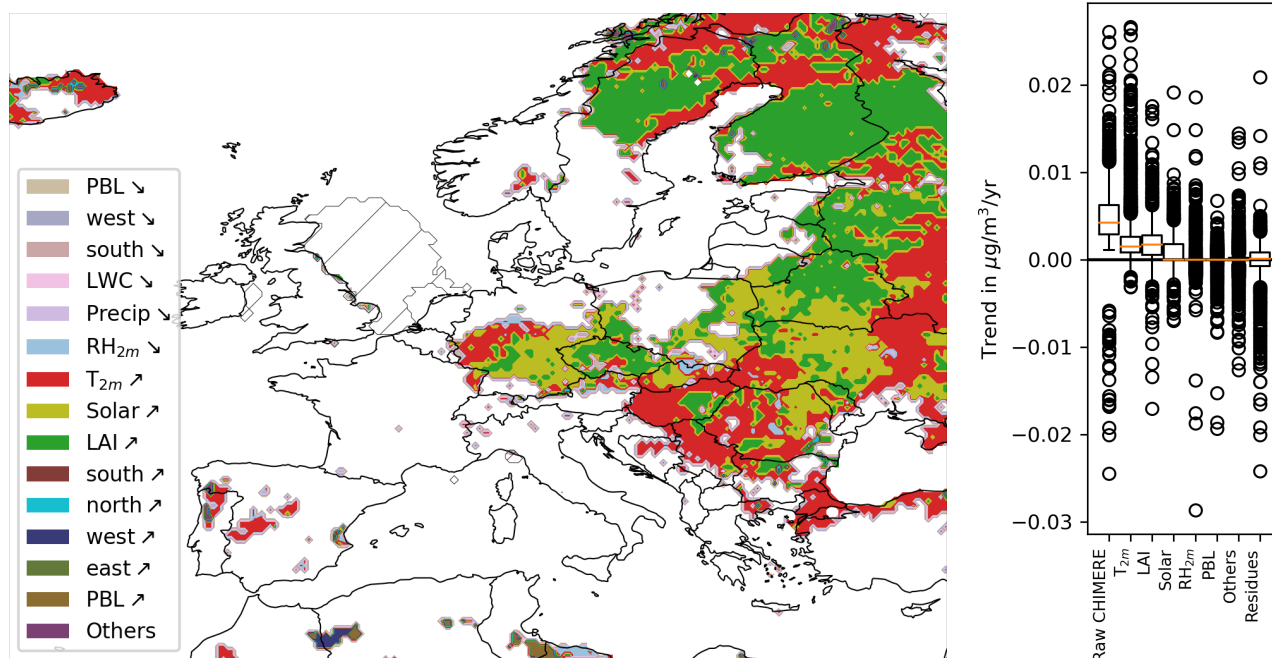


Figure S31. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for PNAT annual concentrations. The 14 first dominant drivers (with a positive or negative trend) are shown in the map of the dominant drivers, cells where the evolution of PNAT concentrations is dominated by other drivers are gathered in "Others". Similarly, only the first 5 dominant drivers in the boxplot, the sum of the contributions of the other drivers are shown in "Others". "Residues" represent the difference between the trend calculated with CHIMERE results and the sum of contributions of all meteorological drivers.

