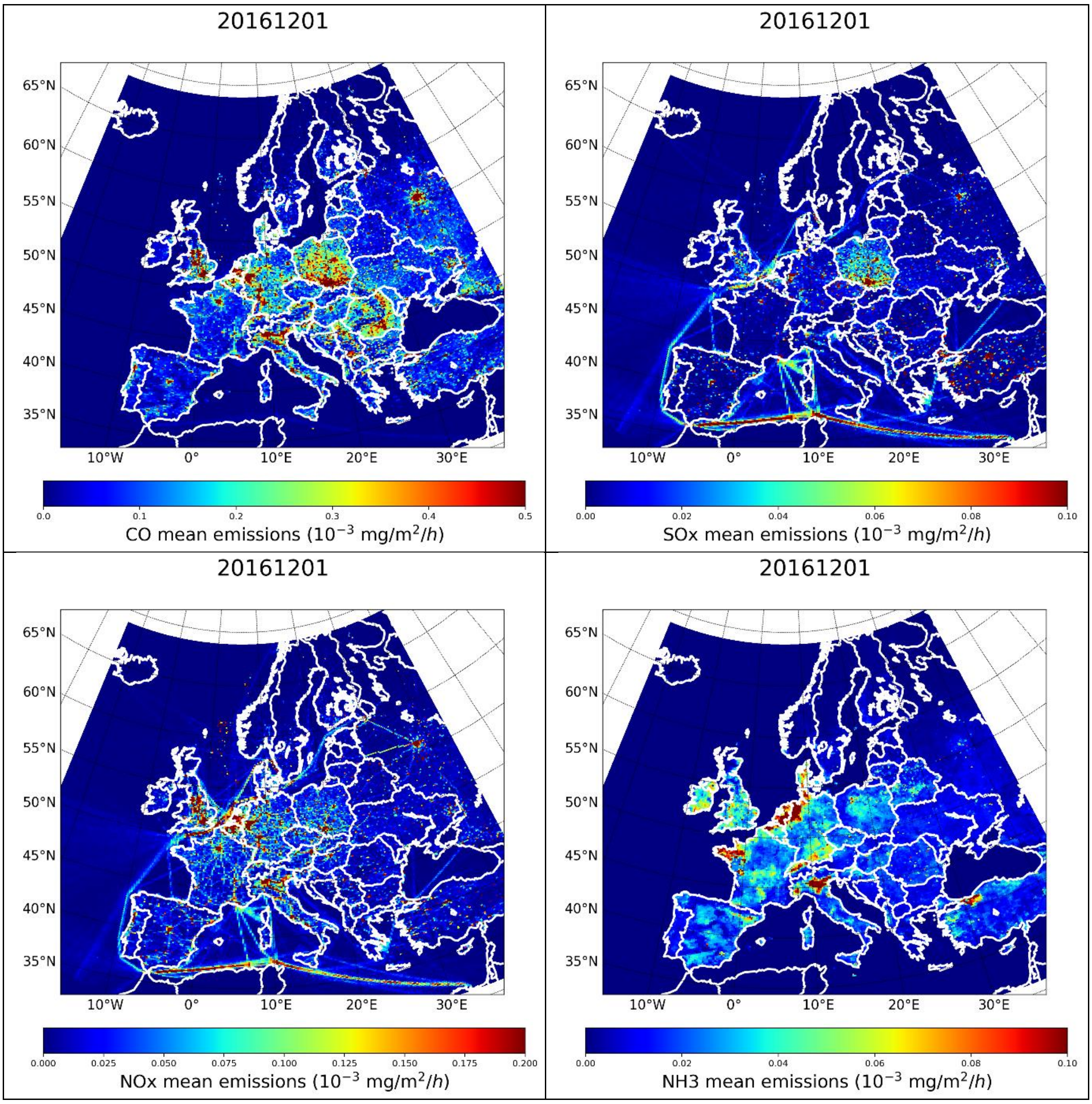
Supplement.


20161201
CO mean emissions (10^{-3} mg/m²/h)
20161201
SOx mean emissions (10^{-3} mg/m²/h)
20161201
NOx mean emissions (10^{-3} mg/m²/h)
20161201
NH3 mean emissions (10^{-3} mg/m²/h)

