

Supplementary Information

National and transboundary contributions to surface ozone concentration across European countries

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1 Supplementary Note 1: Configuration of the CALIOPE system

Supplementary Table S1. Detailed set up of the CALIOPE system

Meteorological model	WRF-ARWv3.5.1
Initial and boundary meteorological conditions	ERA-Interim (79 x 79 km ²)
Meteorological spin-up period	12 hours
WRF domain 18 km resolution (nx,ny,nz)	294 x 294 x 37
WRF parametrizations	Boundary layer: YSU Microphysics scheme: WSM5 Cumulus scheme: no cumulus parametrization Land Surf physics: unified Noah land-surface model Long Wave: RRTM Short Wave: Dudhia Surface Layer: Revised MM5 Monin-Obukhov scheme
Anthropogenic emission model	HERMESv3
Anthropogenic emissions	CAMS-REG-APv4.2 2015, 2016 and 2017
Biogenic emission model	MEGANv2.0.1
Chemical Transport Model	CMAQv5.0.2
Chemical mechanism	cb05-TUCL-aero6
Chemical boundary conditions	CAMS analysis (79 x 79 km ²)
CTM spin-up period	15 days
CTM domain 18 km resolution (nx,ny,nz)	292 x 292 x 37
CMAQ parametrizations	Horizontal advection scheme: Yamartino mass-conserving Vertical advection scheme: Piecewise Parabolic Method (PPM) Vertical diffusion: Asymmetric Convective Model v2 (ACM2) Eddy diffusivity approach Dry deposition routine: Models-3 + Cl species
ISAM tagged species	O ₃ NOx species: NO, NO ₂ , nitrogen trioxide (NO ₃), dinitrogen pentoxide (N ₂ O ₅), nitrous acid (HONO), peroxyacyl nitrates (PAN), higher peroxyacyl nitrates (PANX), peroxyntiric acid (PNA), and organic nitrates (NTR) VOCs species: acetaldehyde (ALD2), higher aldehydes (ALDX), ethene (ETH), ethane (ETHA), ethanol (ETOH), formaldehyde (FORM), internal olefin (IOLE), isoprene (ISOP), methanol (MEOH), olefin (OLE), paraffin (PAR), monoterpene (TERP), toluene (TOL), and xylene (XYL)



Supplementary Figure S1. Boundaries of the study domain.

2 Supplementary Note 2: Quality assurance with GHOST

- 20 We applied a quality assurance screening to the O₃ hourly observations using the metadata available in GHOST (Globally Harmonised Observational Surface in Space and Time). The air quality assurance flags used are described in Table S2.

Supplementary Table S2. Description of the GHOST quality-assurance flags used on the EEA air quality observational dataset.

Flag	Description
0	Measurement is missing (i.e. NaN).
1	Value is infinite – occurs when data values are outside of the range that <i>float32</i> data type can handle (-3.4E+38 to +3.4E+38).
2	Measurement is negative in absolute terms.
3	Measurement is equal to zero.
6	Measurements are associated with data quality flags given by the data provider which have been decreed by the GHOST project architects as being associated with substantial uncertainty/bias.
8	After screening by key QA flags, no valid data remains to average in the temporal window.
10	The measurement methodology used has not yet been mapped to standardised dictionaries of measurement methodologies.
18	The specific name of the measurement method is unknown.
20	The primary sampling is not appropriate to prepare the specific parameter for subsequent measurement.
21	The sample preparation is not appropriate to prepare the specific parameter for subsequent measurement.
22	The measurement methodology used is not known to be able to measure the specific parameter.
72	Measurement is below or equal to the preferential lower limit of detection.
75	Measurement is above or equal to the preferential upper limit of detection.
82	The preferential resolution for the measurement is coarser than a set limit (variable by measured parameter).
83	The resolution of the measurement is analysed month by month. If the minimum difference between observations is coarser than a set limit (variable by measured parameter), measurements are flagged.
90	Check for persistently recurring values. Check is done by using a moving window of 9 measurements. If 5/6 (i.e. 83.33%) of values in the window are the same then the entire window is flagged.
91	Check for persistently recurring values. Check is done by using a moving window of 12 measurements. If 9/12 (i.e. 75%) of values in the window are the same, then the entire window is flagged.
92	Check for persistently recurring values. Check is done by using a moving window of 24 measurements. If 16/24 (i.e. 66.66%) of values in the window are the same, then the entire window is flagged.
110	The measured value is below or greater than scientifically feasible lower/upper limits (variable by parameter).
111	The median of the measurements in a month is greater than a scientifically feasible limit (variable by parameter).
112	Data has been reported to be an outlier through data flags by the network data reporters (and not manually checked and verified as valid).
113	Data has been found and decreed manually to be an outlier.
131	2 out of 3 months' distributions are classed as Zone 6 or higher, suggesting there are potentially systematic reasons for the inconsistent distributions across the 3 months.
132	4 out of 6 months' distributions are classed as Zone 6 or higher, suggesting there are potentially systematic reasons for the inconsistent distributions across the 6 months.
133	8 out of 12 months' distributions are classed as Zone 6 or higher, suggesting there are potentially systematic reasons for the inconsistent distributions across the 12 months.

3 Supplementary Note 3: Model Evaluation

We evaluate the simulated MDA8 O₃ surface concentrations against air quality measurements from the AQ eReporting database of the European Environmental Agency [1]. We first evaluate the model following the benchmarking methodology developed by the Forum for Air Quality Modelling in Europe (FAIRMODE). The evaluation procedure is described in the Guidance Document on Modelling Quality Objectives and Benchmarking [2]. The Modelling Quality Indicator (MQI) is calculated with the following formula: $MQI_i = |M_i - O_i| / (\beta U(O_i))$ where M_i and O_i are the values of the model and the observation, $U(O_i)$ the uncertainty at 95% of the observation O_i , and β a factor set to a value of 2 by FAIRMODE experts. In this context, a prediction is considered of reasonable quality if its MQI is below 1 and the Modelling Quality Objective (MQO) is fulfilled. Considering a time series at one station, the global MQI of the series is calculated with the following formulas:

$$MQI = \frac{\sqrt{\sum_{i=1}^N (O_i - M_i)^2 / N}}{\beta \sqrt{\sum_{i=1}^N U(O_i)^2 / N}} \quad (1a)$$

$$U(O_i) = U_r(RV) \sqrt{(1 - \alpha^2) O_i^2 + \alpha^2 RV^2} \quad (1b)$$

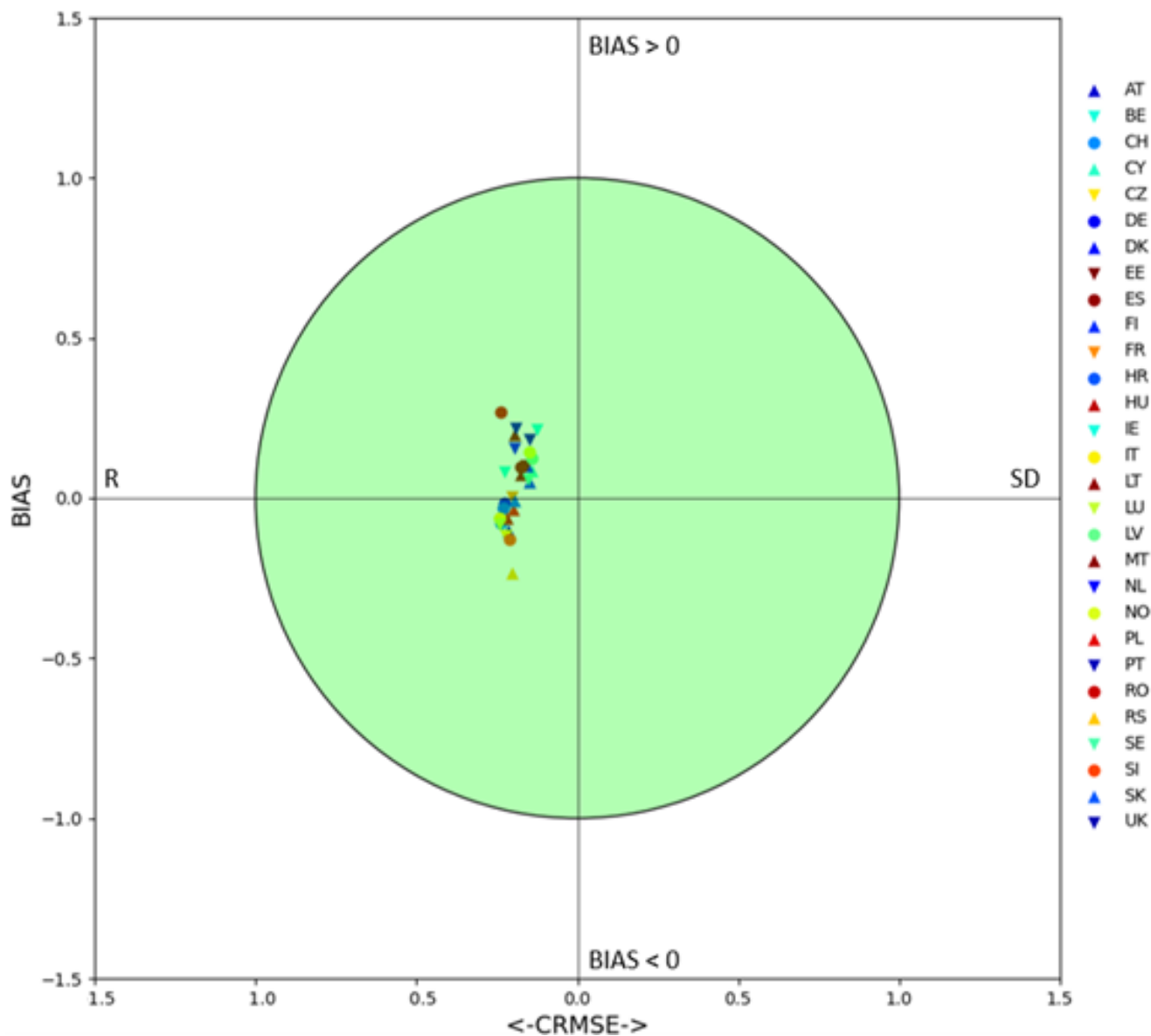
where $RV = 120 \mu\text{g}/\text{m}^3$ (*the reference value*), $\alpha = 0.79$ and $U_r(RV) = 0.18$ for O₃. All the stations evaluated achieved an MQI below 1, accomplishing the MQO. To represent the good performance of the model for each country and aggregate this information, we did a target diagram (Fig. S2). In the target diagram, the X and Y axis represents the BIAS and CRMSE normalized by the measurement uncertainty, RMS_u . The CRMSE is related to the RMSE and the BIAS as follows: $RMSE^2 = BIAS^2 + CRMSE^2$. CRMSE, RMSE, and RMS_u are defined as:

