Supplement to 'Are contributions of emissions to ozone a matter of scale?'

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S1 Definition of the tagging categories

Table S1: Description of the different tagging categories applied in this study.

tagging categories	description	notation for tagged ozone
land transport	emissions of road traffic, inland navigation, railways (IPCC codes 1A3b_c_e)	O_3^{tra}
anthropogenic non-traffic	sectors energy, solvents, waste, industries, residential, agriculture	O_3^{ind}
ship	emissions from ships (IPCC code 1A3d)	O_3^{shp}
aviation	emissions from aircraft	O_3^{air}
lightning	lightning NO_x emissions	O_3^{lig}
biogenic	on-line calculated isoprene and soil-NO_x emissions, off-line emissions from biogenic sources and agricultural waste burning (IPCC code 4F)	O_3^{soi}
biomass burning	biomass burning emissions	O_3^{bio}
CH_4	degradation of CH_4	O_3^{CH4}
N_2O	degradation of N_2O	O_3^{N2O}
stratosphere	downward transport from the stratosphere	O_3^{str}

S2 Supplementary information

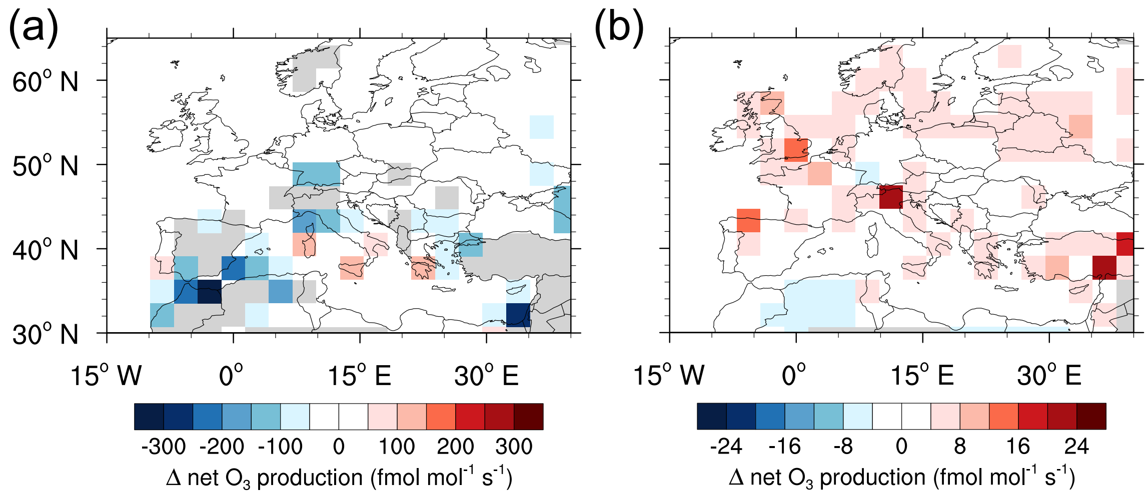

Figure S1: ΔP_{O3} calculated for JJA 2008–2010 at **(a)** 950 hPa and **(b)** 700 hPa (in fmol mol^{-1} s^{-1}). The CM50 data have been transformed on the horizontal and vertical grid of EMAC. Grey areas indicate missing values.

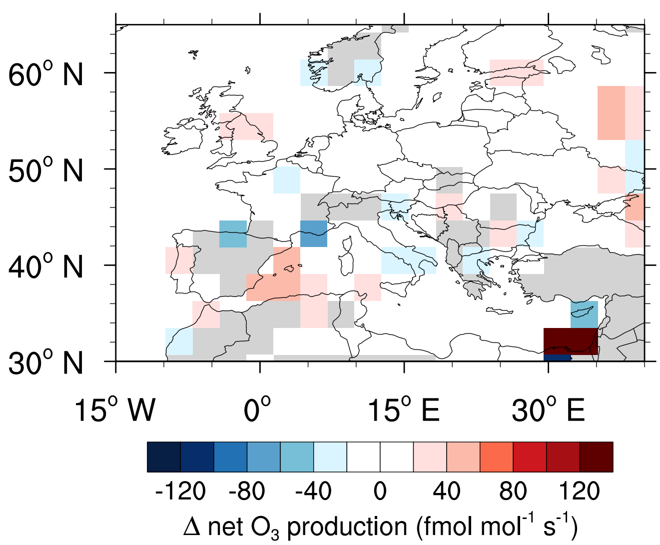

Figure S2: Δ $P_{O3}{}^{ET42}$ 'MINUS' Δ $P_{O3}{}^{REF}$ calculated for JJA 2008 at 950 hPa (in fmol mol^{-1} s^{-1}). The CM50 data have been transformed on the horizontal and vertical grid of EMAC. Grey areas indicate missing values.