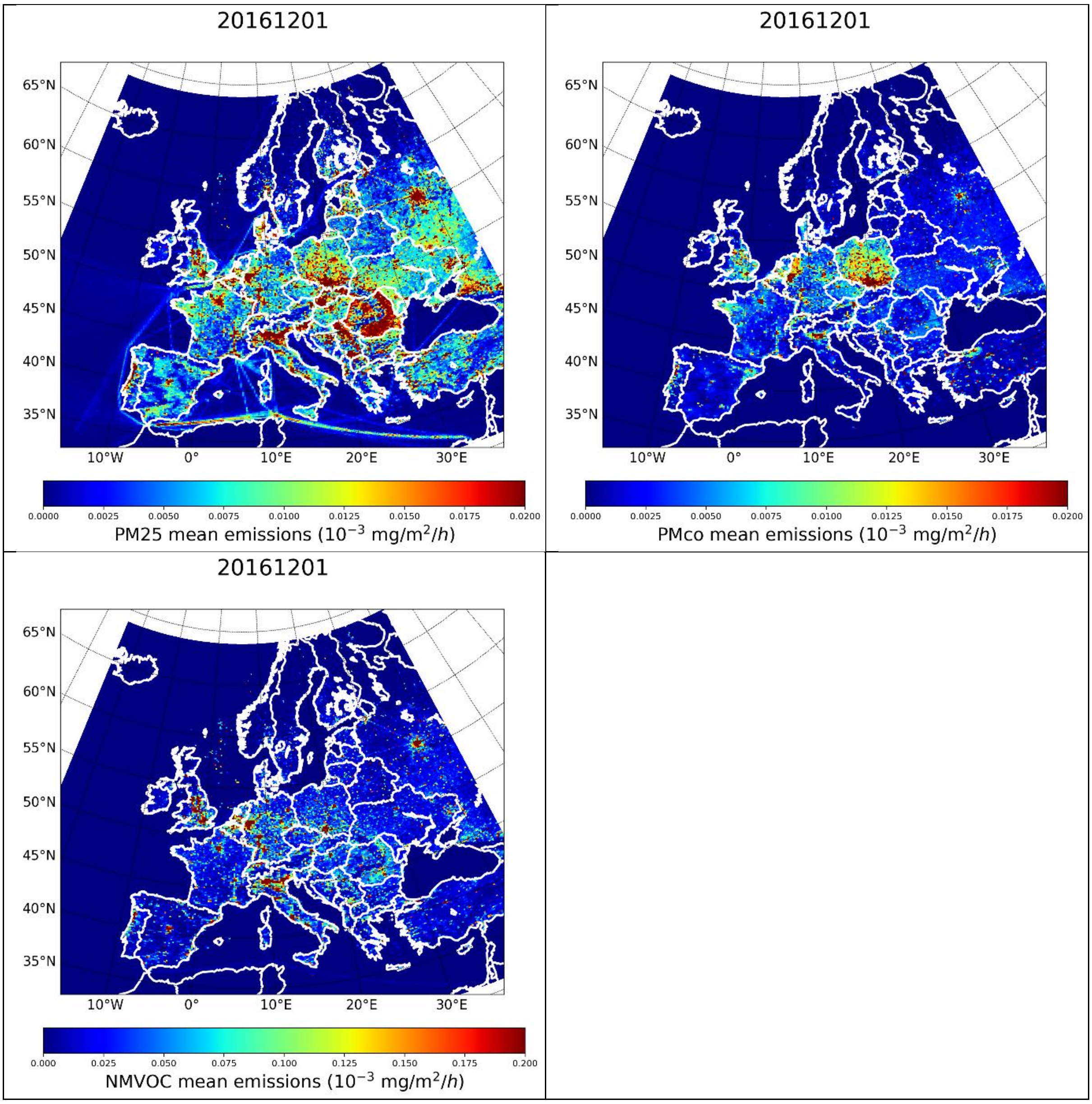
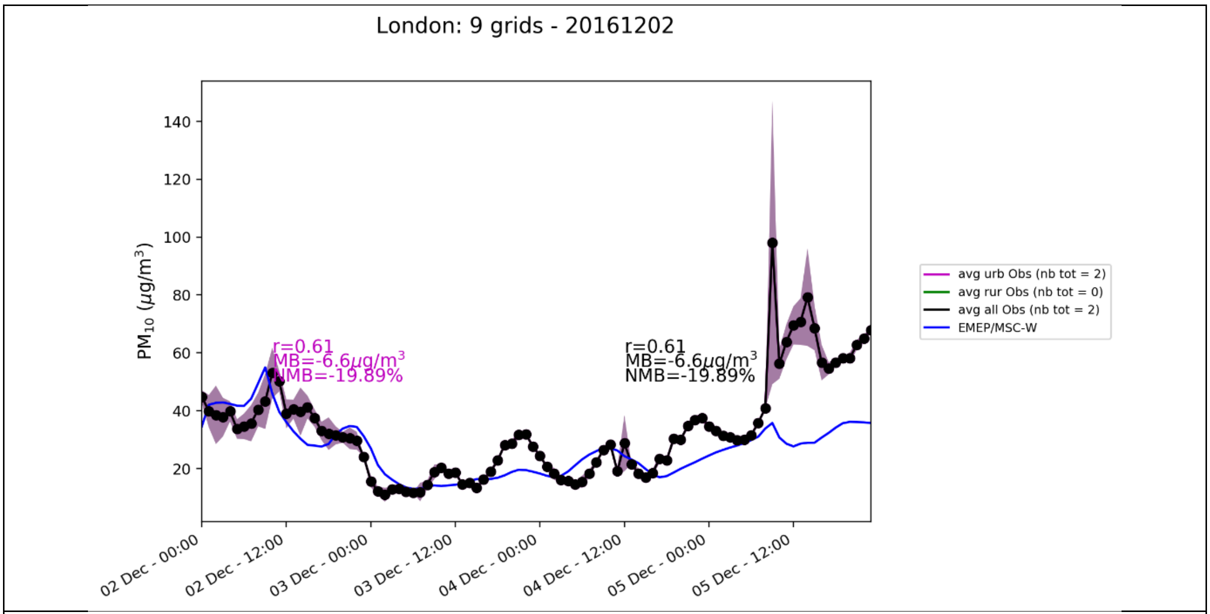
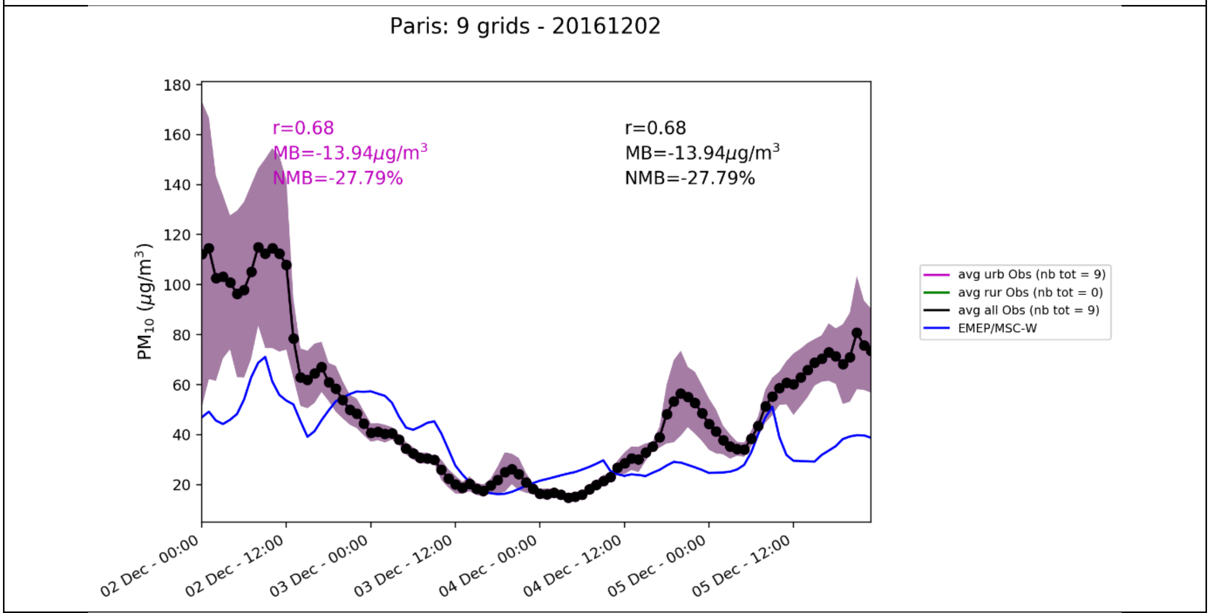
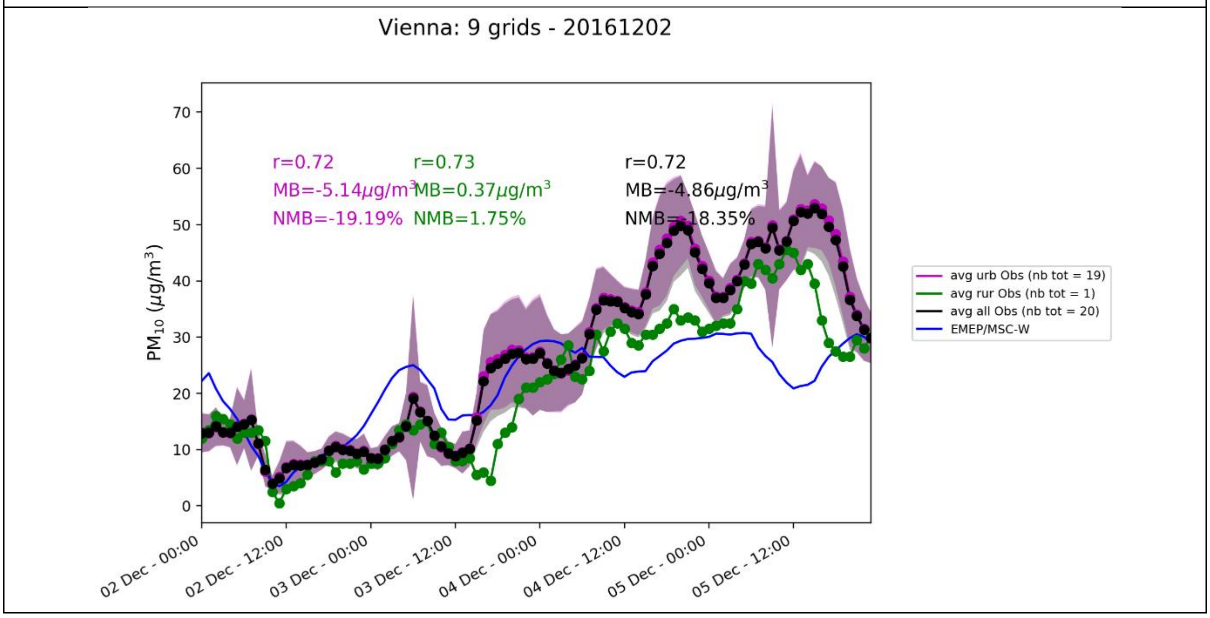
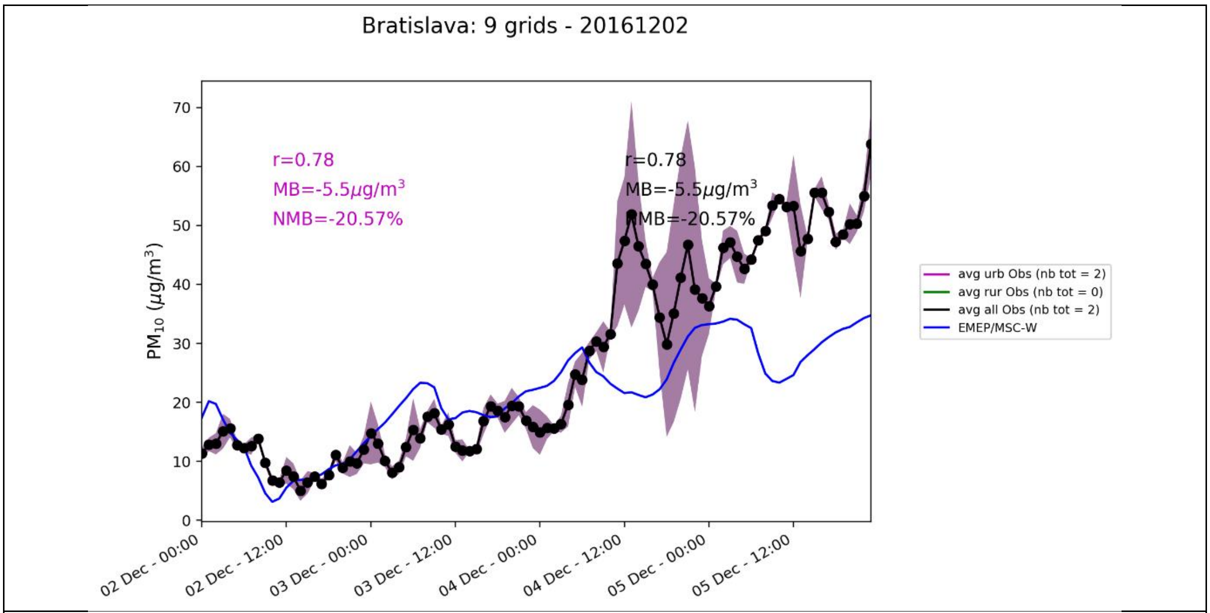


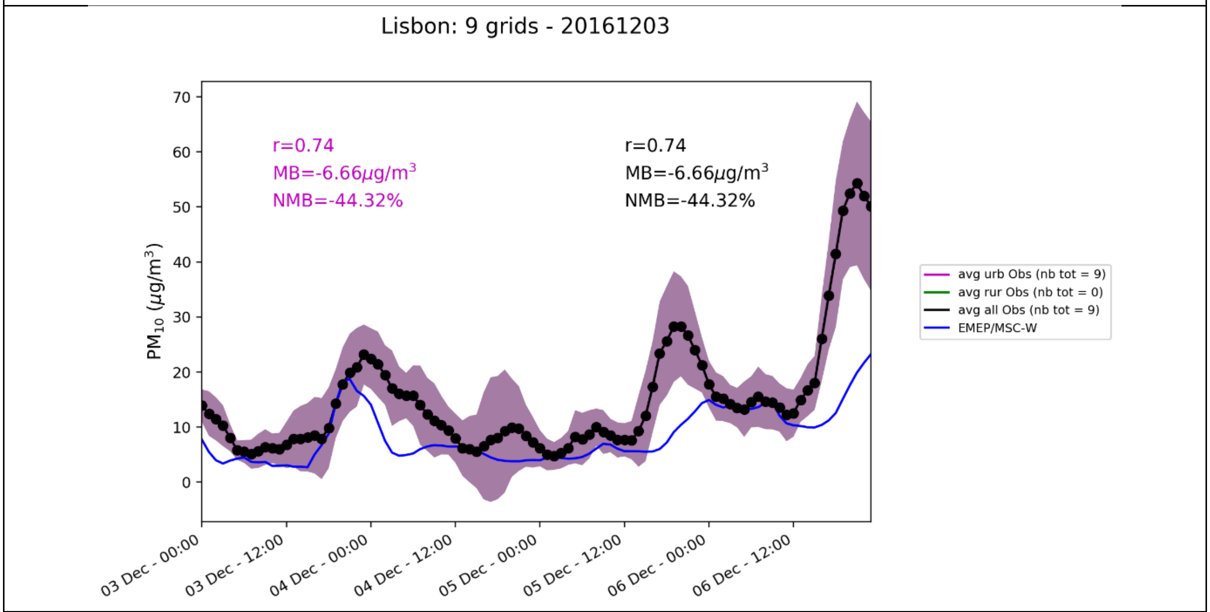
Fig. S1. Daily mean distribution of the anthropogenic emissions of CO, SO_x, NO_x, NH_3, fine primary particulate matter (denoted as $PM_{2.5}$), coarse primary particulate matter (denoted as PMco) and NMVOC as used in the EMEP model for the reference run on 01 December 2016.