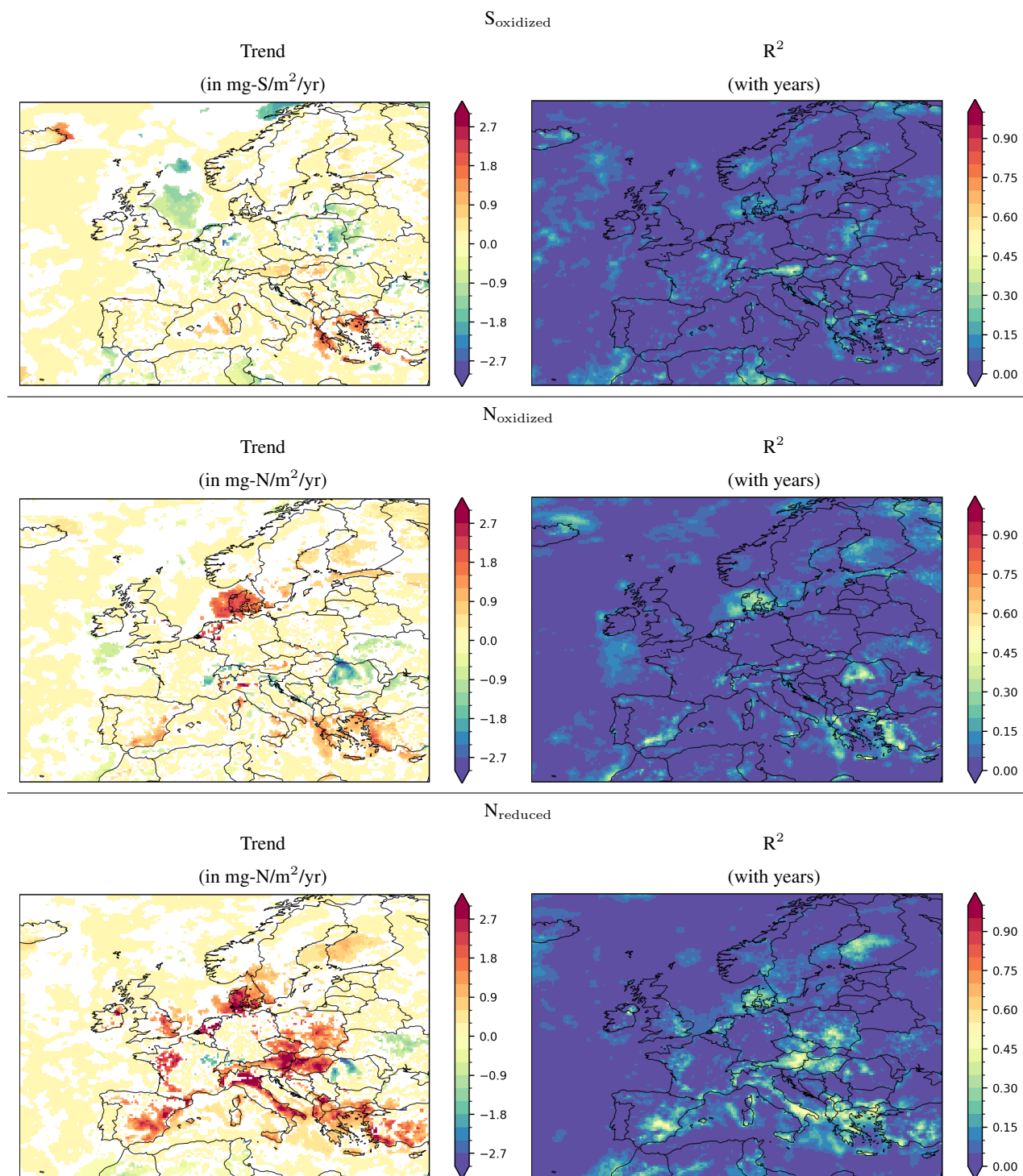


Figure S32. Maps of weather-driven trends and R^2 with years of total (wet+dry) deposition of SO_x , NO_x and NH_x .