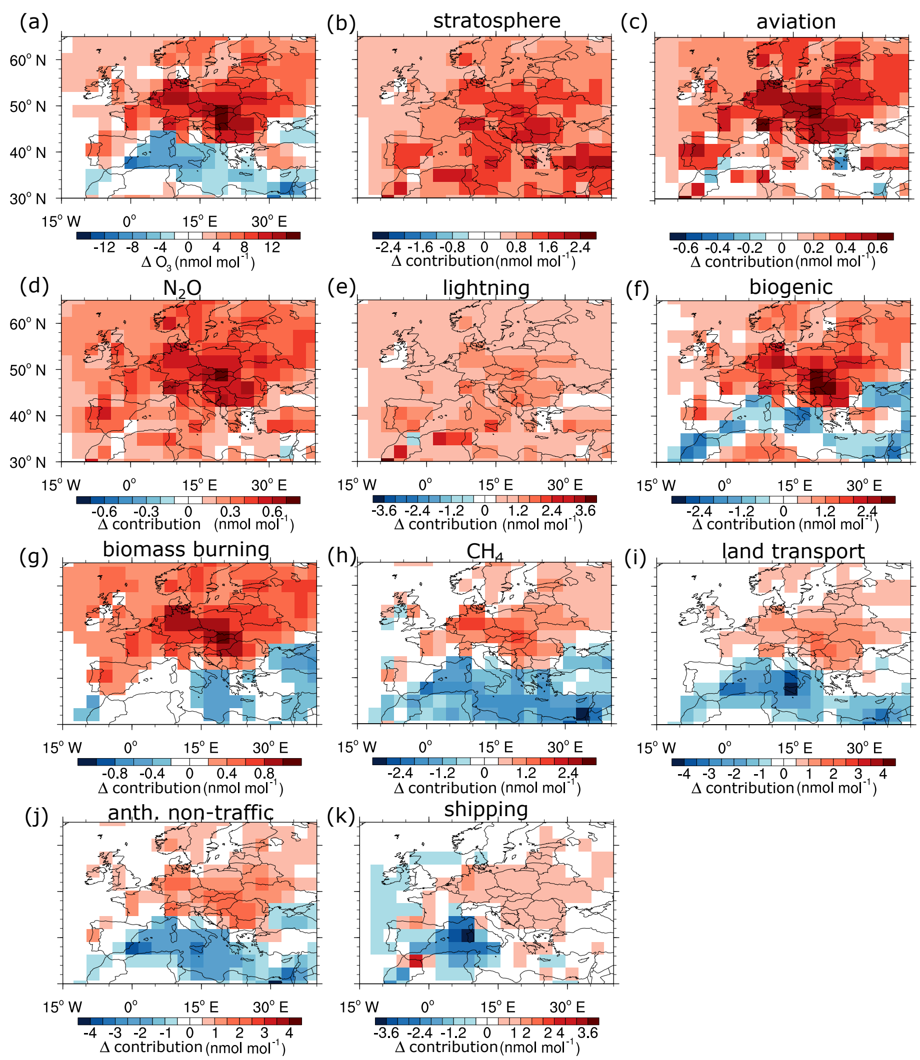

Figure S3: Differences ('CM50 MINUS EMAC') of the JJA averaged ground-level ozone values **(a)** and absolute contributions to groundlevel O_3 (in nmol mol^{-1}) of the categories: **(b)** stratosphere, **(c)** aviation, **(d)** N_2O, **(e)** lightning, **(f)** biogenic, **(g)** biomass burning degradation, **(h)** aviation, **(i)** land transport, **(j)** anthropogenic non-traffic, and **(k)** shipping. Shown are the results of the *REF* simulation, averaged for JJA 2008–2010. The CM50 data were transformed onto the horizontal grid of EMAC.