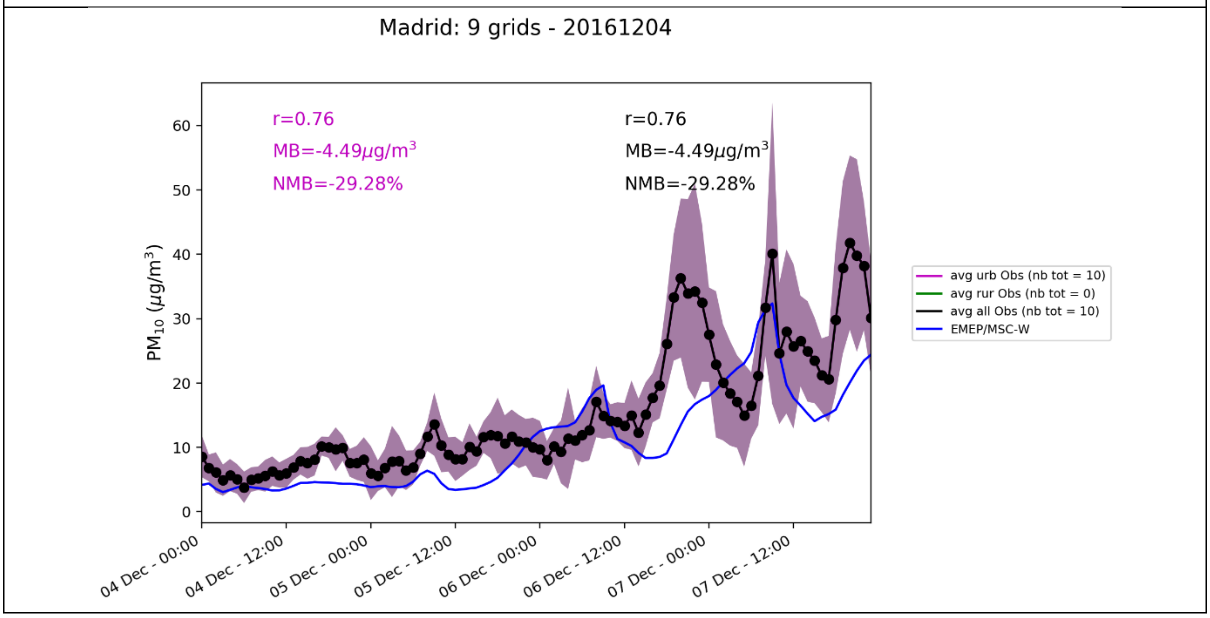

London: 9 grids - 20161202
PM10 (μg/m3)
r=0.61
MB=-6.6μg/m3
NMB=-19.89%
r=0.61
MB=-6.6μg/m3
NMB=-19.89%
avg urb Obs (nb tot = 2)
avg rur Obs (nb tot = 0)
avg all Obs (nb tot = 2)
EMEP/MSC-W
02 Dec - 00:00
02 Dec - 12:00
03 Dec - 00:00
03 Dec - 12:00
04 Dec - 00:00
04 Dec - 12:00
05 Dec - 00:00
05 Dec - 12:00


Paris: 9 grids - 20161202
PM10 (μg/m3)
r=0.68
MB=-13.94μg/m3
NMB=-27.79%
r=0.68
MB=-13.94μg/m3
NMB=-27.79%
avg urb Obs (nb tot = 9)
avg rur Obs (nb tot = 0)
avg all Obs (nb tot = 9)
EMEP/MSC-W
02 Dec - 00:00
02 Dec - 12:00
03 Dec - 00:00
03 Dec - 12:00
04 Dec - 00:00
04 Dec - 12:00
05 Dec - 00:00
05 Dec - 12:00


Vienna: 9 grids - 20161202
PM10 (μg/m3)
r=0.72
MB=-5.14μg/m3
NMB=-19.19%
r=0.73
MB=0.37μg/m3
NMB=1.75%
r=0.72
MB=-4.86μg/m3
NMB=-18.35%
avg urb Obs (nb tot = 19)
avg rur Obs (nb tot = 1)
avg all Obs (nb tot = 20)
EMEP/MSC-W
02 Dec - 00:00
02 Dec - 12:00
03 Dec - 00:00
03 Dec - 12:00
04 Dec - 00:00
04 Dec - 12:00
05 Dec - 00:00
05 Dec - 12:00


Bratislava: 9 grids - 20161202
r=0.78
MB=-5.5μg/m3
NMB=-20.57%
r=0.78
MB=-5.5μg/m3
NMB=-20.57%
PM10 (μg/m3)
avg urb Obs (nb tot = 2)
avg rur Obs (nb tot = 0)
avg all Obs (nb tot = 2)
EMEP/MSC-W
02 Dec - 00:00
02 Dec - 12:00
03 Dec - 00:00
03 Dec - 12:00
04 Dec - 00:00
04 Dec - 12:00
05 Dec - 00:00
05 Dec - 12:00


Lisbon: 9 grids - 20161203
r=0.74
MB=-6.66μg/m3
NMB=-44.32%
r=0.74
MB=-6.66μg/m3
NMB=-44.32%
PM10 (μg/m3)
avg urb Obs (nb tot = 9)
avg rur Obs (nb tot = 0)
avg all Obs (nb tot = 9)
EMEP/MSC-W
03 Dec - 00:00
03 Dec - 12:00
04 Dec - 00:00
04 Dec - 12:00
05 Dec - 00:00
05 Dec - 12:00
06 Dec - 00:00
06 Dec - 12:00


Madrid: 9 grids - 20161204
r=0.76
MB=-4.49μg/m3
NMB=-29.28%
r=0.76
MB=-4.49μg/m3
NMB=-29.28%
PM10 (μg/m3)
avg urb Obs (nb tot = 10)
avg rur Obs (nb tot = 0)
avg all Obs (nb tot = 10)
EMEP/MSC-W
04 Dec - 00:00
04 Dec - 12:00
05 Dec - 00:00
05 Dec - 12:00
06 Dec - 00:00
06 Dec - 12:00
07 Dec - 00:00
07 Dec - 12:00