$$CRMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N [(M_i - \bar{M}) - (O_i - \bar{O})]^2} \quad (2a)$$

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (O_i - M_i)^2} \quad (2b)$$

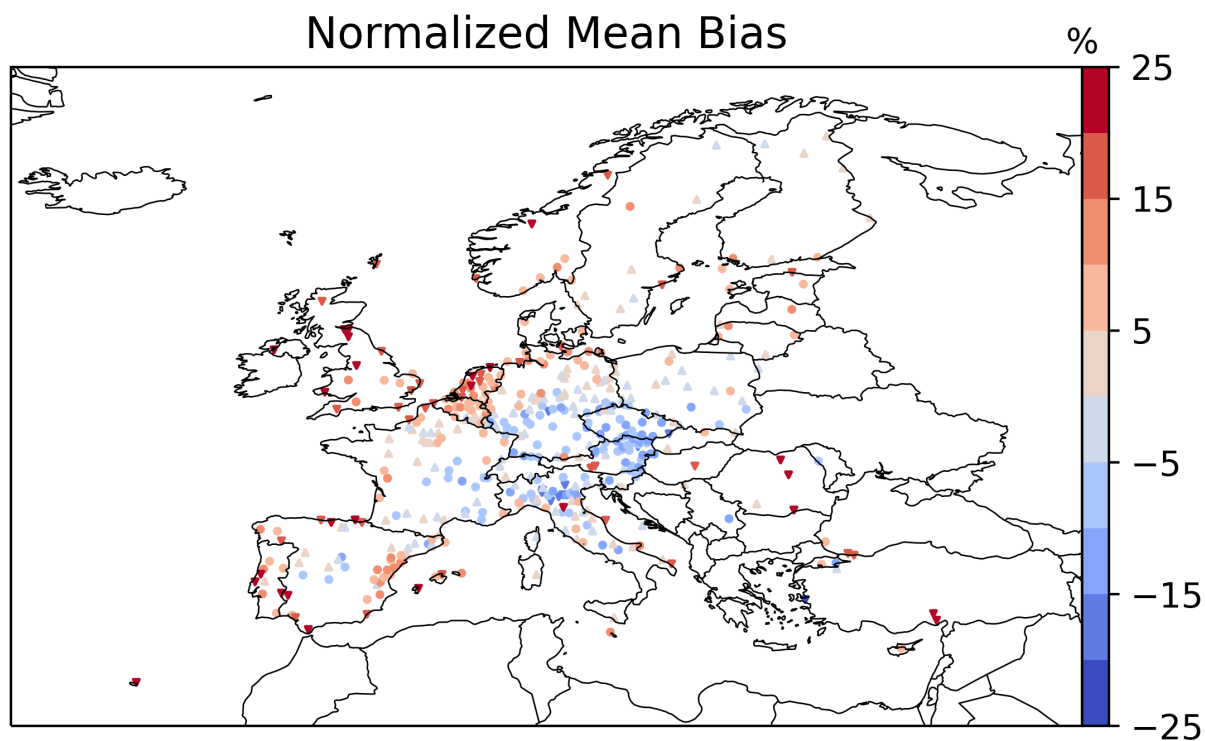
$$RMS_u = U_{95,r}^{RV} \sqrt{(1 - \alpha^2)(\bar{O}^2 + \sigma_o^2) + \alpha^2 RV^2} \quad (2c)$$

For the mean of all the stations of each country, a dot is placed on the target diagram, where the green area of the target plot identifies the fulfillment of the MQO. The graph also indicates if the error is dominated by the SD or the r, where the left side of the plot is dominated by the r and the right side by the SD. We accomplish the MQO of the FAIRMODE, and the error is mainly dominated by the r in all the countries.

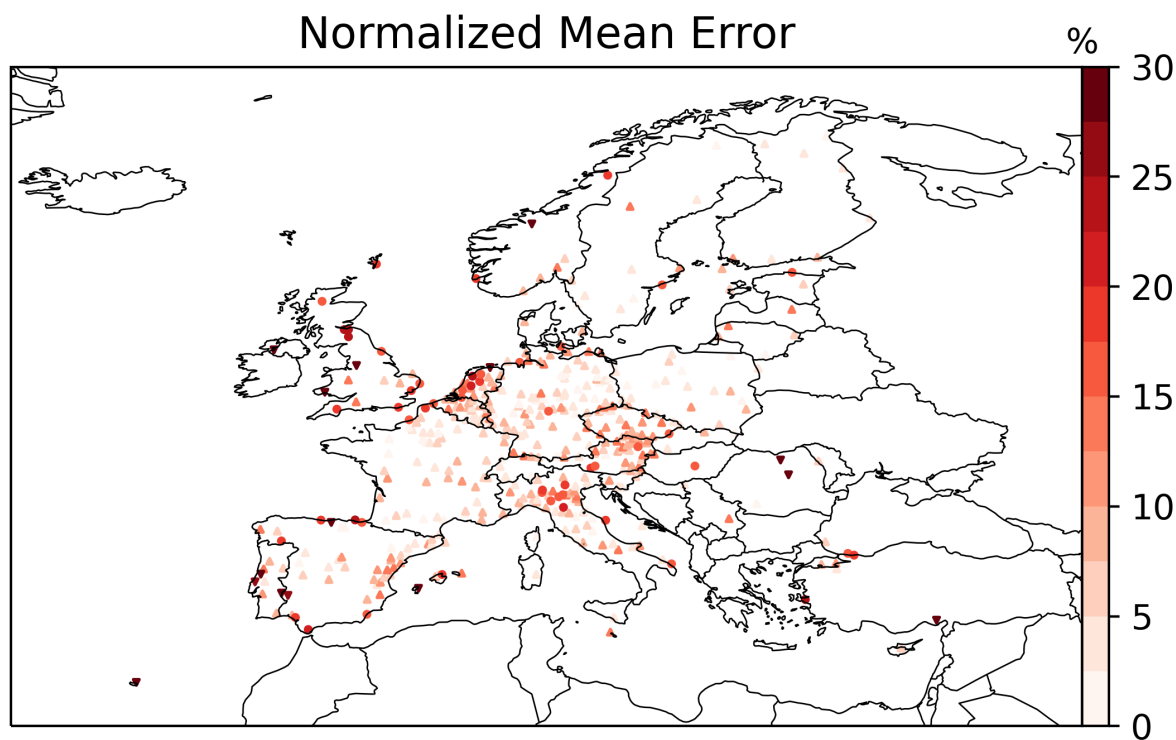


Supplementary Figure S2. Target diagram to visualize the main aspects of the model performance to the MDA8 O₃ with the aggregate information of the mean values of all the stations by European country. The green area represents the achievement of the MQO.