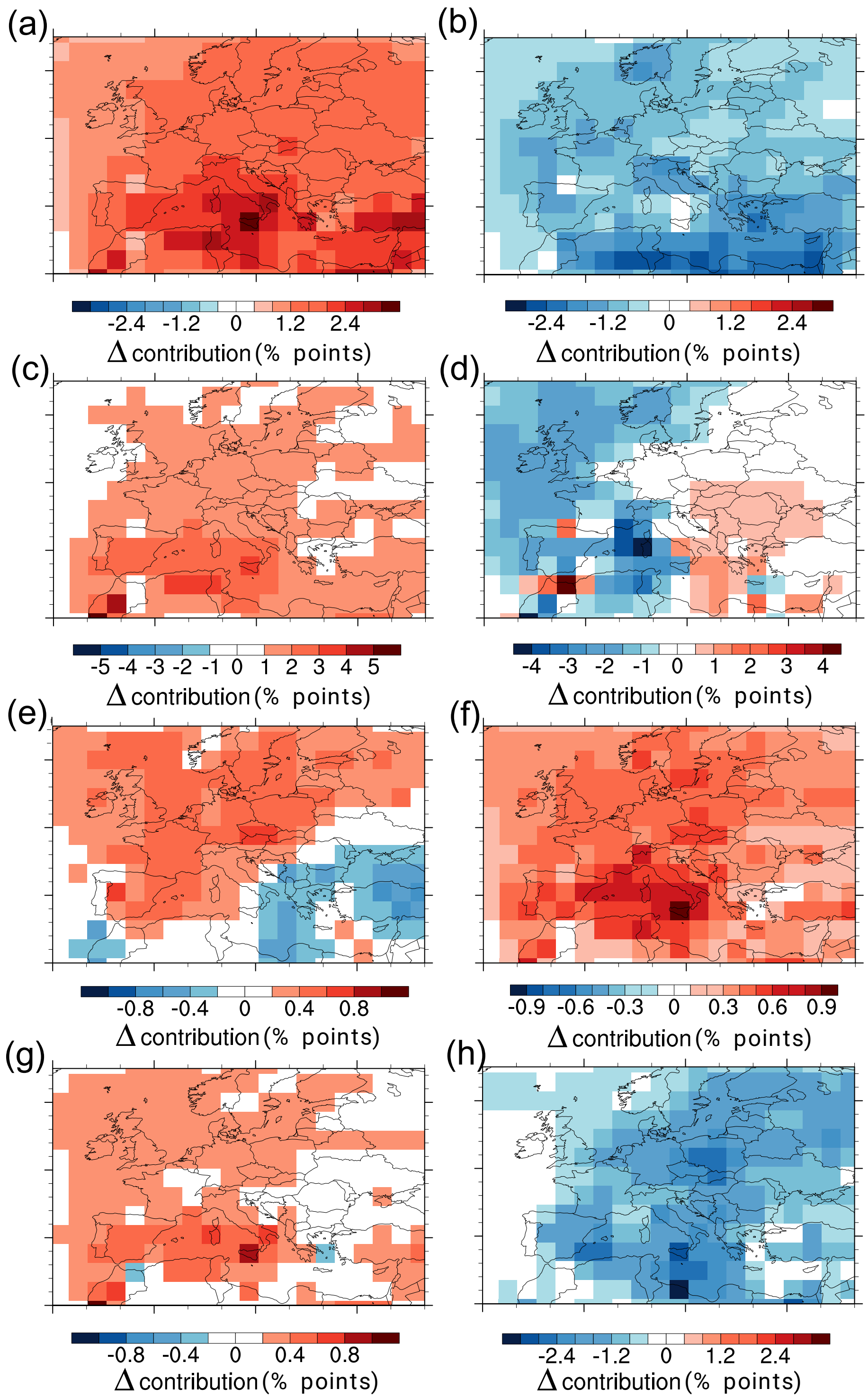

Figure S4: Differences ('CM50 MINUS EMAC') of the JJA averaged relative contributions to groundlevel O_3 (in percentage points) of the categories: **(a)** stratosphere, **(b)** methane degradation, **(c)** lightning, **(d)** shipping, **(e)** biomass burning, **(f)** N_2O degradation, **(g)** aviation and **(h)**anthropogenic non-traffic. Shown are the results of the *REF* simulation, averaged for JJA 2008–2010. The CM50 data were transformed onto the horizontal grid of EMAC.

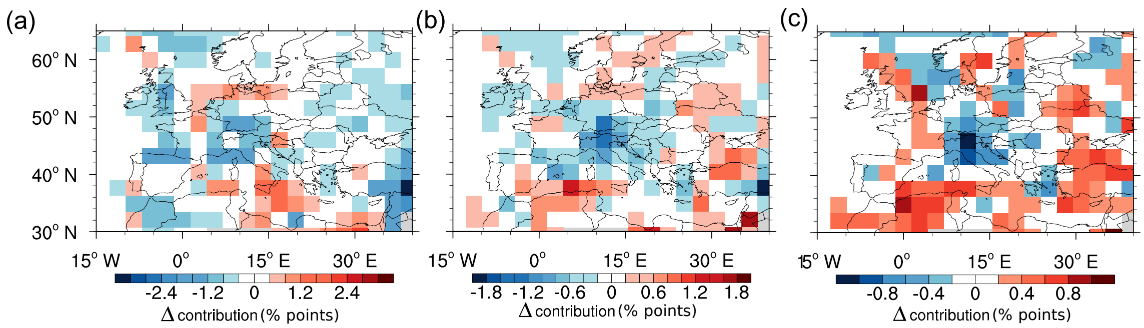

Figure S5: Difference ('CM50 MINUS EMAC') of the relative contribution of O_3^{tra} to ozone at **(a)** 800 hPa,**(b)** 700 hPa and **(c)** 600 hPa (in percentage points). The CM50 results are transformed onto the EMAC grid. Shown are the results of the *REF* simulation, averaged for JJA 2008–2010.

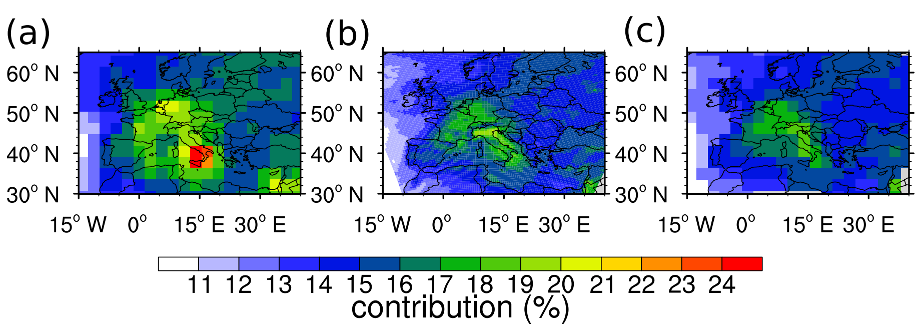

Figure S6: 95th percentile of the contribution of O_3^{tra} to ground-level O_3 (for JJA between 9–18 UTC) for **(a)** EMAC, **(b)** CM50 and **(c)** CM50 transformed onto the EMAC grid ($CM50_E$).

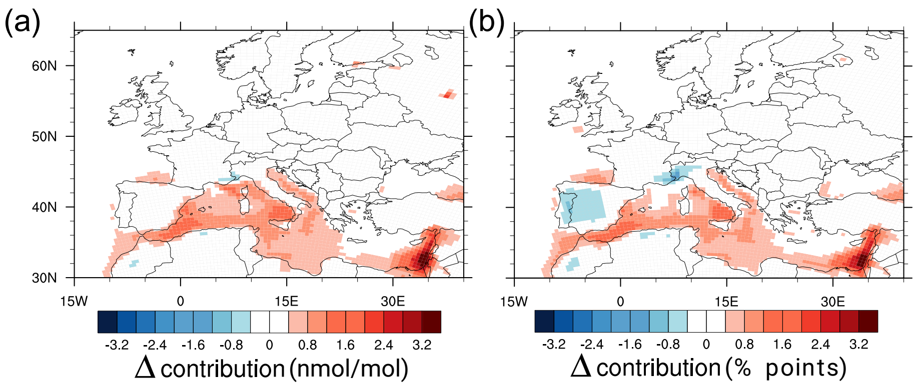

Figure S7: Difference ('*ET42* MINUS *REF*') of the absolute ((**a**), in nmol mol^{-1} and relative (**b**) in percentage points) contribution of O_3^{tra} to ground-level ozone. Shown are the results of CM50 for JJA 2008.