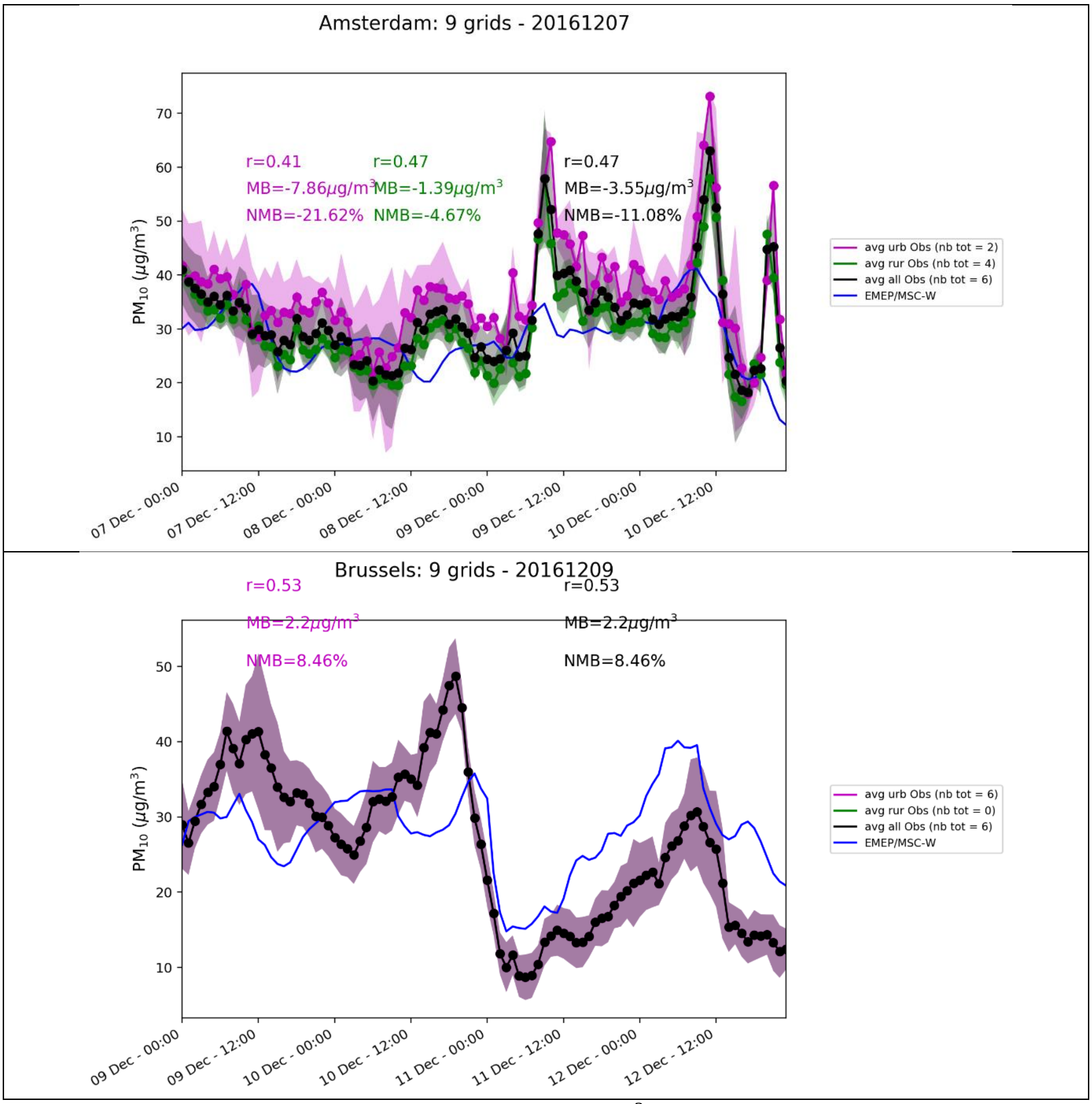


Fig S2 Hourly averaged PM_{10} concentrations (in μg/m^3) measured by the AirBase stations for several examples. The cities are defined by an area using 9 grid cells. The mean of all the stations is plotted in black, the urban stations are plotted in magenta and the rural stations in green. The coloured shade error corresponds to the standard deviation. The number of maximum of stations available during the period is provided. The EMEP model concentrations are plotted in blue. The correlation coefficient, the mean bias and the normalised mean bias for these 4-day forecasts are provided. The mean bias is calculated as MB = $\frac{\sum_{i=1}^{N}(M_i - O_i)}{N}$, and the normalised mean bias as NMB = $\frac{\sum_{i=1}^{N}(M_i - O_i)}{\sum_{i=1}^{N} O_i} \times 100\%$, with M referring to the model, O to the observations and N is the number of observations. The examples are for London, Paris, Vienna, Bratislava for predictions from 02 to 05 December 2016; Lisbon for predictions from 03 to 06 December 2016; Madrid for predictions from 04 to 07 December 2016, Amsterdam for predictions from 07 to 10 December 2016; and Brussels for predictions from 09 to 12 December 2016.

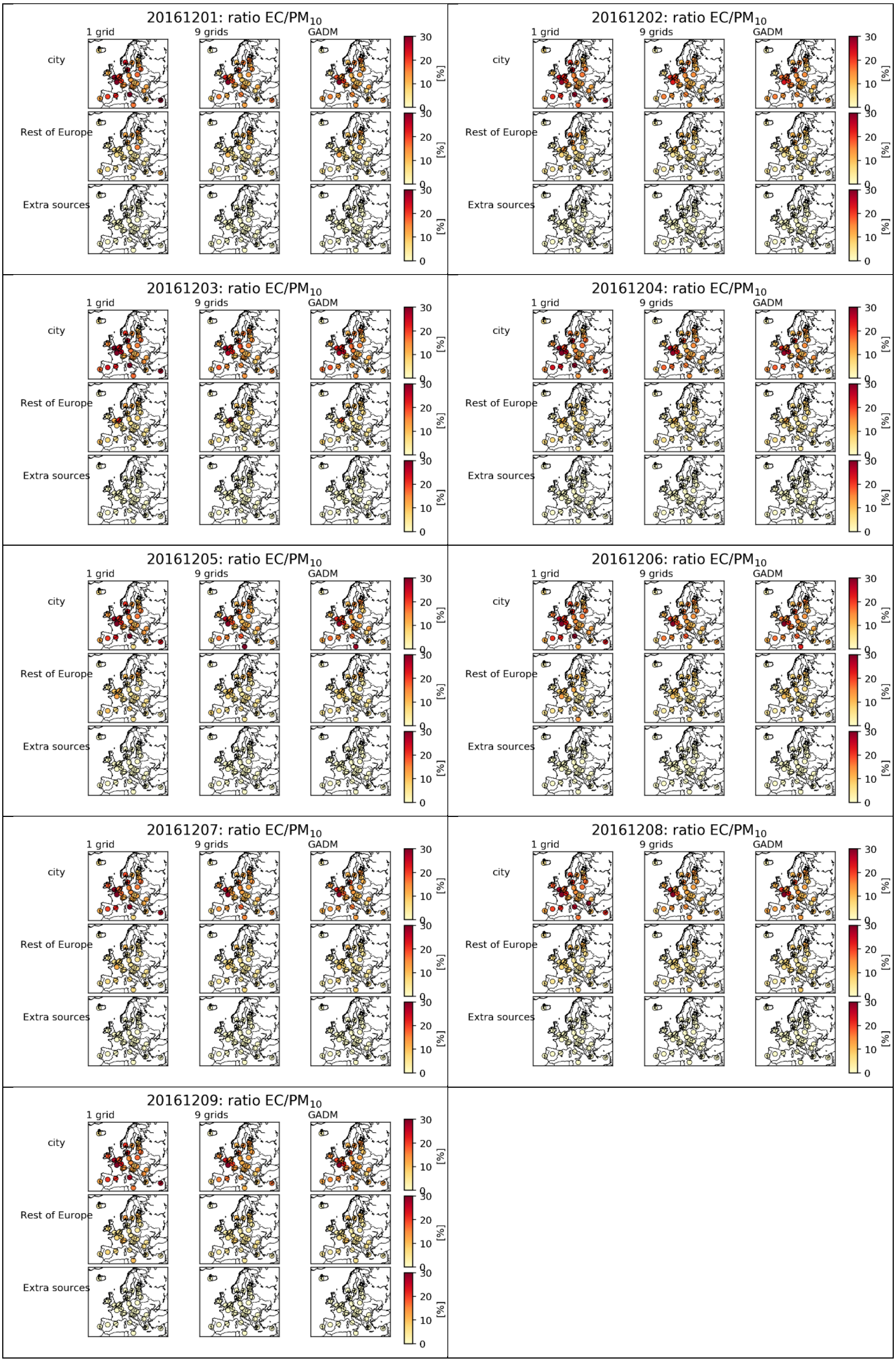
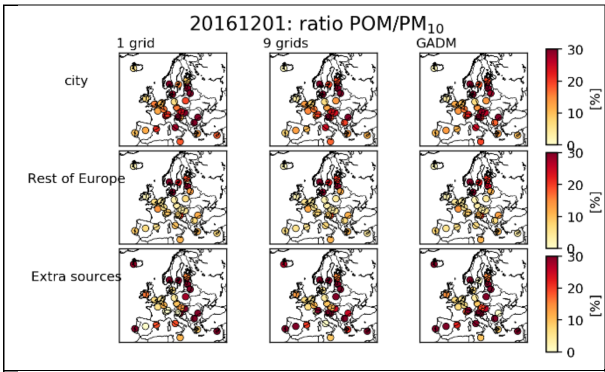
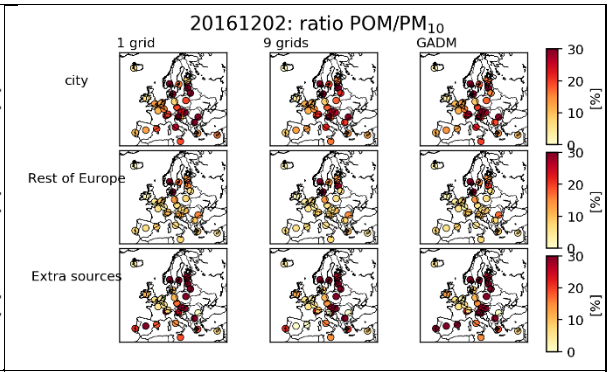
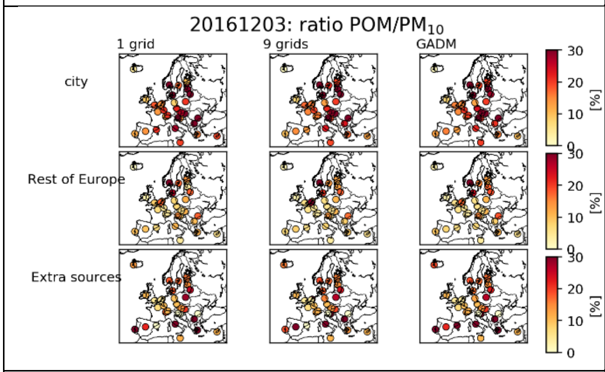
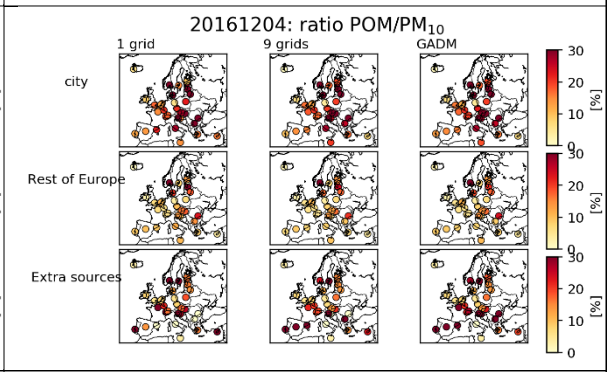

20161201: ratio EC/PM$_{10}$
20161202: ratio EC/PM$_{10}$
20161203: ratio EC/PM$_{10}$
20161204: ratio EC/PM$_{10}$
20161205: ratio EC/PM$_{10}$
20161206: ratio EC/PM$_{10}$
20161207: ratio EC/PM$_{10}$
20161208: ratio EC/PM$_{10}$
20161209: ratio EC/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
0
10
20
30
[%]

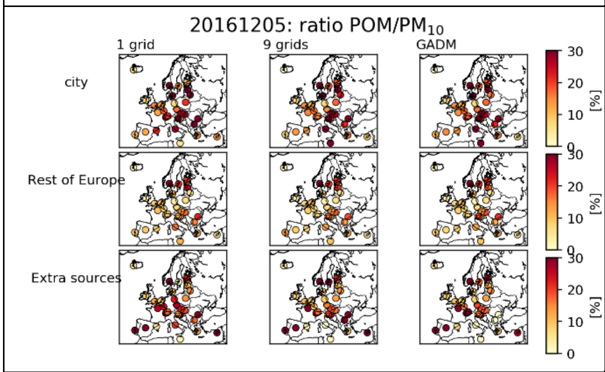
Fig. S3 Mean EC/PM_{10} ratio in percent for each individual forecasted day (from 01 December to 09 December 2016) over each city and for three city definitions (1 grid cell, 9 grid cells and GADM) and for the City, Rest of Europe and Extra Sources contributions. The results shown are for the 15% perturbation runs.