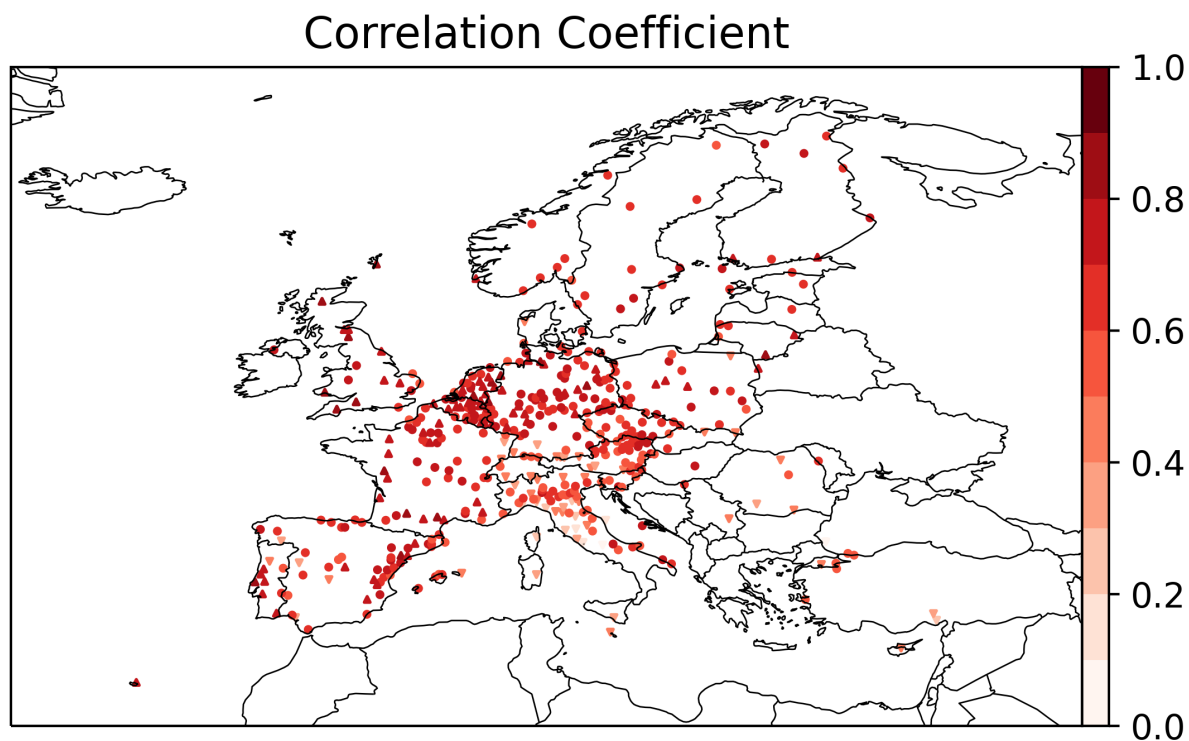
45 Additionally, we evaluate the model with some discrete statistics, including the Normalized Mean Bias (NMB), the Normalized Mean Error (NME), and the correlation coefficient (r). The mean results of the three years of these statistics for each station are shown in Figs. S3, S4 and S5. We aggregate these results for each country and each year at Tables S3, S4, and S5.



Supplementary Figure S3. Normalized Mean Bias (NMB) of the MDA8 O₃ over June-July-August 2015, 2016, and 2017 for all EEA rural background stations below 1000 m a.s.l.. According to the recommendations defined by Emery et al. [3] (NMB; goal: $\pm 5\%$; criteria: $\pm 15\%$), $\triangle$ represents the stations that fulfill the goal, $\circ$ represents the stations between the goal and the criteria, ∇ represents the stations below the criteria.



Supplementary Figure S4. Normalized Mean Error (NME) of the MDA8 O₃ over June-July-August 2015, 2016, and 2017 for all EEA rural background stations below 1000 m a.s.l.. According to the recommendations defined by Emery et al. [3] (NME; goal: 15%; criteria: <25%), $\triangle$ represents the stations that fulfill the goal, $\bigcirc$ represents the stations between the goal and the criteria, ∇ represents the stations below the criteria.



Supplementary Figure S5. Correlation Coefficient (r) of the MDA8 O_3 over June-July-August 2015, 2016, and 2017 for all EEA rural background stations below 1000 m a.s.l.. According to the recommendations defined by Emery et al. [3] (r ; goal: >0.75 ; criteria: > 0.50), $\triangle$ represents the stations that fulfill the goal, $\circ$ represents the stations between the goal and the criteria, ∇ represents the stations below the criteria.

Supplementary Table S3. Statistical overview of MDA8 O₃ concentrations at rural background stations located below 1000m above sea level in each tagged country over June-July-August 2015. The metrics are mean bias (MB), normalized mean bias (NMB), mean error (ME), normalized mean error (NME) and correlation coefficient (r). Also shown are the number of observations (N), the mean of the observations (MO), and the modelled mean (MM).

Country	MB (ppb)	NMB (%)	ME (ppb)	NME (%)	r	N	MO (ppb)	MM (ppb)
Albania (AL)	nan	nan	nan	nan	nan	0	nan	nan
Austria (AT)	-6.66	-11.8	9.81	17.35	0.71	2882	56.53	49.87
Belgium (BE)	1.46	3.1	7.29	15.68	0.82	1681	46.50	47.96
Bulgaria (BG)	nan	nan	nan	nan	nan	0	nan	nan
Switzerland (CH)	-5.05	-8.7	9.37	16.17	0.62	634	57.93	52.88
Czech Republic (CZ)	-7.02	-12.6	10.15	18.28	0.78	1860	55.52	48.51
Cyprus (CY)	2.70	4.7	4.22	7.42	0.59	89	56.89	59.58
Germany (DE)	-3.53	-6.8	8.94	17.24	0.79	6647	51.86	48.34
Denmark (DK)	1.62	3.7	5.58	12.85	0.49	181	43.42	45.04
Estonia (EE)	2.54	6.5	4.89	12.47	0.69	272	39.25	41.79
Greece (EL)	-0.16	-0.3	3.72	6.80	0.48	76	54.76	54.60
Spain (ES)	3.31	6.3	7.01	13.40	0.65	4530	52.34	55.65
Finland (FI)	1.39	4.2	4.45	13.41	0.69	969	33.15	34.54
France (FR)	-1.57	-3.1	7.00	13.90	0.73	4613	50.36	48.79
Croatia (HR)	1.08	2.2	9.68	19.92	0.67	259	48.58	49.66
Hungary (HU)	-6.44	-11.6	9.74	17.52	0.75	183	55.56	49.12
Ireland (IE)	8.68	29.3	8.82	29.78	0.40	77	29.61	38.29
Iceland (IS)	nan	nan	nan	nan	nan	0	nan	nan
Italy (IT)	-3.09	-5.1	8.88	14.63	0.55	5118	60.71	57.62
Liechtenstein (LI)	nan	nan	nan	nan	nan	0	nan	nan
Lithuania (LT)	0.11	0.2	5.64	13.35	0.63	366	42.25	42.36
Luxembourg (LU)	-4.95	-9.4	8.14	15.51	0.82	272	52.50	47.55
Latvia (LV)	2.90	8.1	5.06	14.16	0.64	92	35.71	38.62
Montenegro (ME)	nan	nan	nan	nan	nan	0	nan	nan
Malta (MT)	9.69	17.5	10.76	19.47	0.26	74	55.26	64.95
Netherlands (NL)	4.62	10.8	7.74	18.06	0.77	1569	42.88	47.50
Norway (NO)	4.15	11.4	6.04	16.60	0.51	707	36.36	40.50
Poland (PL)	-5.35	-10.4	7.88	15.29	0.76	1613	51.53	46.18
Portugal (PT)	8.67	20.2	9.89	23.00	0.79	1004	43.02	51.69
Romania (RO)	8.22	20.9	14.48	36.90	0.43	520	39.26	47.48
Serbia (RS)	-10.17	-16.8	11.09	18.32	0.65	88	60.54	50.38
Sweden (SE)	1.79	4.9	4.94	13.49	0.63	903	36.62	38.42
Slovenia (SI)	-5.23	-9.4	7.87	14.10	0.71	270	55.85	50.62
Slovakia (SK)	-2.31	-4.6	8.08	15.95	0.50	321	50.69	48.38
United Kingdom (UK)	5.86	16.0	7.01	19.11	0.69	1781	36.66	42.52
Europe	-0.78	-1.6	8.27	16.37	0.71	41278	50.53	49.75

Supplementary Table S4. Statistical overview of MDA8 O₃ concentrations at rural background stations located below 1000m above sea level in each tagged country over June-July-August 2016. The metrics are mean bias (MB), normalized mean bias (NMB), mean error (ME), normalized mean error (NME) and correlation coefficient (r). Also shown are the number of observations (N), the mean of the observations (MO), and the modelled mean (MM).