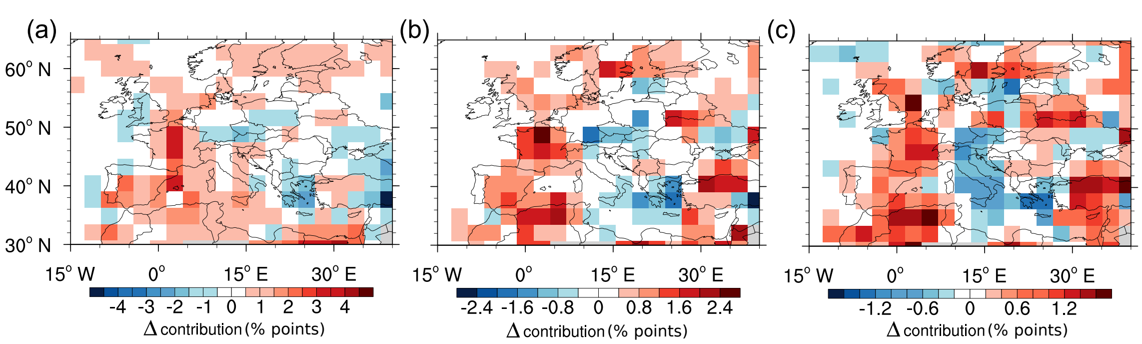

Figure S8: Difference ('CM50 MINUS EMAC') of the relative contribution of O_3^{soi} to ozone at (**a**) 800 hPa,(**b**) 700 hPa, and (**b**) 600 hPa, (in percentage points). The CM50 results are transformed onto the EMAC grid. Shown are the results of the *REF* simulation, averaged for JJA 2008–2010.

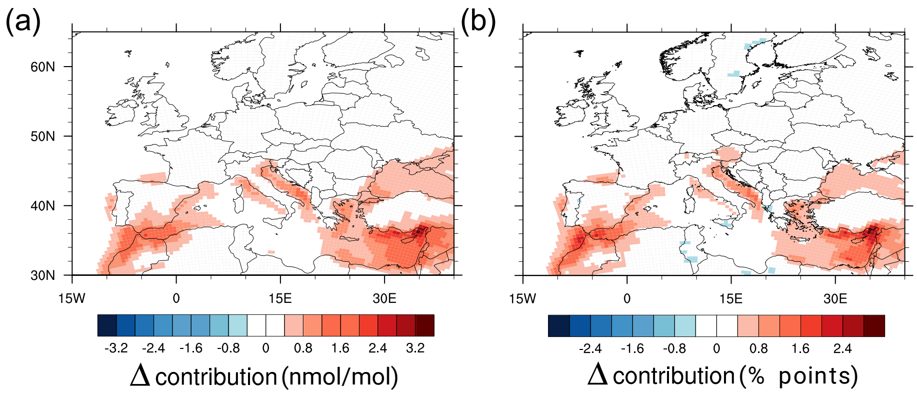

Figure S9: Difference ('*EBIO* MINUS *REF*') of the absolute ((**a**), in nmol mol^{-1} and relative (**b**) in percentage points) contribution of O_3^{soi} to ground-level ozone. Shown are the results of CM50 for JJA 2008.

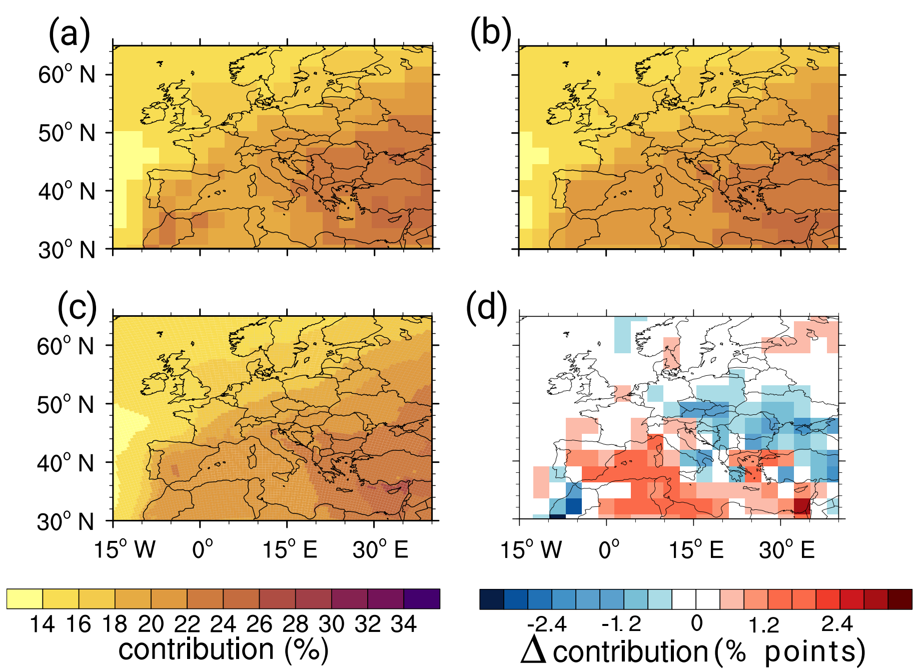

Figure S10: Comparison of the JJA averaged ground-level contribution of O_3^{soi} to O_3 (in %) of EMAC and CM50: (**a**) results of EMAC, (**b**) results of CM50 transformed onto the EMAC grid, (**c**) results of CM50 on the original grid and (**d**) difference ('CM50 MINUS EMAC' in percentage points) on the coarse grid. (**a**)– (**c**) use the same (left) colour bar. Shown are the results of the *EBIO* simulation for 2008.