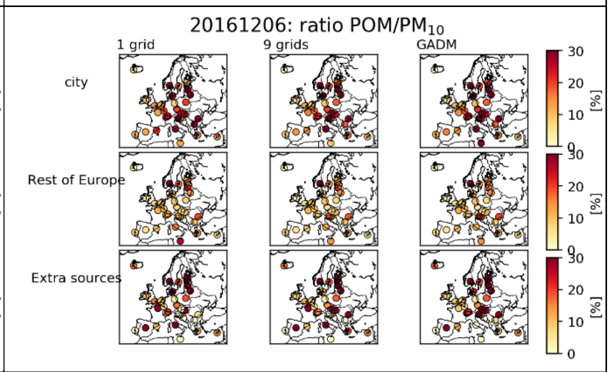

20161201: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]

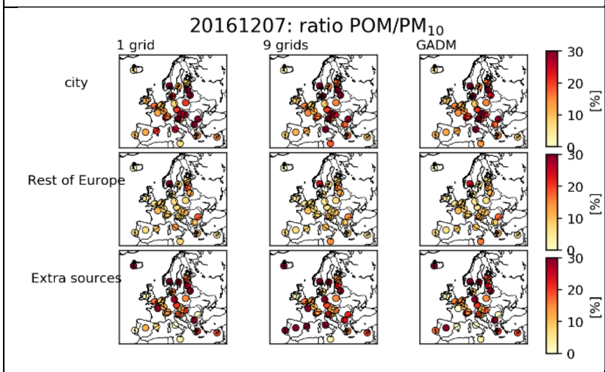

20161202: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]

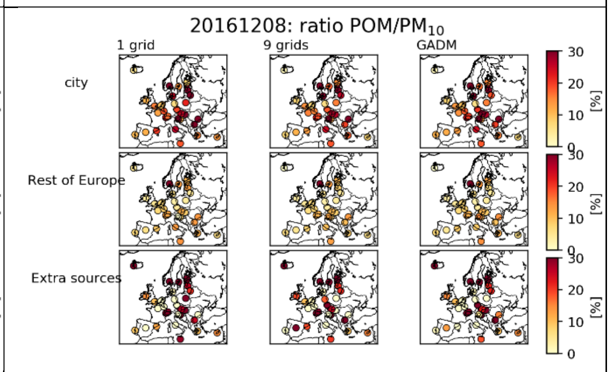

20161203: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]


20161204: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]


20161205: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]


20161206: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]


20161207: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]


20161208: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]

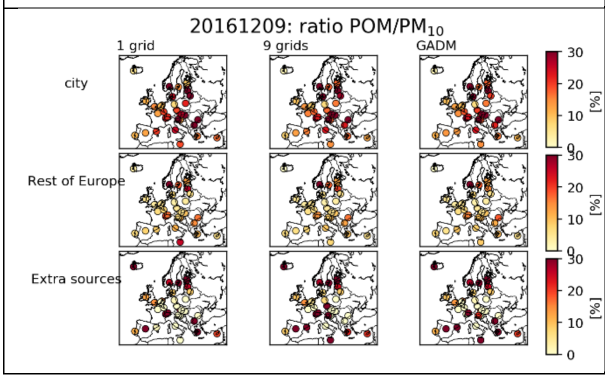
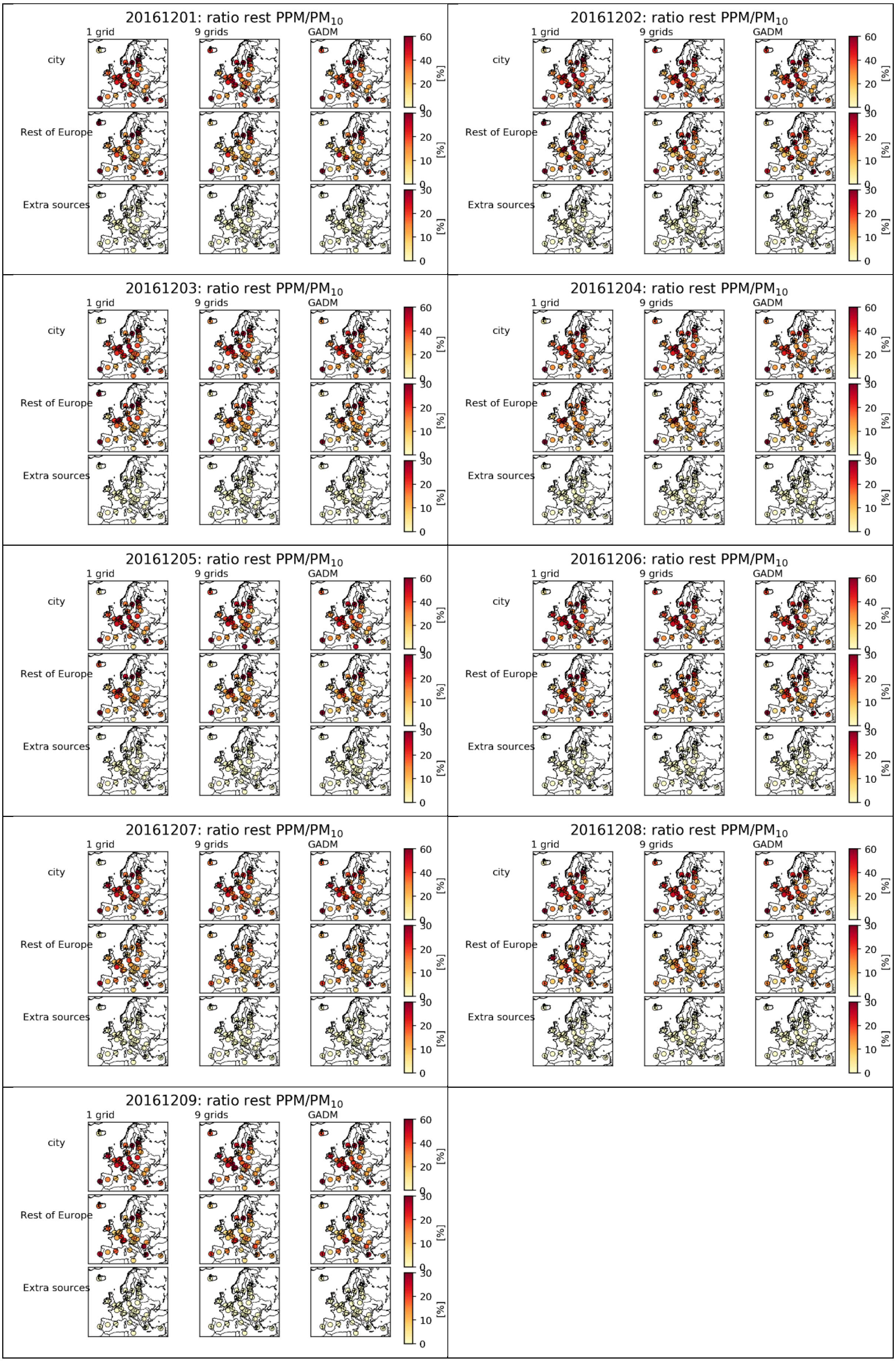

20161209: ratio POM/PM10
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
30
20
10
0
[%]

Fig. S4 Mean POM/PM_{10} ratio in percent for each individual forecasted day (from 01 December to 09 December 2016) over each city and for three city definitions (1 grid cell, 9 grid cells and GADM) and for the City, Rest of Europe and Extra Sources contributions. The results shown are for the 15% perturbation runs.



20161201: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
60
40
20
0
30
20
10
0
20161202: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161203: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161204: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161205: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161206: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161207: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161208: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
20161209: ratio rest PPM/PM$_{10}$
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]

Fig. S5 Mean rest PPM/PM_{10} ratio in percent for each individual forecasted day (from 01 December to 09 December 2016) over each city and for three city definitions (1 grid cell, 9 grid cells and GADM) and for the City, Rest of Europe and Extra Sources contributions. The results shown are for the 15% perturbation runs. Note the different colour scale for the city contribution.