Country	MB (ppb)	NMB (%)	ME (ppb)	NME (%)	r	N	MO (ppb)	MM (ppb)
Albania (AL)	nan	nan	nan	nan	nan	0	nan	nan
Austria (AT)	-0.49	-1.1	6.12	13.15	0.50	2881	46.58	46.09
Belgium (BE)	5.38	14.1	7.79	20.40	0.81	1678	38.18	43.56
Bulgaria (BG)	nan	nan	nan	nan	nan	0	nan	nan
Switzerland (CH)	-0.22	-0.5	7.02	14.56	0.60	640	48.23	48.01
Czech Republic (CZ)	-1.72	-3.7	6.07	13.12	0.62	1973	46.30	44.57
Cyprus (CY)	3.25	5.7	4.84	8.51	0.47	89	56.88	60.12
Germany (DE)	1.04	2.4	6.62	15.36	0.74	6584	43.11	44.15
Denmark (DK)	4.30	11.2	5.93	15.47	0.73	174	38.36	42.67
Estonia (EE)	5.38	14.7	6.68	18.21	0.64	272	36.68	42.06
Greece (EL)	nan	nan	nan	nan	nan	0	nan	nan
Spain (ES)	3.19	6.4	6.87	13.72	0.61	4699	50.12	53.30
Finland (FI)	1.98	6.0	4.63	13.95	0.63	845	33.19	35.17
France (FR)	2.48	5.8	6.83	16.03	0.78	4585	42.59	45.07
Croatia (HR)	0.68	1.4	6.04	12.76	0.43	287	47.32	48.00
Hungary (HU)	-1.84	-3.8	6.45	13.49	0.53	164	47.81	45.97
Ireland (IE)	7.20	23.6	7.66	25.09	0.77	346	30.53	37.73
Iceland (IS)	nan	nan	nan	nan	nan	0	nan	nan
Italy (IT)	-0.16	-0.3	8.16	14.96	0.41	5340	54.58	54.42
Liechtenstein (LI)	nan	nan	nan	nan	nan	0	nan	nan
Lithuania (LT)	2.98	7.7	6.12	15.80	0.65	385	38.70	41.68
Luxembourg (LU)	0.94	2.2	6.61	15.84	0.82	269	41.71	42.65
Latvia (LV)	5.94	18.2	6.61	20.28	0.59	92	32.60	38.54
Montenegro (ME)	nan	nan	nan	nan	nan	0	nan	nan
Malta (MT)	7.96	15.1	8.62	16.31	0.43	92	52.87	60.83
Netherlands (NL)	6.18	16.7	7.87	21.19	0.78	1573	37.13	43.32
Norway (NO)	5.87	17.9	7.20	21.93	0.52	720	32.83	38.69
Poland (PL)	0.46	1.1	5.88	13.58	0.70	1639	43.28	43.74
Portugal (PT)	6.63	14.8	9.71	21.63	0.70	996	44.91	51.54
Romania (RO)	12.33	37.5	17.07	51.89	0.06	380	32.90	45.23
Serbia (RS)	-6.50	-12.1	8.81	16.44	0.42	86	53.58	47.08
Sweden (SE)	2.29	6.5	5.17	14.69	0.62	967	35.17	37.46
Slovenia (SI)	-2.14	-4.3	5.58	11.13	0.55	276	50.16	48.01
Slovakia (SK)	2.09	4.8	6.54	15.07	0.40	348	43.38	45.47
United Kingdom (UK)	7.64	23.1	8.22	24.90	0.76	1854	33.03	40.67
Europe	2.27	5.1	7.16	16.11	0.72	41771	44.42	46.69

Supplementary Table S5. Statistical overview of MDA8 O₃ concentrations at rural background stations located below 1000m above sea level in each tagged country over June-July-August 2016. The metrics are mean bias (MB), normalized mean bias (NMB), mean error (ME), normalized mean error (NME) and correlation coefficient (r). Also shown are the number of observations (N), the mean of the observations (MO), and the modelled mean (MM).

Country	MB (ppb)	NMB (%)	ME (ppb)	NME (%)	r	N	MO (ppb)	MM (ppb)
Albania (AL)	nan	nan	nan	nan	nan	0	nan	nan
Austria (AT)	-3.88	-7.6	7.55	14.75	0.55	2887	51.16	47.28
Belgium (BE)	1.87	4.4	6.35	15.08	0.83	1723	42.12	43.99
Bulgaria (BG)	nan	nan	nan	nan	nan	0	nan	nan
Switzerland (CH)	-3.29	-6.5	7.34	14.56	0.63	724	50.40	47.10
Czech Republic (CZ)	-3.82	-7.7	6.60	13.35	0.67	2009	49.45	45.64
Cyprus (CY)	3.46	6.1	4.79	8.50	0.41	76	56.37	59.83
Germany (DE)	0.50	1.1	6.30	14.36	0.73	6682	43.85	44.35
Denmark (DK)	4.70	12.5	5.48	14.62	0.62	181	37.50	42.21
Estonia (EE)	2.86	7.9	4.99	13.81	0.67	275	36.17	39.03
Greece (EL)	nan	nan	nan	nan	nan	0	nan	nan
Spain (ES)	3.73	7.6	7.30	14.81	0.61	4597	49.32	53.05
Finland (FI)	1.85	6.0	4.60	14.77	0.59	715	31.12	32.98
France (FR)	-0.52	-1.2	5.71	12.69	0.79	4531	45.03	44.51
Croatia (HR)	-5.49	-9.9	8.65	15.59	0.47	294	55.47	49.97
Hungary (HU)	-2.93	-5.8	9.33	18.36	0.41	284	50.80	47.86
Ireland (IE)	6.28	21.1	6.68	22.47	0.62	343	29.72	35.99
Iceland (IS)	nan	nan	nan	nan	nan	0	nan	nan
Italy (IT)	-3.95	-6.6	9.02	15.13	0.37	5539	59.59	55.64
Liechtenstein (LI)	nan	nan	nan	nan	nan	0	nan	nan
Lithuania (LT)	4.80	13.6	6.01	17.09	0.56	403	35.15	39.95
Luxembourg (LU)	-4.64	-9.8	6.82	14.47	0.85	276	47.12	42.48
Latvia (LV)	4.17	12.9	5.18	15.98	0.45	81	32.40	36.57
Montenegro (ME)	nan	nan	nan	nan	nan	0	nan	nan
Malta (MT)	3.95	6.8	6.91	11.96	0.50	92	57.72	61.68
Netherlands (NL)	5.66	14.7	7.69	19.97	0.72	1523	38.48	44.14
Norway (NO)	5.23	15.8	6.49	19.62	0.26	698	33.05	38.28
Poland (PL)	1.01	2.4	5.62	13.05	0.64	1748	43.07	44.08
Portugal (PT)	7.26	17.2	8.99	21.31	0.68	900	42.18	49.44
Romania (RO)	8.08	20.9	10.74	27.76	0.27	739	38.68	46.76
Serbia (RS)	-8.75	-15.1	9.67	16.71	0.57	70	57.89	49.13
Sweden (SE)	1.93	5.6	4.39	12.69	0.59	999	34.63	36.56
Slovenia (SI)	-6.65	-11.7	8.25	14.57	0.65	266	56.63	49.99
Slovakia (SK)	-1.08	-2.3	6.51	13.59	0.39	358	47.88	46.79
United Kingdom (UK)	5.38	16.0	6.57	19.50	0.69	1631	33.67	39.05
Europe	0.47	1.0	7.04	15.17	0.73	42126	46.42	46.88

4 Supplementary Note 4: MDA8 O₃ contribution in absolute values over 35 European countries

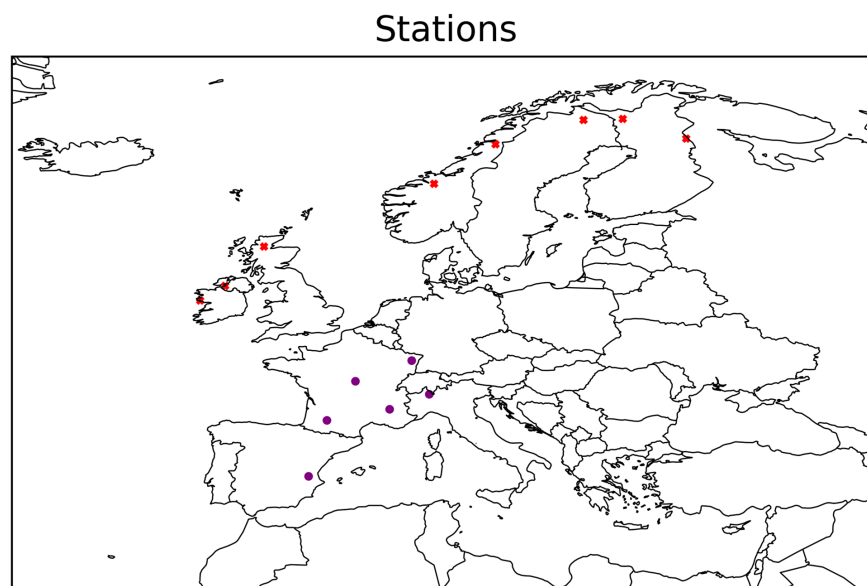
Supplementary Table S6. Average MDA8 O₃ contribution in µg/m³ over 35 European countries for summer 2015, 2016, and 2017. The contributions include the national O₃ contribution (NATIONAL), the imported O₃ in each country originated from each of the remaining 34 countries (EUC), along with the contributions from hemispheric transport (BCON), non-European neighboring countries (NOEU35) and maritime sources (SEA). In parenthesis, the percentage contribution of each source.