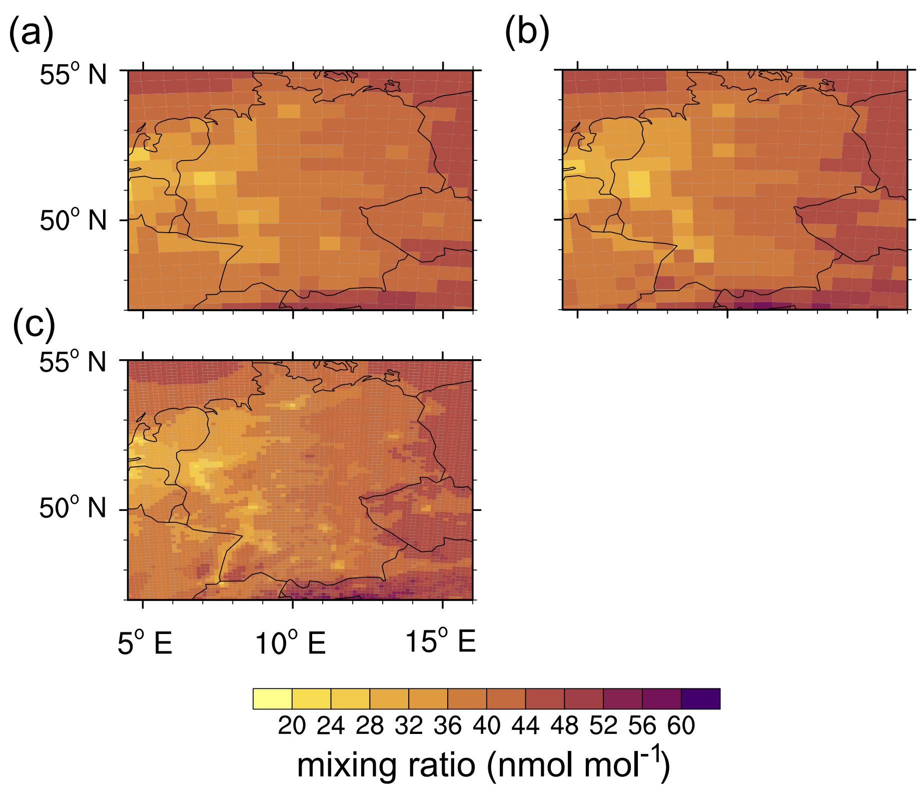

Figure S11: Ground-level ozone mixing ratios averaged for JJA 2008. (a) The values of CM50, (b) the values of CM12 transformed onto the CM50 grind and (c) the CM12 values on the original grid. Results are from the *EVEU* simulation.

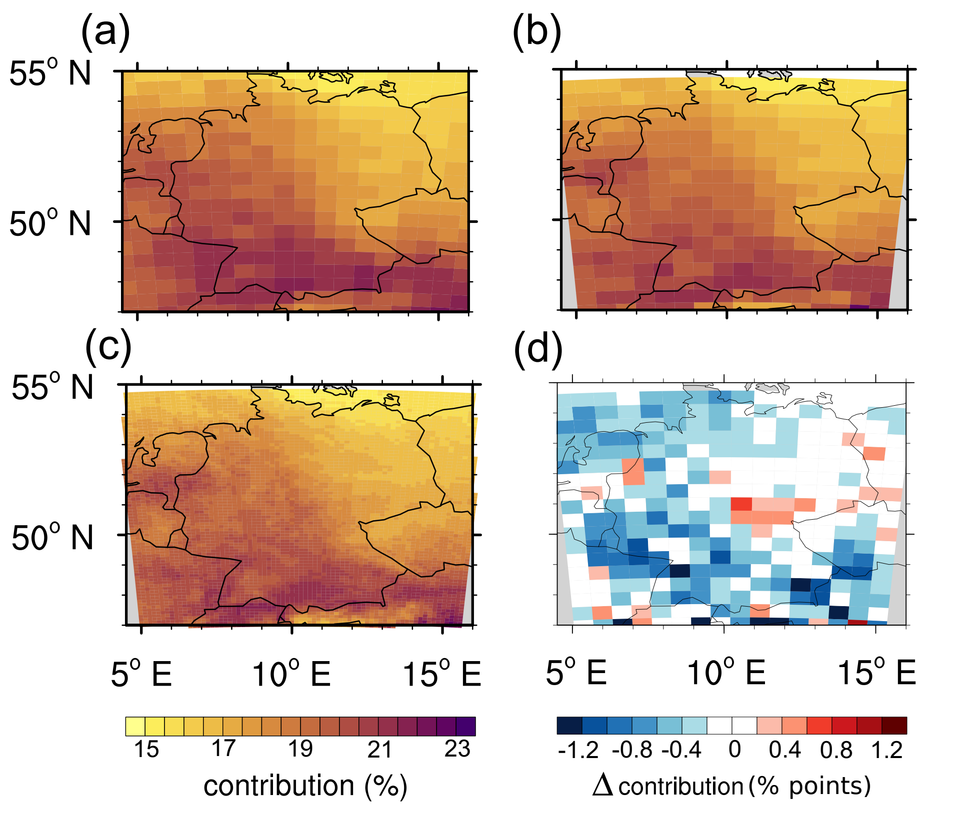

Figure S12: 95th percentile of the relative contribution of O_3^{tra} to ground-level O_3 (for JJA between 9–18 UTC) for **(a)** CM50, **(b)** CM12 transformed onto the CM50 grid, **(c)** CM50, and **(d)** the difference between CM50 and CM12 ('CM12 MINUS CM50')).