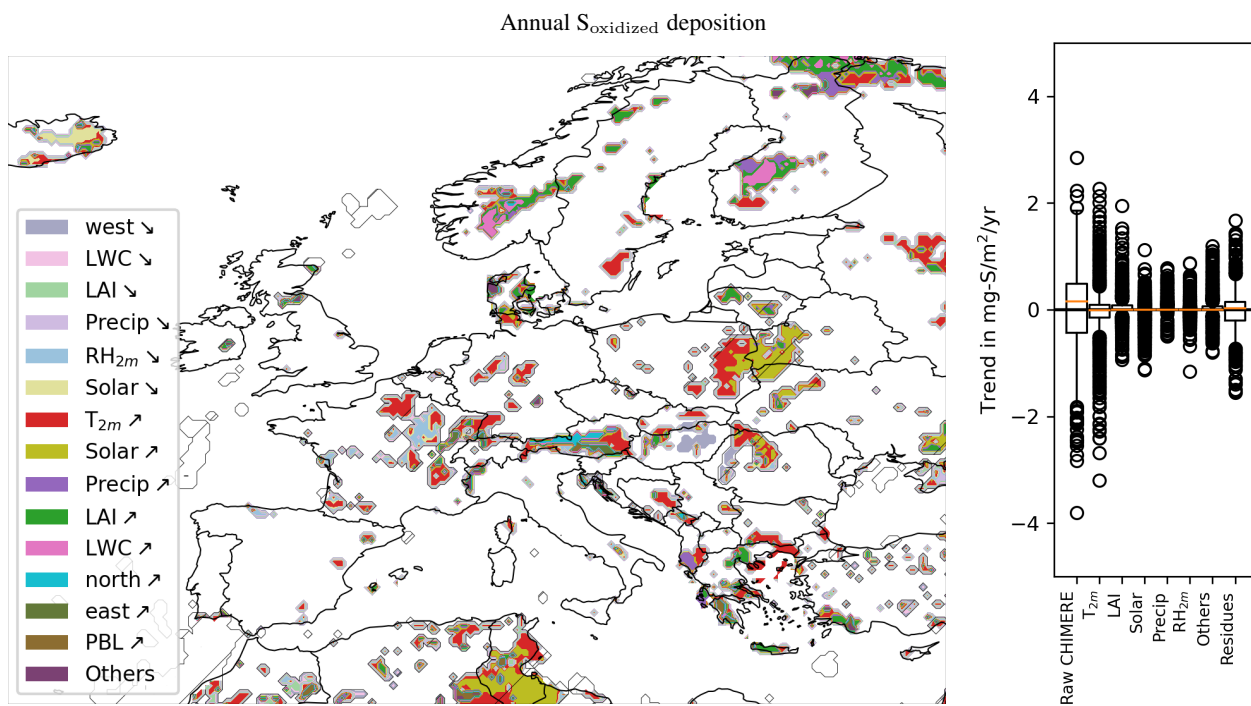


Figure S33. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for annual total (dry+wet) deposition of SO_x .