Countries	NATIONAL (µg/m ³)	EUC (µg/m ³)	NOEU35 (µg/m ³)	SEA (µg/m ³)	BCON (µg/m ³)	Total (µg/m ³)
Italy (IT)	20.69 (17%)	27.42 (22%)	4.35 (4%)	12.80 (10%)	58.62 (47%)	123.86
France (FR)	16.30 (17%)	16.01 (16%)	1.16 (1%)	8.02 (8%)	56.68 (58%)	98.17
Germany (DE)	16.31 (16%)	24.72 (25%)	1.20 (1%)	5.76 (6%)	51.74 (52%)	99.73
Spain (ES)	17.13 (15%)	13.78 (12%)	3.69 (3%)	12.31 (11%)	68.65 (59%)	115.57
Poland (PL)	12.39 (13%)	24.30 (25%)	3.89 (4%)	5.55 (6%)	51.66 (53%)	97.79
Portugal (PT)	13.68 (12%)	16.09 (15%)	2.02 (2%)	13.50 (12%)	65.51 (59%)	110.80
Romania (RO)	9.67 (10%)	20.50 (20%)	11.35 (11%)	3.48 (3%)	55.77 (56%)	100.77
United Kingdom (UK)	7.58 (9%)	8.08 (10%)	0.55 (1%)	6.42 (8%)	61.41 (73%)	84.04
Serbia (RS)	8.65 (8%)	31.63 (29%)	8.44 (8%)	3.92 (4%)	55.09 (51%)	107.73
Switzerland (CH)	8.44 (8%)	35.99 (32%)	1.64 (1%)	5.11 (5%)	60.87 (54%)	112.04
Austria (AT)	8.12 (7%)	39.22 (36%)	2.53 (2%)	4.43 (4%)	56.16 (51%)	110.45
Czech Republic (CZ)	7.54 (7%)	37.63 (36%)	2.30 (2%)	4.33 (4%)	51.65 (50%)	103.44
Bulgaria (BG)	7.45 (7%)	23.33 (22%)	12.85 (12%)	4.32 (4%)	58.88 (55%)	106.83
Netherlands (NL)	6.66 (7%)	27.97 (29%)	0.77 (1%)	7.91 (8%)	54.03 (55%)	97.35
Belgium (BE)	6.24 (6%)	31.16 (32%)	0.78 (1%)	6.92 (7%)	53.04 (54%)	98.14
Hungary (HU)	6.48 (6%)	36.62 (35%)	5.33 (5%)	3.94 (4%)	52.28 (50%)	104.65
Greece (EL)	7.17 (6%)	25.46 (21%)	14.04 (12%)	9.86 (8%)	63.69 (53%)	120.21
Slovenia (SI)	6.41 (6%)	44.12 (38%)	3.75 (3%)	6.32 (6%)	54.05 (47%)	114.65
Croatia (HR)	5.52 (5%)	39.94 (35%)	6.77 (7%)	7.75 (7%)	54.98 (48%)	114.95
Albania (AL)	5.70 (5%)	33.13 (27%)	11.12 (9%)	7.91 (6%)	64.93 (53%)	122.80
Slovakia (SK)	4.87 (5%)	38.94 (37%)	4.58 (4%)	4.02 (4%)	53.60 (51%)	106.01
Finland (FI)	2.91 (4%)	8.86 (13%)	2.52 (2%)	4.67 (4%)	51.96 (73%)	70.92
Norway (NO)	2.82 (4%)	7.87 (10%)	1.09 (1%)	5.78 (7%)	62.01 (78%)	79.57
Ireland (IE)	2.66 (3%)	5.69 (7%)	0.38 (0%)	4.53 (6%)	65.49 (83%)	78.75
Lithuania (LT)	2.93 (3%)	21.12 (24%)	6.03 (7%)	6.71 (8%)	51.12 (58%)	87.92
Malta (MT)	4.42 (3%)	38.90 (28%)	6.33 (5%)	36.25 (26%)	51.37 (37%)	137.28
Sweden (SE)	2.30 (3%)	10.44 (14%)	1.42 (2%)	5.81 (8%)	56.10 (74%)	76.07
Latvia (LV)	2.25 (3%)	19.65 (23%)	4.93 (6%)	7.24 (9%)	50.51 (60%)	84.59
Denmark (DK)	2.39 (3%)	21.86 (23%)	1.59 (2%)	9.93 (10%)	58.95 (62%)	94.72
Estonia (EE)	1.84 (2%)	18.92 (22%)	4.06 (4%)	8.16 (10%)	51.89 (61%)	84.87
Montenegro (ME)	2.34 (2%)	34.84 (30%)	9.46 (8%)	6.30 (5%)	64.40 (55%)	117.34
Luxembourg (LU)	1.95 (2%)	38.24 (39%)	0.85 (1%)	5.71 (6%)	51.80 (53%)	98.56
Cyprus (CY)	2.47 (2%)	14.66 (11%)	26.75 (20%)	19.67 (15%)	67.83 (52%)	131.37
Iceland (IS)	1.30 (2%)	3.86 (5%)	0.50 (1%)	3.33 (5%)	64.00 (88%)	73.00
Liechtenstein (LI)	0.45 (0%)	47.57 (41%)	1.69 (1%)	4.85 (4%)	60.17 (52%)	114.72

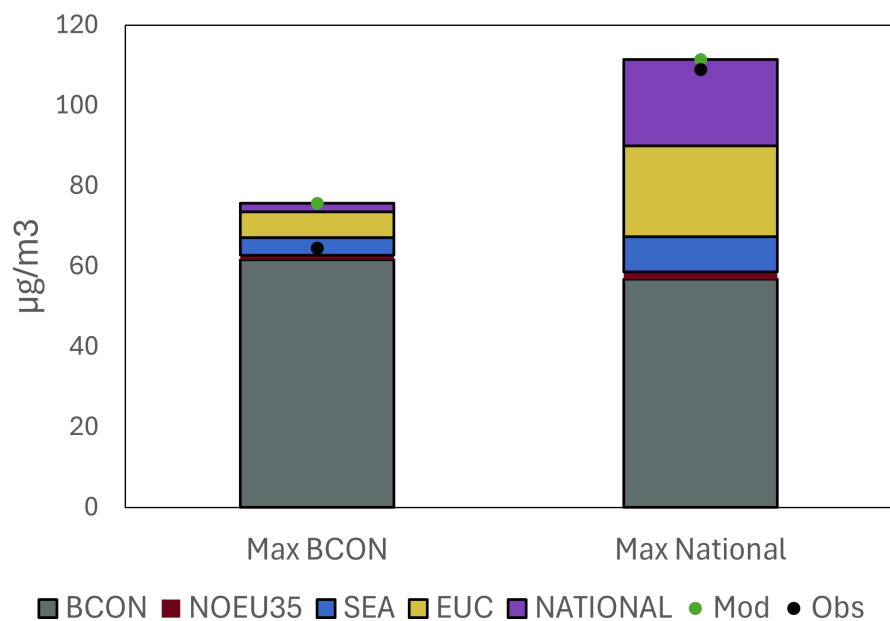
5 Supplementary Note 5: Evaluation of the hemispheric and local/regional photochemical contributions

50 We used O_3 concentrations obtained from remote and regional background EMEP stations across Europe. We specifically assessed the model's performance at stations where the simulated hemispheric contribution is at its peak, as well as stations where the model exhibits the greatest national influence (Fig. S7). This approach allowed scrutinizing the model's ability to capture background conditions and local/regional photochemical enhancements, respectively.

As depicted in Fig. S6, stations mainly influenced by background O_3 (Max BCON) are located along the Atlantic coast. On
55 average there is a positive bias which remains below 17 %. For reference, our model closely aligns with the results obtained by the Copernicus Programme CAMS global reanalysis (CAMSRA), which combines both model and observation-based information to provide a comprehensive estimation of the atmospheric conditions [4]. On the other hand, the model presents a very good agreement in stations heavily influenced by local/regional photochemical production (Max National). Most notably, this comparison highlights that overall the model captures well the transition from background conditions to photochemical O_3
60 production regimes, indicating its ability to reproduce O_3 variability effectively. The good agreement between the background and photochemical increment derived from observations and the numerical simulations instills again stronger confidence in all the discussion and findings of our results.



Supplementary Figure S6. Geographical distribution of the stations with higher contributions from the hemispheric ozone (red) and the stations with higher contributions from national photochemical ozone production (purple).



Supplementary Figure S7. Average simulated source contributions to O₃ and total simulated and measured O₃ in EMEP stations where the contributions of hemispheric O₃ are above the 90th percentile (Max BCON) and where the national contributions are above the 90th percentile (Max National) of all EMEP stations. The geographical distribution of those stations is presented in Figure S6. Contributions include the national O₃ contribution (NATIONAL), the imported O₃ in each country originated from each of the remaining 34 countries (EUC), the contributions from hemispheric transport (BCON), non-European neighboring countries (NOEU35) and maritime sources (SEA). The green dot represents model total O₃ concentration, the black dot the O₃ concentrations from observations, and the red dot the O₃ concentrations from the CAMS reanalysis (CAMSRA).