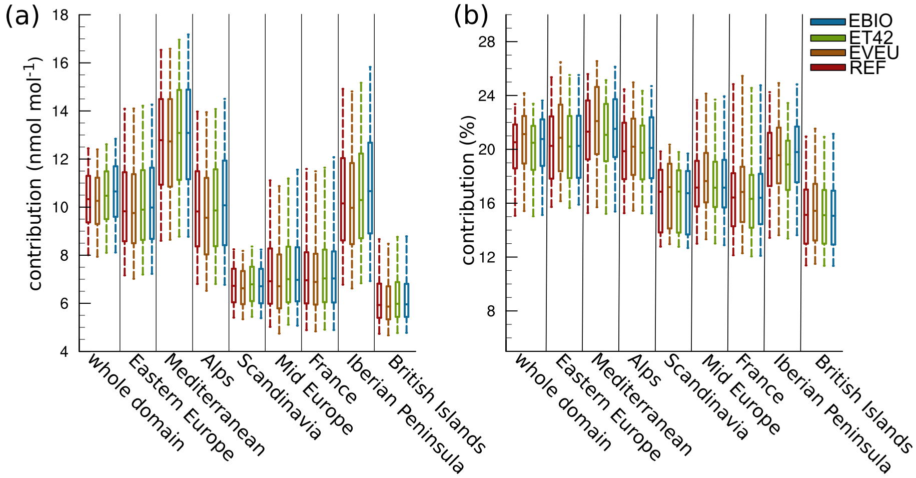

Figure S13: Comparison of the contributions of O_3^{soi} to ground-level ozone for JJA 2008 between the four simulations. (a) displays the absolute contribution in nmol mol^{-1} and (b) the relative contribution to ground-level ozone (in %). All values are area averaged over the respective region and are calculated using the results of the CM50 instance. The lower and upper end of the box indicate the 25th and 75th percentile, respectively, the bar the median, and the whiskers the 5th and 95th percentile of the timeseries for the JJA values from 2008 based on 3-hourly model output.

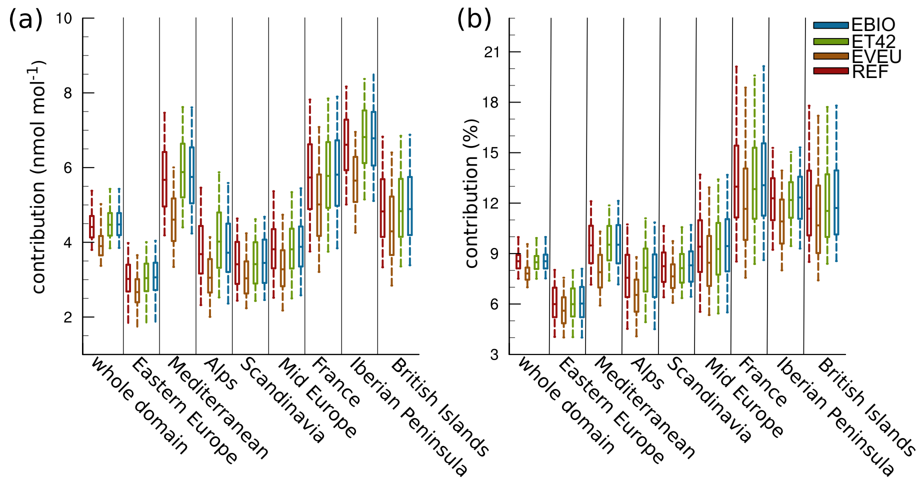

Figure S14: Comparison of the contributions of O_3^{shp} to ground-level ozone for JJA 2008 between the four simulations. (a) displays the absolute contribution in nmol mol^{-1} and (b) the relative contribution to ground-level ozone (in %). All values are area averaged over the respective region and are calculated using the results of the CM50 instance. The lower and upper end of the box indicate the 25th and 75th percentile, respectively, the bar the median, and the whiskers the 5th and 95th percentile of the timeseries for the JJA values from 2008 based on 3-hourly model output.

Table S2: Root-mean-square error (RMSE, in µg m^{-3} and normalized mean-bias error (MBE, in %) of O_3 for CM50 and CM12 in comparison to ground-level observations. Shown are the averaged values for June to August 2008 for all stations located in the CM12 domain. The values are calculated from monthly mean values. The model values are height corrected as discussed in detail by Mertens et al. (2016).

	RMSE (in µg m^{-3})		MB (in %)	
	CM50	CM12	CM50	CM12
REF	15.68	15.43	10.89	13.98
EVEU	14.96	14.03	6.82	11.15

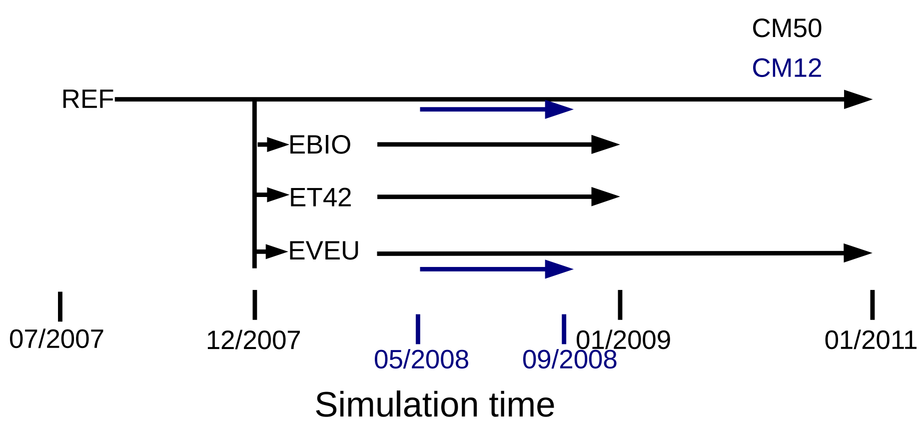

Figure S15: Sketch visualising the simulation period of the individual simulations. All simulations were branched off from the *REF* simulations. The black arrows indicate the simulation period of the CM50 instance, blue indicates the periods were additionally also the CM12 model instance was activated.