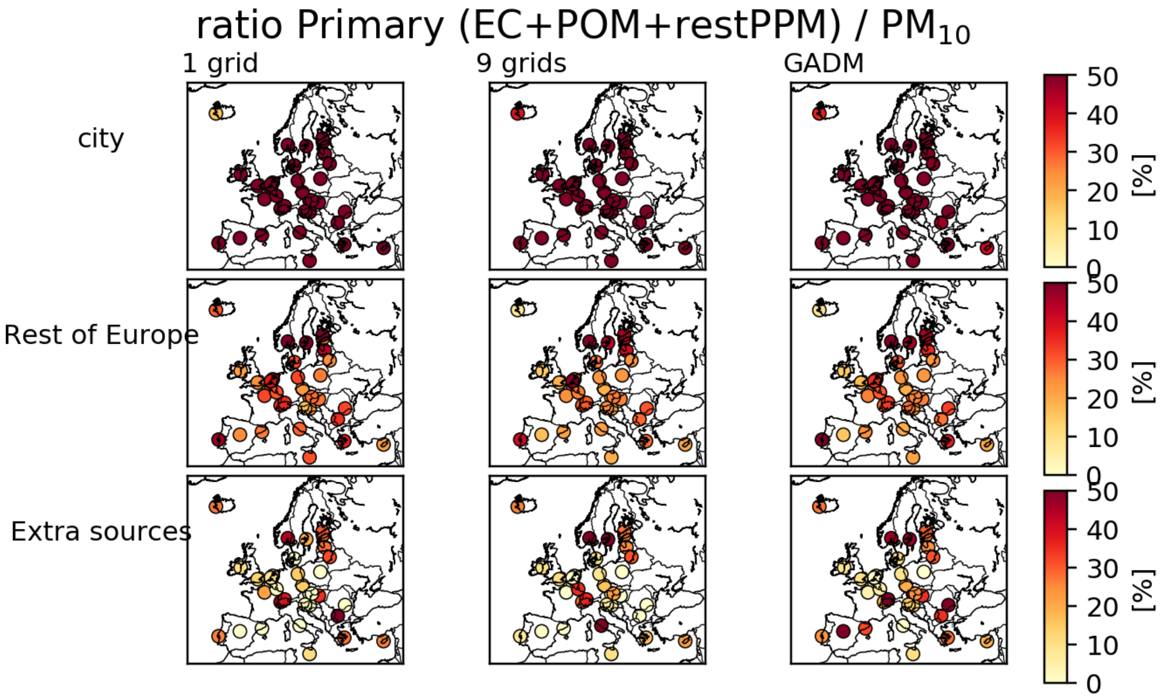


Fig. S6 Mean Primary aerosols/PM_{10} ratio in percent over the studied cities for three city definitions (1 grid cell, 9 grid cells and GADM) for all predicted PM_{10} concentrations (for all 4-day forecasts) and for the City, Rest of Europe and Extra Sources contributions, based on the calculations performed by the 15% perturbation runs.

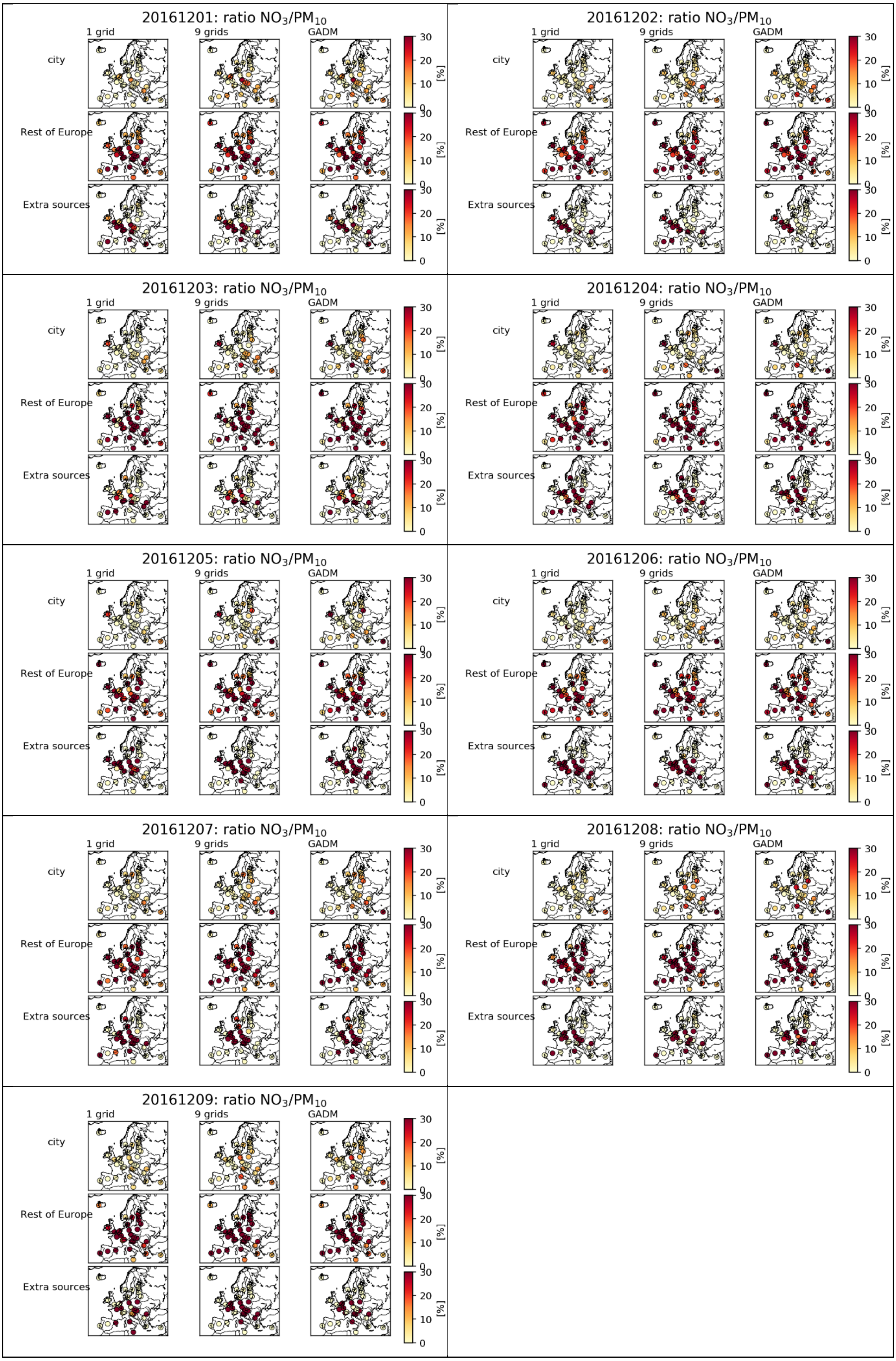
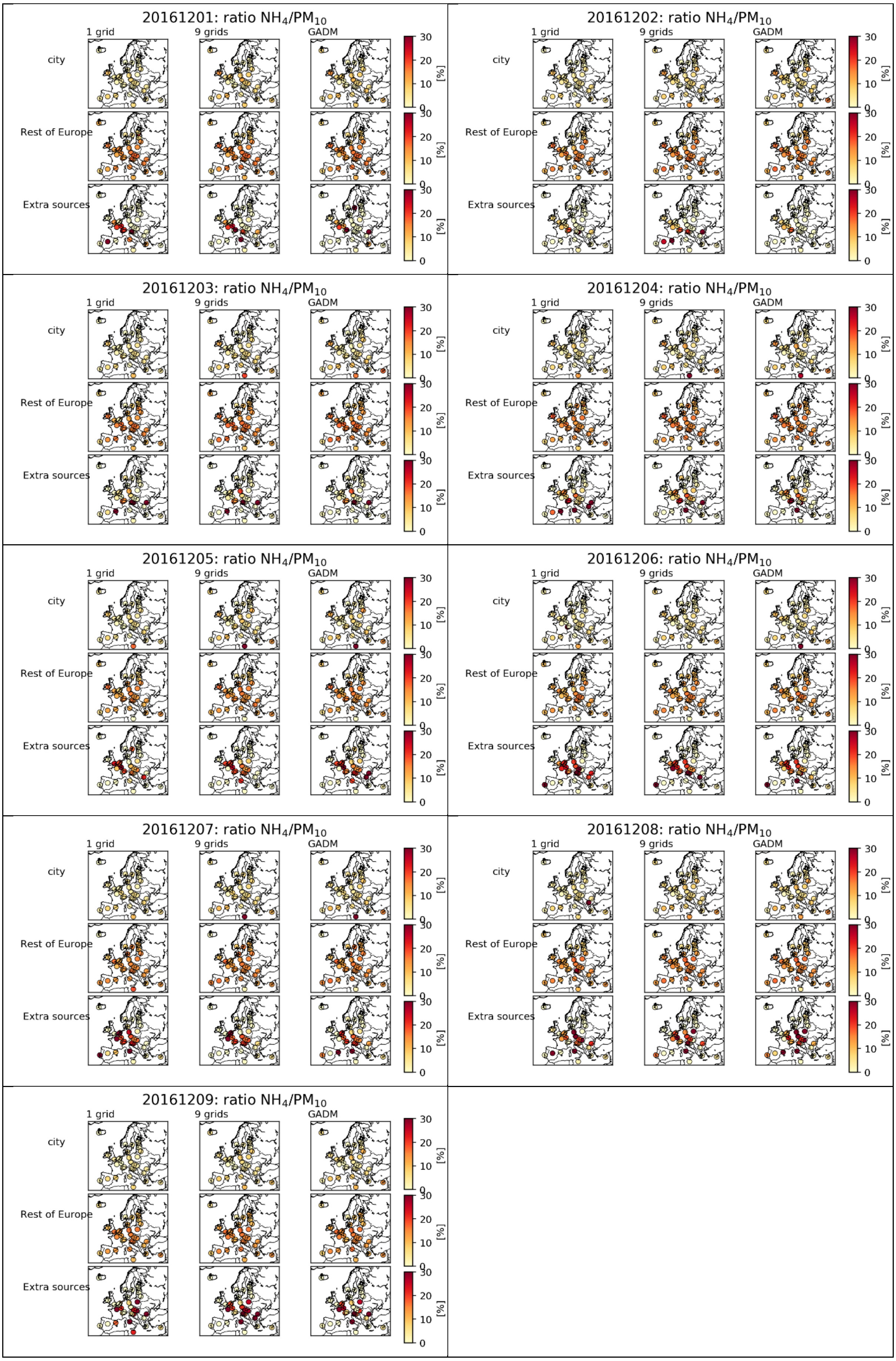