6 Supplementary Note 6: Comparison of the regional photochemical production with an observational-based analysis

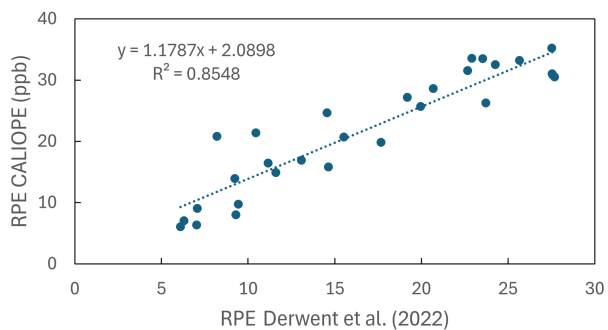
65 Derwent and Parrish [5] investigated long-term changes in the annual maximum 8-hourly (AM8) O₃ mixing ratios from 1989 to 2018 at 32 low-elevation EMEP stations in northwestern Europe. At Mace Head, two contributions were estimated by fitting their sum to the AM8 time series: a background O₃ transported into the region (TB), modeled with a three-term power series, and a regional photochemical enhancement (RPE), modeled as the 2000 RPE multiplied by an exponential decay function of time. The fitted TB contribution at Mace Head was then subtracted from the AM8 at each location, and the resulting RPEs were
70 fitted to the exponential decay function, representing the contribution of local and regional anthropogenic precursor emissions to O₃.

We compared the RPE fitted values from Derwent and Parrish [5] with our simulations at these EMEP stations (Table S7). Due to the short duration of our study, we could not derive AM8 fitted values. Instead, our model-derived RPEs are based on the mean MDA8 for June-August 2015-2016, considering national sources (NATIONAL), imported O₃ from 34 other countries
75 (EUC), and maritime sources (SEA). Our simulated RPEs show a strong correlation with the observationally-derived values from Derwent and Parrish [5] across the 32 stations ($R^2 = 0.85$) (Fig. S8 a).

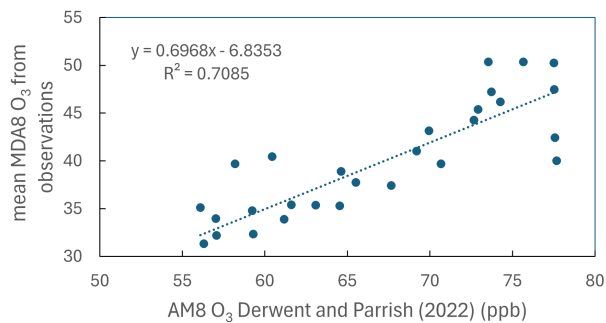
Despite using a different metric, the strong correlation between the AM8 fitted values from Derwent and Parrish [5] and the mean MDA8 observed at each station (Fig. S8 b) demonstrates the relevance of our comparison. This comparison with observational analysis underscores the robustness of our modeling method in disentangling contributions from long-range
80 transport and photochemical production driven by local and regional anthropogenic precursor emissions.

Supplementary Table S7. Comparison of Regional Photochemical Enhancement (RPE) values from Derwent and Parrish [5] and CALIOPE. The RPE values from Derwent and Parrish [5] are based on the average RPE values for 2015, 2016, and 2017, while the CALIOPE values represent the mean RPE for the same years, specifically from June to August.

Station code	Longitude, °E	Latitude, °N	RPE Derwent and Parrish (2022) (ppb)	RPE CALIOPE (ppb)
AT0002R	16.767	47.767	24	33.48
AT0042R	15.047	48.879	23	31.57
AT0043R	15.919	48.106	28	35.22
AT0045R	15.547	48.371	23	33.55
AT0046R	16.731	48.335	24	32.51
AT0047R	16.677	48.051	26	33.22
CH0002R	6.945	46.813	24	26.28
CH0003R	8.905	47.48	28	31.05
DE0001R	8.31	54.926	20	25.69
DE0002R	10.759	52.802	28	30.78
DE0007R	13.033	53.167	21	28.61
DE0009R	12.733	54.433	19	27.20
DK0031R	8.433	56.283	18	19.85
DK0041R	12.126	55.687	8	20.82
FI0009R	21.377	59.779	10	21.41
FI0017R	27.686	60.527	13	16.91
FI0022R	29.402	66.32	6	7.02
IE0031R	9.5	53.167	6	6.09
IT0004R	8.633	45.8	31	38.17
NL0009R	6.277	53.334	15	24.65
NL0010R	5.854	51.451	28	30.52
NO0001R	8.25	58.383	9	13.91
NO0015R	13.917	65.833	9	8.04
NO0039R	8.883	62.783	9	9.74
NO0042G	11.883	78.9	0	1.65
NO0043R	11.533	59	12	14.92
SE0011R	13.15	56.017	16	20.67
SE0012R	17.383	58.8	11	16.43
SE0013R	21.067	67.883	7	6.37
SE0032R	15.567	57.817	15	15.80
SE0035R	19.767	64.25	7	9.03



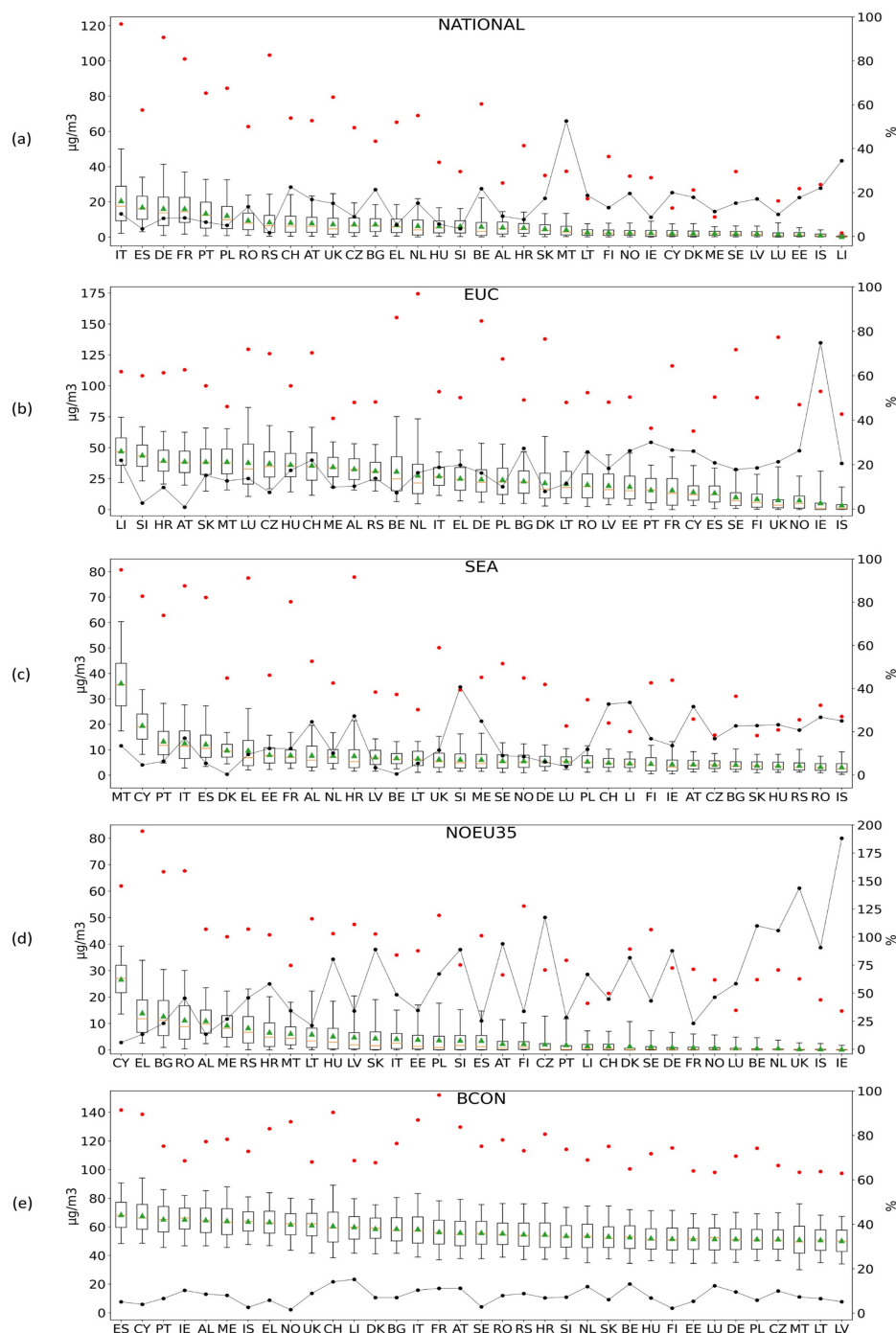
(a)