S3 Total emissions of the different simulations

The following Tables summaries the emissions of NO_x, CO and VOCs from all anthropogenic and natural sources applied in the different simulations. Please note, that for every specie only these emission sectors are given where emissions occur. These sector are:

- **road t.**: Road traffic emissions
- **anth. nt**: Anthropogenic non-traffic emissions
- **shipping**: Shipping emissions
- **aviation**: Aviation emissions
- **Soil-**NO_x: NO_x emissions from soils calculated by the model
- **LNOX**: Lightning NO_x emissions
- **AWB**: Agricultural waste burning emissions
- **BB** : Biomass burning emissions
- **biog.**: Further biogenic emissions, not calculated by the model but prescribed as annual climatology
- **biog.** C_5H_8: Biogenic isoprene emissions, calculated by the model

Tables S3–S5 give the total emissions of EMAC of NO_x, CO and VOC, respectively. Tables S6–S8 give the total emissions of CM50 and Table S9–S11 of CM12. Please note, that the emissions of C_5H_8 from biogenic origin listed in the tables are already scaled with factors of 0.6 (EMAC and COSMO-CLM/MESSy for the *EBIO* simulation) and 0.45 (COSMO-CLM/MESSy).

Table S3: Total emissions of NO_x (in Tg a^{-1} in amount of NO) for EMAC. Given are the annual totals for the year 2008.

Simulation	road t.	anth. nt	shipping	aviation	Soil NO_x	LNOX	AWB	BB	Sum
REF	20.4	36.9	12.7	2.14	12.5	12.1	0.416	9.47	97.2
ET42	20.4	36.9	12.7	2.14	12.5	12.1	0.416	9.47	97.2
EBIO	20.4	36.9	12.7	2.14	12.5	12.1	0.416	9.47	97.2
EVEU	20.4	36.9	12.7	2.14	12.5	12.1	0.416	9.47	97.2

Table S4: Total emissions of CO (in (in Tg a^{-1}) for EMAC. Given are the annual totals for the year 2008.

Simulation	road t.	anth. nt	shipping	AWB	biog.	BB	Sum
REF	153	412	1.33	20.2	113	328	1030
ET42	153	412	1.33	20.2	113	328	1030
EBIO	153	412	1.33	20.2	113	328	1030
EVEU	153	412	1.33	20.2	113	328	1030

Table S5: Total emissions of VOC (in Tg a^{-1} in amount of C) for EMAC. Given are the annual totals for the year 2008.

Simulation	road t.	anth. nt	shipping	biog. C_5H_8	AWB	biog.	BB	Sum
REF	17.5	73.4	2.19	270	0.943	108	14.5	667
ET42	17.5	73.4	2.19	270	0.943	108	14.5	667
EBIO	17.5	73.4	2.19	270	0.943	108	14.5	667
EVEU	17.5	73.4	2.19	270	0.943	108	14.5	667

Table S6: Total emissions of NO_x (in Tg a^{-1} in amount of NO) for CM50. Given are the annual totals for the year 2008.

Simulation	road t.	anth. nt	shipping	aviation	Soil NO_x	LNOX	AWB	BB	Sum
REF	5.19	7.6	2.35	0.595	1.57	0.715	0.0762	0.28	18.4
ET42	5.20	7.65	2.35	0.591	1.57	0.715	0.0762	0.276	18.4
EBIO	5.20	7.65	2.35	0.591	1.66	0.715	0.0762	0.276	18.5
EVEU	5.35	5.08	1.81	0.534	1.57	0.715	0.0762	0.28	15.4

Table S7: Total emissions of CO (in Tg a^{-1}) for CM50. Given are the annual totals for the year 2008.

Simulation	road t.	anth. nt	shipping	AWB	biog.	BB	Sum
REF	30.9	28.7	0.241	2.89	4.84	8.87	76.4
ET42	30.9	28.9	0.246	2.88	4.84	9.03	76.8
EBIO	30.9	28.9	0.246	2.88	4.84	9.03	76.8
EVEU	23.6	3.1	0.299	2.89	4.84	8.87	43.6

Table S8: Total emissions of VOC (in Tg a^{-1} in amount of C) for CM50. Given are the annual totals for the year 2008.

Simulation	road t.	anth. nt	shipping	biog. C_5H_8	AWB	biog.	BB	Sum
REF	3.29	14.2	0.343	14.3	0.0994	4.59	0.377	54.7
ET42	3.31	14.4	0.351	14.3	0.0994	4.59	0.377	54.9
EBIO	3.29	14.4	0.351	20.3	0.0994	4.59	0.377	56.7
EVEU	3.39	6.56	0.0954	14.3	0.0994	4.59	0.377	46.9

Table S9: Total emissions of NO_x (in Tg in amount of NO) for CM12. Given are the annual totals for the May–August 2008.

Simulation	road t.	anth. nt	shipping	aviation	Soil NO_x	LNOX	AWB	BB	Sum
REF	0.342	0.335	0.0333	0.0642	0.167	0.0456	0.000761	0.00361	0.988
EVEU	0.441	0.277	0.0263	0.0555	0.167	0.0456	0.000761	0.00361	1.01

Table S10: Total emissions of CO (in Tg) for CM12. Given are the annual totals for the May–August 2008.

Simulation	road t.	anth. nt	shipping	AWB	biog.	BB	Sum
REF	1.27	1.41	0.00348	0.0333	4.81	0.12	7.65
EVEU	1.18	0.842	0.000719	0.0333	4.81	0.12	6.98

Table S11: Total emissions of VOC (in Tg in amount of C) for CM12. Given are the annual totals for the May–August 2008.