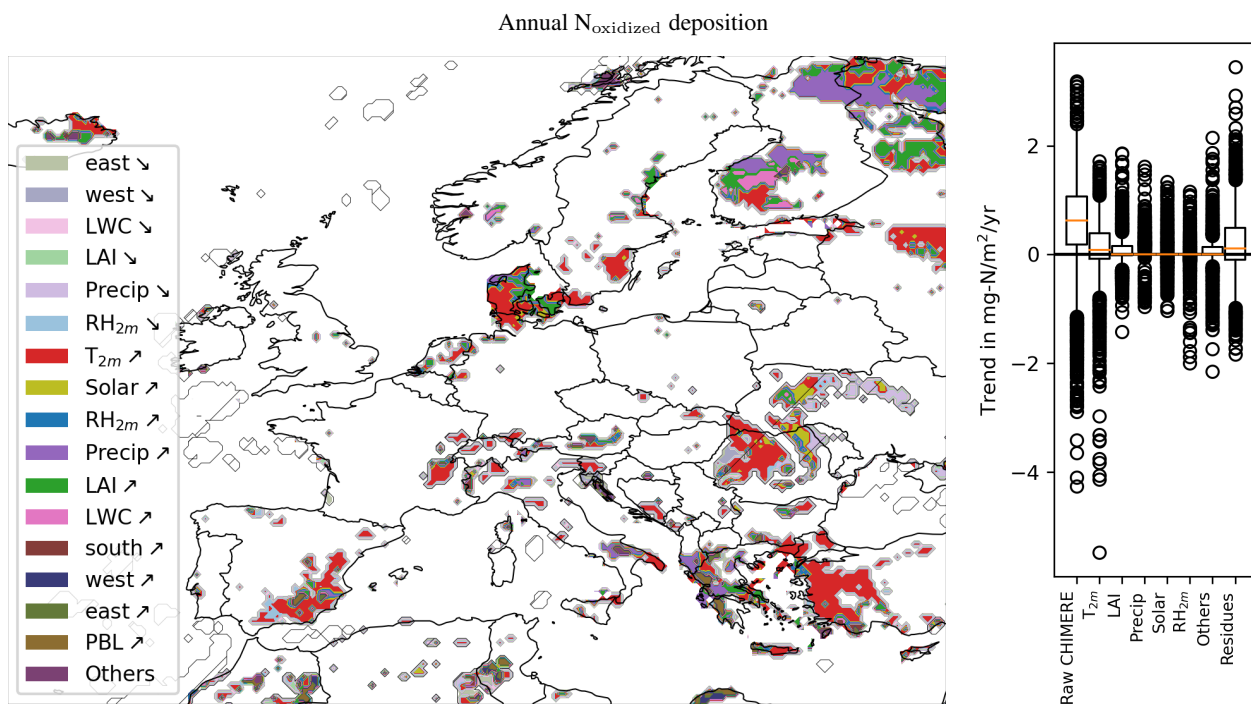


Figure S34. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for annual total (dry+wet) deposition of NO_x .

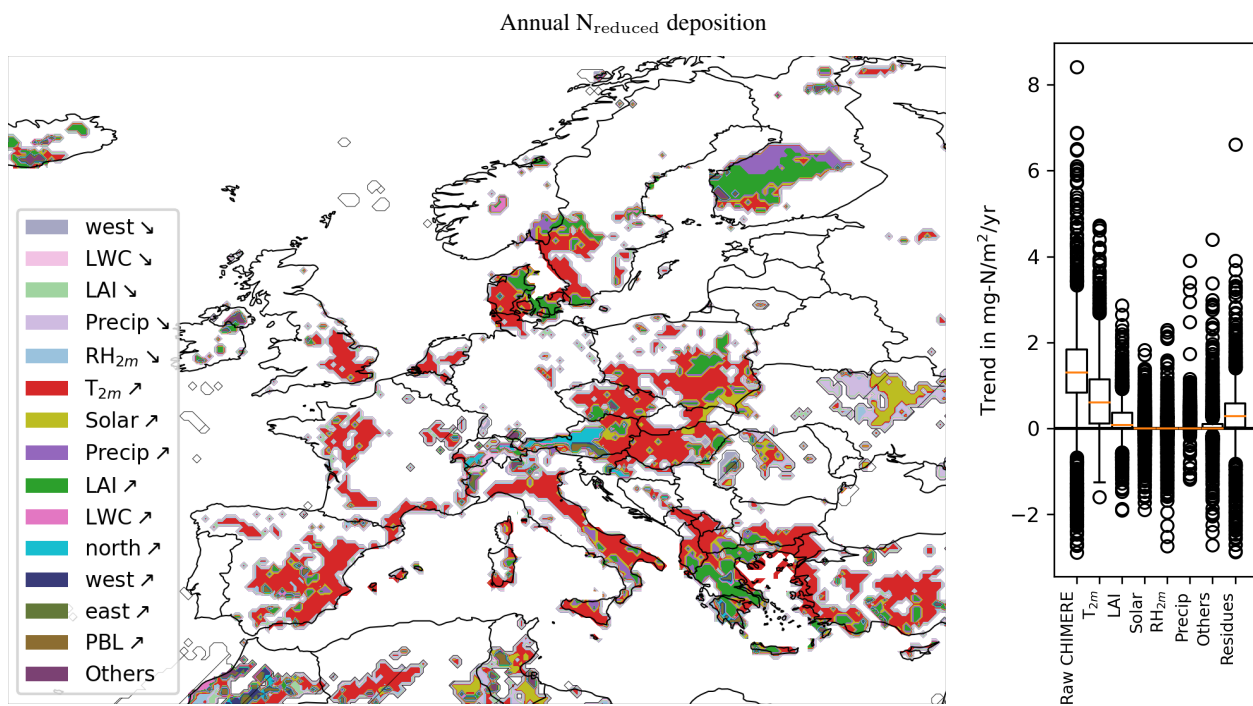


Figure S35. Map of the dominant meteorological driver (left) and box plot of the estimated contribution over EUR of each driver (right) for annual total (dry+wet) deposition of NH_x .

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