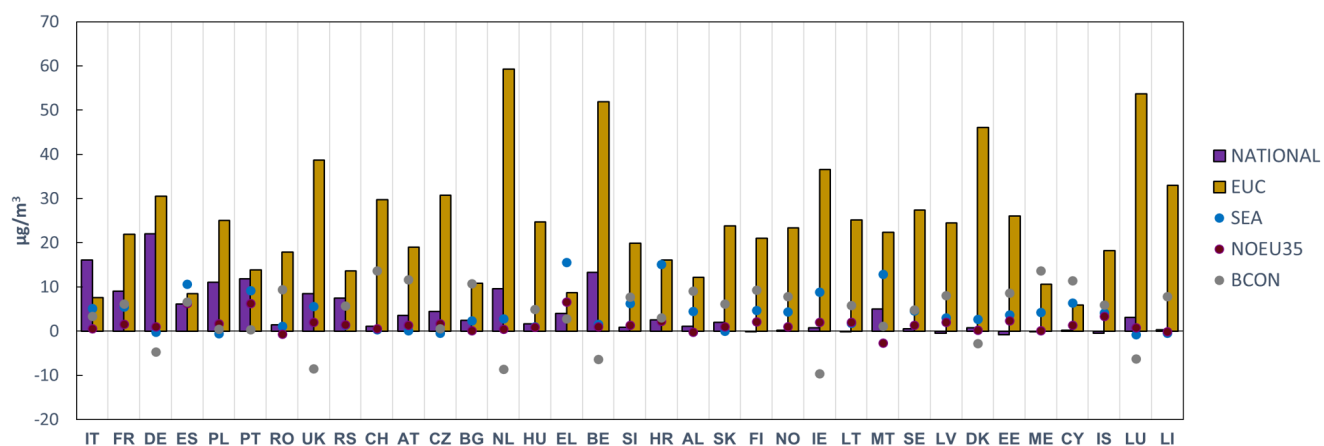
(b)

Supplementary Figure S8. (a) RPEs in Derwent and Parrish [5] vs RPEs estimated from simulated contributions. The RPE values from Derwent and Parrish [5] are based on the average RPE values for 2015, 2016, and 2017, while the CALIOPE values represent the mean RPE for the same years, specifically from June to August. (b) Correlation between the AM8 O₃ fit by Derwent and Parrish [5] and the mean MDA8 O₃ from the observations at each monitoring station from June to August for 2015, 2016 and 2017.

7 Supplementary Note 7: Daily and intracountry variability in absolute values

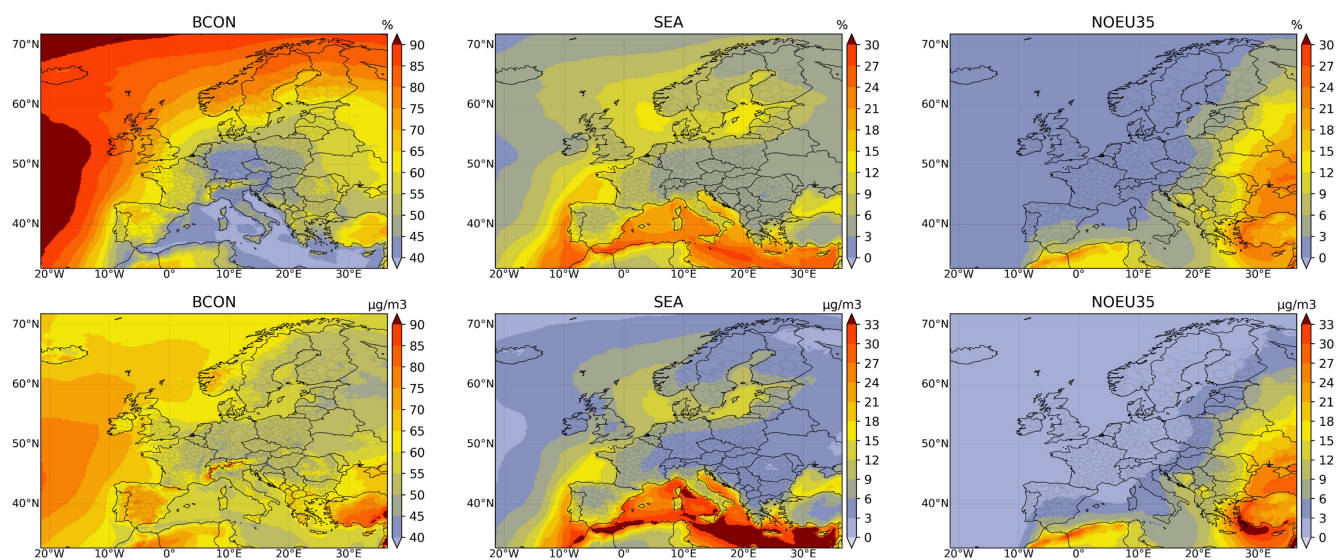


Supplementary Figure S9. Distribution of the model results in absolute values over the whole simulated period and grid cells for each source of contribution (NATIONAL, EUC, SEA, NOEU35 and BCON). Each whisker plot shows the average values (green triangle), the median (yellow line), the lower extreme (p5), the lower quartile (p25), the upper quartile (p75), the upper extreme (p95), and the maximum value (red dot) in absolute values. The black continuous line shows the difference between the maximum and minimum yearly (3-monthly) averages over the three-yearly average.



Supplementary Figure S10. Difference in absolute values between the MDA8 O₃ when O₃ concentrations are above the 95th percentile in each country and the mean MDA8 O₃ by source. The countries are arranged in descending order of their mean NATIONAL contribution as Fig. 4a.

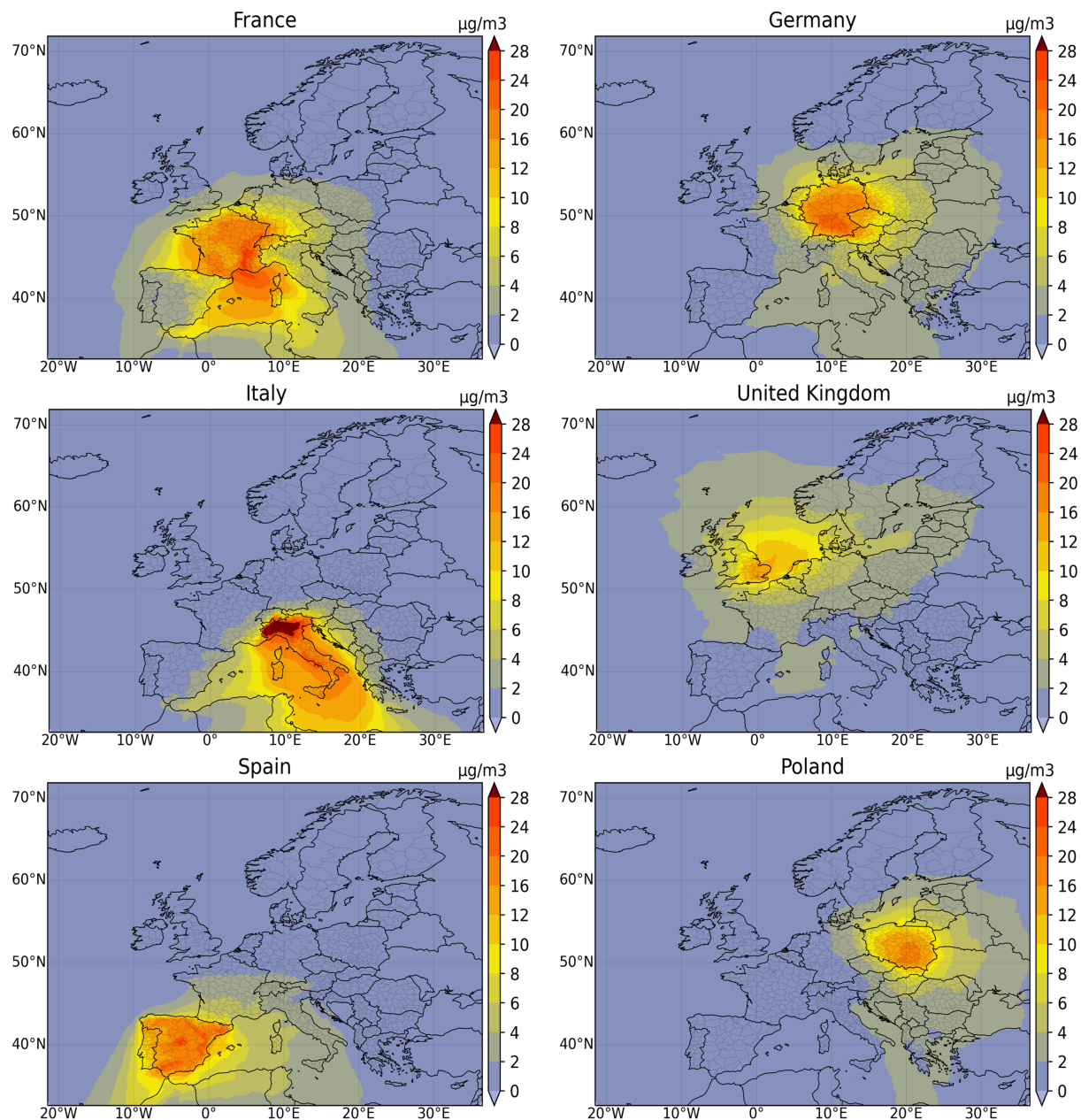
8 Supplementary Note 8: Contributions from hemispheric transport, non-European neighbouring countries and maritime sources to local ozone at model resolution



Supplementary Figure S11. BCON, SEA, and NOEU35 contribution to the MDA8 O₃ over Europe in absolute values (upper panel) and percentage (bottom panel) at model resolution, an average of summer 2015, 2016, and 2017.