20161201: ratio NO_3/PM_{10}
20161202: ratio NO_3/PM_{10}
20161203: ratio NO_3/PM_{10}
20161204: ratio NO_3/PM_{10}
20161205: ratio NO_3/PM_{10}
20161206: ratio NO_3/PM_{10}
20161207: ratio NO_3/PM_{10}
20161208: ratio NO_3/PM_{10}
20161209: ratio NO_3/PM_{10}
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
[%]
0
10
20
30

Fig. S7 Mean NO_3/PM_{10} ratio in percent for each individual forecasted day (from 01 December to 09 December 2016) over each city and for three city definitions (1 grid cell, 9 grid cells and GADM) and for the City, Rest of Europe and Extra Sources contributions. The results shown are for the 15% perturbation runs.



20161201: ratio NH_4/PM_{10}
20161202: ratio NH_4/PM_{10}
20161203: ratio NH_4/PM_{10}
20161204: ratio NH_4/PM_{10}
20161205: ratio NH_4/PM_{10}
20161206: ratio NH_4/PM_{10}
20161207: ratio NH_4/PM_{10}
20161208: ratio NH_4/PM_{10}
20161209: ratio NH_4/PM_{10}
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
0
10
20
30
[%]

Fig. S8 Mean NH_4/PM_{10} ratio in percent for each individual forecasted day (from 01 December to 09 December 2016) over each city and for three city definitions (1 grid cell, 9 grid cells and GADM) and for the City, Rest of Europe and Extra Sources contributions. The results shown are for the 15% perturbation runs.

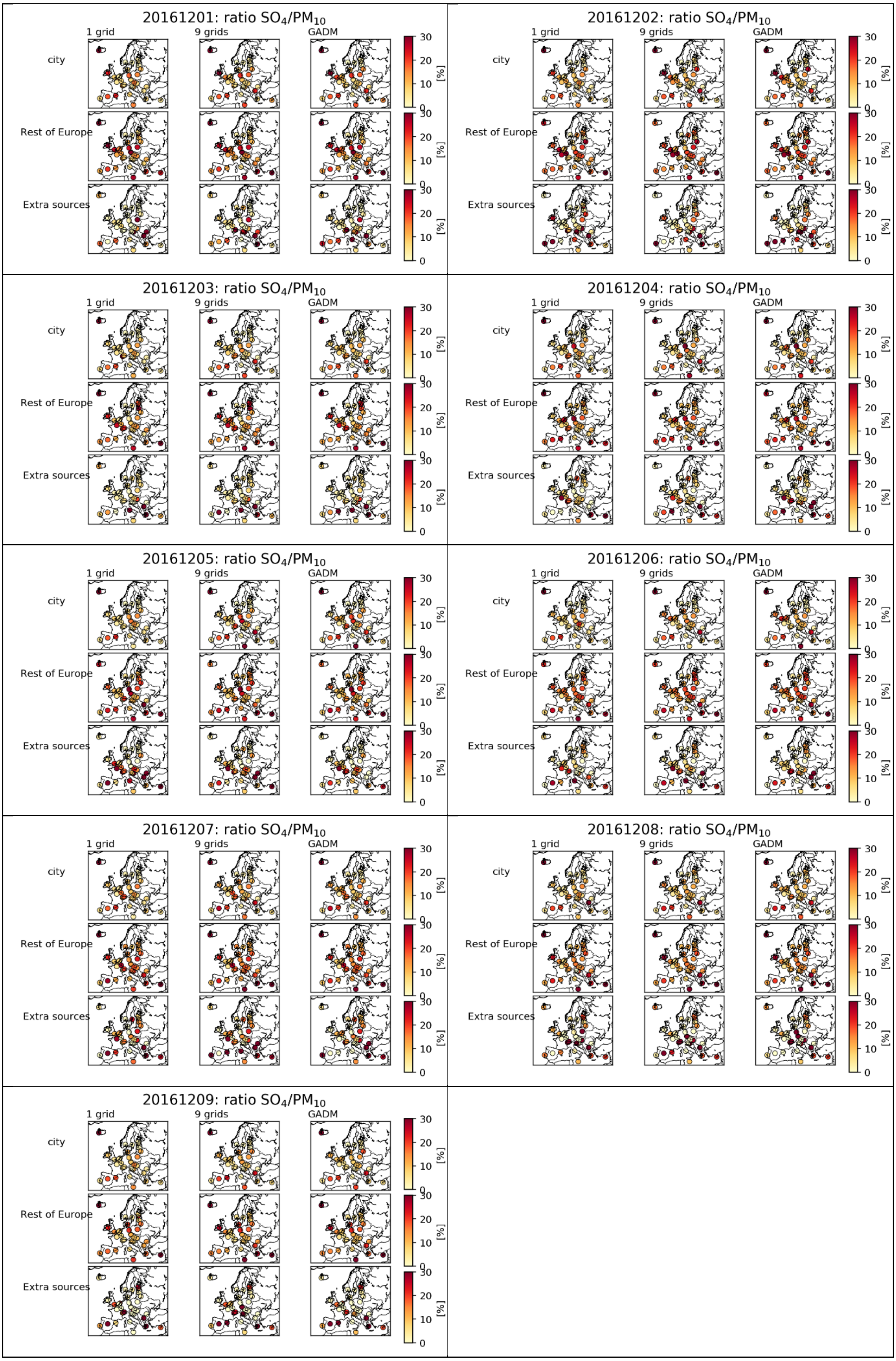

20161201: ratio SO_4/PM_{10}
20161202: ratio SO_4/PM_{10}
20161203: ratio SO_4/PM_{10}
20161204: ratio SO_4/PM_{10}
20161205: ratio SO_4/PM_{10}
20161206: ratio SO_4/PM_{10}
20161207: ratio SO_4/PM_{10}
20161208: ratio SO_4/PM_{10}
20161209: ratio SO_4/PM_{10}
1 grid
9 grids
GADM
city
Rest of Europe
Extra sources
0
10
20
30
[%]

Fig. S9 Mean SO_4/PM_{10} ratio in percent for each individual forecasted day (from 01 December to 09 December 2016) over each city and for three city definitions (1 grid cell, 9 grid cells and GADM) and for the City, Rest of Europe and Extra Sources contributions. The results shown are for the 15% perturbation runs.

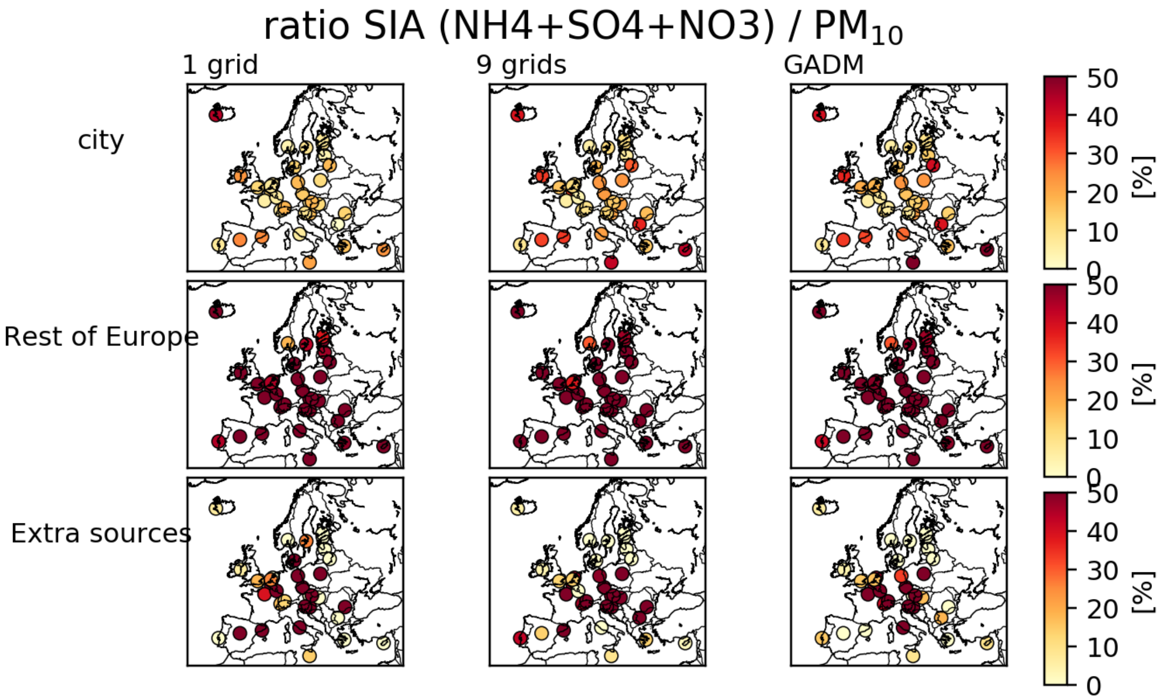


Fig. S10 Mean SIA/PM_{10} ratio in percent over the studied cities for three city definitions (1 grid cell, 9 grid cells and GADM) for all predicted PM_{10} concentrations (for all 4-day forecasts), and for the City, Rest of Europe and Extra Sources contributions, based on the calculations performed by the 15% perturbation runs.