Simulation	road t.	anth. nt	shipping	biog. C_5H_8	AWB	biog.	BB	Sum
REF	0.141	0.538	0.00413	0.190	0.0111	0.372	0.00516	1.49
EVEU	0.143	0.383	0.00175	0.190	0.0111	0.372	0.00516	1.34

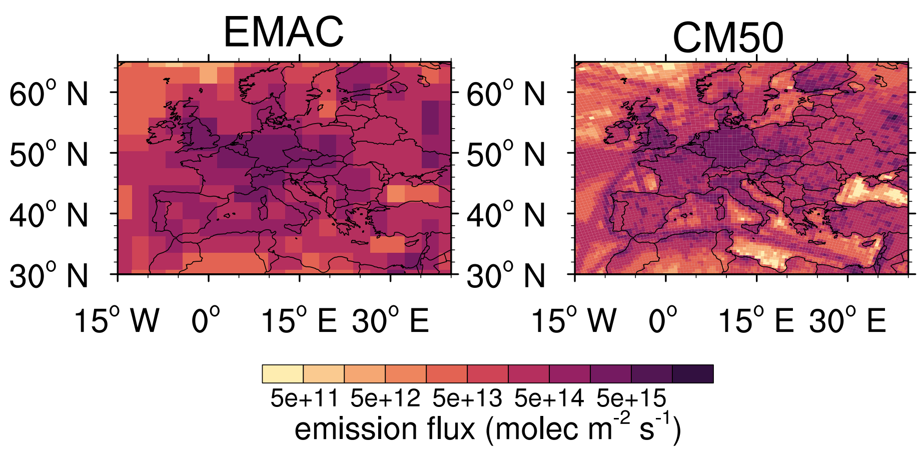

Figure S16: Annual averaged emissions flux (in molec m^{-2} s^{-1}) of NOx due to all anthropogenic emission sources (land transport, anthropogenic non-traffic, shipping; *REF* simulation) for EMAC and CM50.

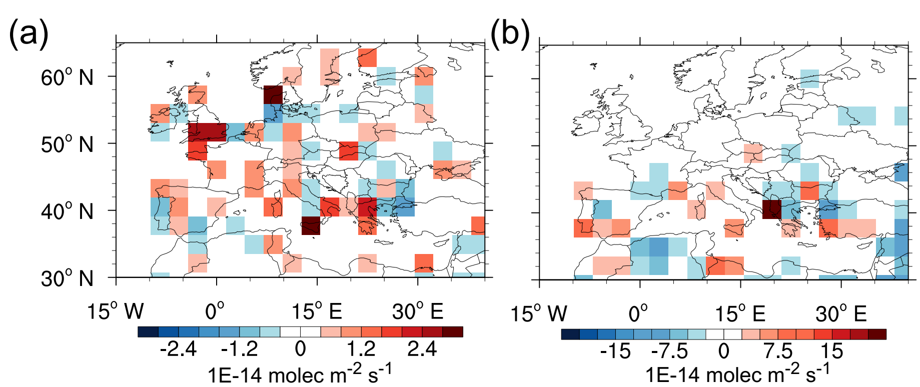

Figure S17: Difference ('EMAC MINUS CM50') of the on-line calculated **(a)** soil-NO_x and **(b)** isoprene emission flux (in 1E-14 molec $m^{-2}s^{-1}$). The CM50 results are transformed onto the EMAC grid. Shown are the results of the *REF* simulation, averaged for JJA 2008–2010. Please note that the different scaling factors of the isoprene emissions in EMAC and CM50 are not considered.

S4 List of used stations

Table S12: Overview of stations from the EMEP network used for the comparison.

Name	lat (in °)	lon (in °)	height (in m)	variable
Aliartos	38.37	23.08	110	O3
Ayia Marina	35.04	33.06	532	O3
Barcorotta	38.48	-6.92	393	O3,
Birkesens	58.38	8.25	190	O3
Eupen	50.63	6.00	295	O3
Forsthof	48.11	15.92	581	O3
Giordon Lighthouse	36.07	14.22	167	O3
Graz Platte	47.11	15.47	651	O3
Iskraba	45.57	14.87	520	O3
Ispra	45.80	8.63	209	O3
Jarczew	51.82	21.98	180	O3
Jungfraujoch	46.54	7.98	3578	O3
La Coulande	48.63	-0.45	304	O3
Ladybower Research	53.40	-1.75	420	O3
Lahemaa	59.50	25.90	32	O3
Lazaropole	41.54	20.69	1332	O3
Le Casset	45.00	6.47	1790	O3
Lullington Heathrow	50.79	0.18	120	O3
Mace Head	53.17	-9.50	15	O3
Montellipreti	42.10	12.63	48	O3
Narberth	51.78	-4.69	160	NO2
Neuglobsow	53.17	13.03	62	O3
Niembro	43.44	-4.85	134	O3
Noraa-Kvill	57.82	15.57	261	O3
Pic du Midi	42.94	0.14	2887	O3
Preila	55.35	21.06	5	O3
Puy de Dome	45.77	2.95	1468	O3
Revin	49.90	4.63	390	O3
Rojen Peak	41.70	24.74	1750	O3
Rucava	56.16	21.17	18	O3
Schmücke	50.65	10.77	937	O3
Starina	49.05	22.27	345	O3
Sulzberg	47.53	9.93	1020	O3
Ulborg	56.28	8.43	10	O3
Vilsandi	58.38	21.81	6	O3
Vizna	37.23	-3.53	1296	O3
Westerland	54.93	8.31	12	O3
Zingst	54.43	12.73	1	O3
Zoesni	57.14	25.91	188	O3

Table S13: Overview of stations providing vertical ozone profiles used for comparison. The last column shows for which variables the station data were used.

Name	lat (in °)	lon (in °)	height (in m)	variable
De Bilt	52.10	5.18 E	2	O3
Hohenpeisenberg	47.80	11.01	985	O3
Leginowo	52.40	29.97	96	O3
Lindenberg	52.22	14.11	112	O3
Madrid	40.45	3.72	680	O3
Payerne	46.81	6.94	490	O3
Uccle	50.80	4.36	100	O3
Valentia	51.94	10.25	14	O3

References

M. Mertens, A. Kerkweg, P. Jöckel, H. Tost, and C. Hofmann. The 1-way on-line coupled model system meco(n) – part 4: Chemical evaluation (based on messy v2.52). *Geosci. Model Dev.*, 9 (10):3545–3567, 2016. doi: 10.5194/gmd-9-3545-2016. URL `http://www.geosci-model-dev.net/9/3545/2016/`.