9 Supplementary Note 9: Contributions from the principal countries to MDA8 O₃ concentration over Europe in absolute values

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Supplementary Figure S12. Contribution of the countries with the highest contribution to MDA8 O₃ concentration over Europe in absolute values, an average of summer 2015, 2016, and 2017.

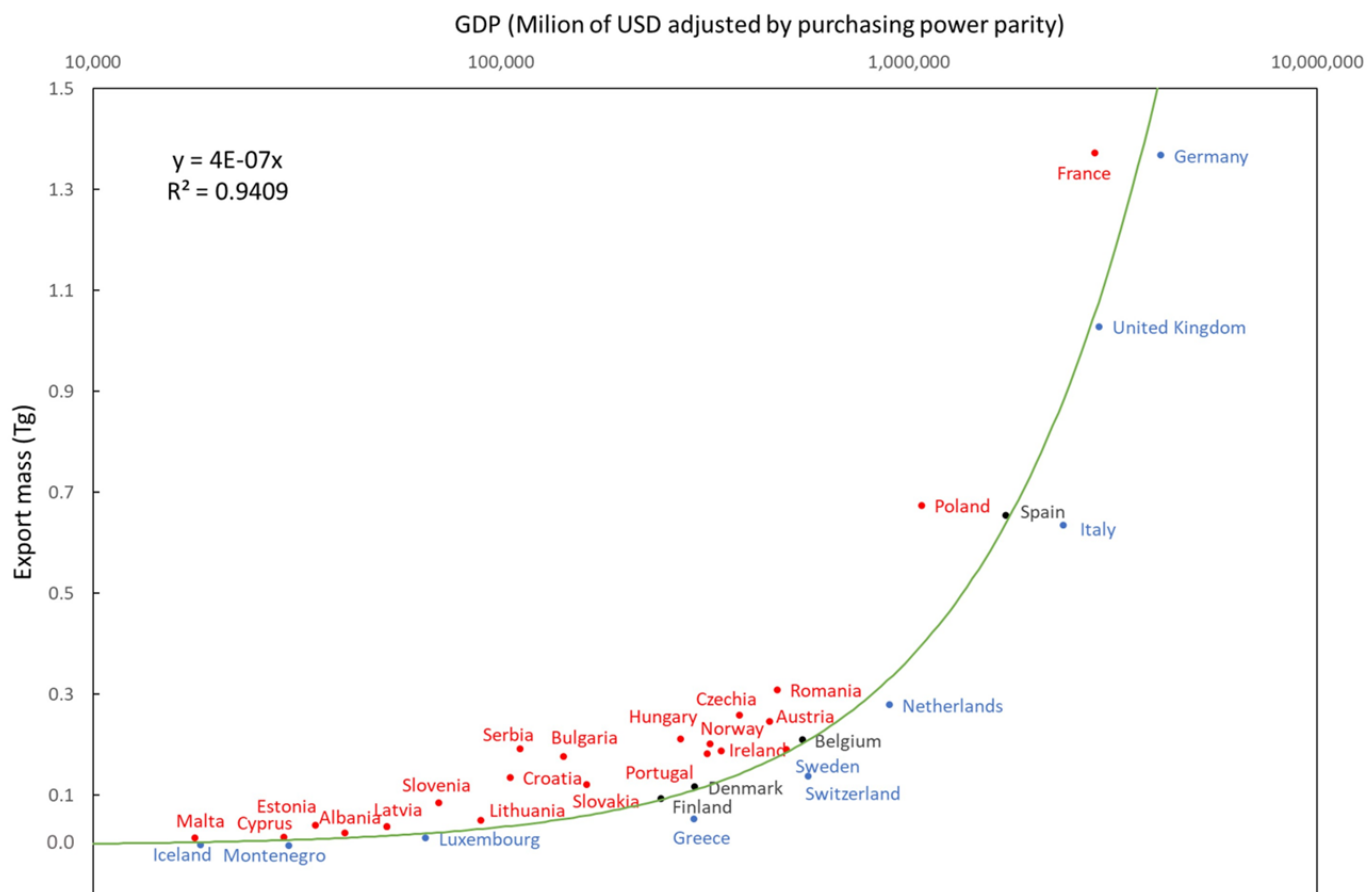
10 Supplementary Note 10: NO_x and NMVOC anthropogenic emissions by country

Supplementary Table S8. Annual NO_x and NMVOC anthropogenic emissions by country used in this study (mean of 2015, 2016, and 2017).

Country	NO _x emissions (t/year)	NMVOC emissions (t/year)
Germany (DE)	1.10×10^9	8.57×10^8
United Kingdom (UK)	8.17×10^8	7.08×10^8
France (FR)	8.15×10^8	6.09×10^8
Poland (PL)	6.90×10^8	5.76×10^8
Italy (IT)	6.16×10^8	7.92×10^8
Spain (ES)	6.09×10^8	5.45×10^8
Netherlands (NL)	2.23×10^8	1.83×10^8
Romania (RO)	2.08×10^8	2.11×10^8
Greece (EL)	2.04×10^8	1.43×10^8
Belgium (BE)	1.76×10^8	8.57×10^7
Czechia (CZ)	1.69×10^8	2.09×10^8
Portugal (PT)	1.44×10^8	1.58×10^8
Austria (AT)	1.40×10^8	9.47×10^7
Finland (FI)	1.17×10^8	7.29×10^7
Norway (NO)	1.16×10^8	1.45×10^8
Bulgaria (BG)	1.13×10^8	7.22×10^7
Sweden (SE)	1.09×10^8	1.26×10^8
Hungary (HU)	1.01×10^8	1.20×10^8
Serbia (RS)	9.54×10^7	8.60×10^7
Denmark (DK)	7.77×10^7	6.74×10^7
Ireland (IE)	7.21×10^7	6.81×10^7
Slovakia (SK)	6.22×10^7	8.94×10^7
Switzerland (CH)	5.85×10^7	6.15×10^7
Croatia (HR)	4.79×10^7	5.83×10^7
Lithuania (LT)	4.32×10^7	3.47×10^7
Slovenia (SI)	3.29×10^7	2.41×10^7
Latvia (LV)	3.25×10^7	3.29×10^7
Estonia (EE)	3.04×10^7	1.77×10^7
Albania (AL)	2.90×10^7	2.43×10^7
Cyprus (CY)	1.43×10^7	1.13×10^7
Luxembourg (LU)	1.21×10^7	8.02×10^6
Montenegro (ME)	7.22×10^6	8.13×10^6
Iceland (IS)	6.61×10^6	3.15×10^6
Malta (MT)	4.28×10^6	2.39×10^6

11 Supplementary Note 11: Example of potential applications of the results: Relationship between the exported mass of O₃ and the gross Domestic Product – Purchase power parity (GDP-PPP)

The results of the source apportionment study can be used for broader applications. In this section, we show an overview of the relationship between a country's wealth and its exported mass of O₃. In this case, we use each country's 2015, 2016, and 2017 mean Gross Domestic Product – Purchase power parity (GDP – PPP) as an economic indicator of wealth (The World Bank, <https://data.worldbank.org/>, last access: 15 of September 2023). Fig. S13 shows the relationship between the GDP - PPP and the export of O₃ of the countries, where the trend line has been forced to 0 export of O₃ and 0 GDP - PPP. Countries that deviate significantly from the trend line export either more or less O₃ than expected based on their GDP-PPP. Specifically, countries on the right of the trend line (blue color Fig. S13) are exporting less O₃ than the wealth they generate compared to other European countries. On the other hand, the countries at the left of the trend line (red color Fig. S13) have the opposite behavior, as they are less efficient in producing goods in terms of emissions of ozone and their precursors. According to these results, most Eastern European countries are exporting much O₃, considering the goods and services they produce, with countries such as Serbia, Romania, Czechia, and Poland standing out. In contrast, the main emitters of O₃ precursors, such as Italy and Germany, export less O₃ than other countries if the wealth they generate is considered (Fig. S13).



Supplementary Figure S13. Relationship between the exported mass of O₃ and the GDP-PPP of each country.

Supplementary References

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