Table S1. Input meteorological data used in the EMEP Model.

Parameter	Unit	Description
3D fields		**for σ levels**
u,v	*m/s*	Horizontal wind velocity components
q	*kg/kg*	Specific humidity
θ	K	Potential temperature
CW	kg/kg	Cloud water
CL	%	3D Cloud cover
cnvuf	kg/sm^2	Convective updraft flux
cnvdf	kg/sm^2	Convective downdraft flux
PR	*mm*	Precipitation
2D fields		**for surface**
PS	*hPa*	Surface pressure
T2	*K*	Temperature at 2 m height
RH2	%	Relative humidity at 2 m height
SR	W/m^2	Surface flux of sensible heat
τ	N/m^2	Surface stress
SST	*K*	Sea Surface Temperature
SWC	m^3/m^3	Soil water content
lspr	*m*	Large scale precipitation
cpr	*m*	Convective precipitation
sdepth	*m*	Snow depth
ice	%	Fraction of ice
SMI1		Soil moisture index level 1
SMI3		Soil moisture index level 3
u10, v10	*m/s*	Wind at 10 m height

Table S2. Correlation coefficient, bias and normalised mean bias between the observations and the corresponding EMEP model value, for the wind speed at 10 m, relative humidity at 2m and the temperature at 2m, using each individual day between 01 and 09 December 2016. The mean bias is calculated as $MB = \frac{\sum_{i=1}^{N}(M_i - O_i)}{N}$, and the normalised mean bias as $NMB = \frac{\sum_{i=1}^{N}(M_i - O_i)}{\sum_{i=1}^{N} O_i} \times 100\%$, with M referring to the model, O to the observations and N is the number of observations. The numbers in square brackets correspond to the numbers of stations available within the area of 9 grid cells used to define each city. The numbers in parentheses below the correlation coefficients are the numbers of hourly observations available during the period. If some stations are located in a same model grid, the hourly observations of these stations are averaged before to be compared to the model.

City	wind speed at 10 m			relative humidity at 2 m			temperature at 2 m		
	r	bias (m/s)	NMB (%)	r	bias (%)	NMB (%)	r	bias (K)	NMB (%)
Amsterdam [1]	0.91 (216)	0.03	0.8	0.65 (216)	-0.16	-0.19	0.95 (216)	-0.55	-0.2
Athens [5]	0.83 (574)	0.5	16.27	0.77 (572)	7.02	10.77	0.85 (574)	-1.4	-0.49
Barcelona [2]	0.69 (422)	0.39	12.56	0.8 (422)	0.22	0.27	0.89 (422)	0.3	0.11
Berlin [1]	0.95 (216)	-0.23	-5.53	0.78 (216)	-2.97	-3.36	0.98 (216)	-0.27	-0.1
Bern [5]	0.25 (1069)	0.13	11.52	0.49 (1066)	-23.68	-28.32	0.65 (1066)	0.78	0.29
Bratislava [3]	0.78 (424)	0.71	21.62	0.5 (541)	-5.51	-6.73	0.81 (541)	0.1	0.04
Brussels [4]	0.78 (758)	0.74	28.94	0.5 (604)	-2.51	-2.85	0.93 (793)	-0.25	-0.09
Bucharest [3]	0.72 (643)	0.91	36.89	0.7 (647)	-0.71	-0.97	0.93 (647)	-0.73	-0.27
Budapest [2]	0.91 (216)	-0.16	-4.63	0.72 (216)	-4.06	-4.9	0.91 (216)	-0.04	-0.01
Copenhagen [4]	0.88 (854)	0.62	10.79	0.79 (854)	-1.4	-1.58	0.92 (854)	0.05	0.02
Dublin [2]	0.81 (432)	-0.55	-13.29	0.67 (432)	2.07	2.37	0.93 (432)	-0.4	-0.14
Helsinki [6]	0.92 (859)	-0.18	-2.84	0.81 (1079)	-3.32	-3.78	0.96 (1079)	0.16	0.06
Lisbon [6]	0.69 (306)	0.42	12.96	0.82 (306)	-1.6	-1.81	0.84 (306)	0.28	0.1
Ljubljana [2]	0.01 (160)	-0.31	-29.58	0.66 (215)	-4.2	-5.22	0.82 (233)	-0.3	-0.11
London [4]	0.8 (556)	-0.06	-1.63	0.81 (553)	5.17	6.02	0.95 (556)	-1.49	-0.53
Luxembourg [1]	0.84 (216)	-0.15	-3.18	0.69 (216)	2.07	2.3	0.96 (216)	-1.19	-0.44
Madrid [4]	0.65 (486)	0.39	24.83	0.76 (486)	-0.14	-0.16	0.9 (486)	-0.54	-0.19
Nicosia [5]	0.55 (658)	-0.26	-9.32	0.79 (658)	2.91	4.23	0.9 (658)	-0.61	-0.21
Oslo [5]	0.42 (379)	1.03	60.64	0.65 (600)	1.8	2.23	0.88 (600)	-0.68	-0.25
Paris [7]	0.63 (864)	0.58	29.17	0.64 (864)	9.51	11.95	0.87 (864)	-1.21	-0.44

Prague [5]	0.83 (647)	0.07	2.05	0.74 (647)	-4.27	-5.22	0.94 (647)	-0.46	-0.17
Reykjavik [3]	0.68 (214)	-0.94	-18.61	0.84 (214)	8.61	10.45	0.6 (214)	-1.65	-0.59
Riga [1]	0.59 (216)	1.44	38.38	0.76 (216)	-6.42	-7.16	0.94 (216)	-0.14	-0.05
Rome [4]	0.28 (604)	0.27	13.13	0.73 (604)	3.55	4.81	0.89 (604)	-0.13	-0.05
Rotterdam [2]	0.85 (430)	-0.4	-10.42	0.72 (216)	0.21	0.24	0.93 (216)	-0.53	-0.19
Sofia [1]	0.7 (216)	-0.5	-16.69	0.53 (216)	-5.76	-7.69	0.77 (216)	-0.99	-0.36
Stockholm [3]	0.85 (216)	0.21	5.82	0.77 (216)	5.46	6.83	0.97 (216)	-1.42	-0.52
Tallinn [1]	0.91 (216)	-0.39	-9.03	0.74 (216)	0.13	0.15	0.94 (216)	-1.05	-0.39
Valetta [1]	0.75 (216)	1.25	36.22	0.86 (216)	-4.67	-6.02	0.85 (216)	0.79	0.27
Vienna [5]	0.77 (819)	0.42	10.99	0.51 (862)	-4.82	-6.07	0.79 (862)	-0.47	-0.17
Vilnius [1]	0.84 (216)	-0.15	-3.18	0.69 (216)	2.07	2.3	0.96 (216)	-1.19	-0.44
Warsaw [2]	0.77 (306)	0.62	14.92	0.73 (306)	2.06	2.36	0.94 (306)	-0.58	-0.21
Zagreb [4]	0.22 (396)	-0.33	-20.23	0.65 (396)	-2.72	-3.62	0.79 (396)	0.44	0.16
Zurich [7]	0.14 (1065)	0.34	38.13	0.31 (1065)	-7.66	-8.72	0.62 (1065)	0.03	0.01
All cities [112]	0.84 (16085)	0.25	8.08	0.59 (16369)	-1.95	-2.38	0.95 (16581)	-0.35	-0.13

Table S3. Total anthropogenic emissions over Paris and London for each day from 01 to 09 December 2016, for CO, NH_3, NO_x, fine primary particulate matter (denoted as $PM_{2.5}$), coarse primary particulate matter (denoted as PMco) SO_x, and NMVOC as used in the EMEP model.

Emission (kg)	**01 Dec**	**02 Dec**	**03 Dec**	**04 Dec**	**05 Dec**	**06 Dec**	**07 Dec**	**08 Dec**	**09 Dec**
Paris									
CO	82.7	86.2	73.5	72.9	83.0	81.7	82.1	82.4	85.9
NH_3	0.7	0.7	0.6	0.6	0.7	0.7	0.7	0.7	0.7
NO_x	53.3	55.9	47.2	46.5	53.8	52.9	53.2	53.6	56.2
$PM_{2.5}$	5.9	6.1	4.6	4.3	5.9	5.8	5.9	5.9	6.1
PMco	3.8	3.9	2.4	1.8	3.8	3.8	3.8	3.8	3.8
SO_x	8.6	8.6	7.5	7.2	8.8	8.8	8.9	9.0	9.1
NMVOC	52.6	53.2	35.9	21.4	52.2	51.9	51.9	51.7	52.3
London									
CO	116.8	120.3	101.2	100.6	115.7	114.7	116.4	115.7	119.2
NH_3	1.2	1.3	1.0	1.0	1.2	1.2	1.2	1.2	1.3
NO_x	43.9	45.1	38.8	38.8	43.3	43.1	43.7	43.4	44.6
$PM_{2.5}$	2.8	2.9	2.3	2.3	2.8	2.8	2.8	2.8	2.9
PMco	1.5	1.5	1.1	0.9	1.5	1.5	1.5	1.5	1.5
SO_x	3.8	3.7	3.1	3.1	3.7	3.7	3.8	3.7	3.7
NMVOC	46.2	46.3	30.7	17.4	45.8	45.8	45.9	45.6	45.6