



## Supplement of

### Sensitivity of air quality model responses to emission changes: comparison of results based on four EU inventories through FAIRMODE benchmarking methodology

Alexander de Meij et al.

Correspondence to: Philippe Thunis (philippe.thunis@ec.europa.eu)

The copyright of individual parts of the supplement might differ from the article licence.

# Sensitivity of air quality model responses to emission changes: comparison of results based on four EU inventories through FAIRMODE benchmarking methodology.

Alexander de Meij<sup>1</sup>, Cornelis Cuvelier<sup>2</sup>, Philippe Thunis<sup>2</sup>, Enrico Pisoni<sup>2</sup>, Bertrand Bessagnet<sup>2</sup>
 <sup>1</sup>MetClim, Varese, 21025, Italy

<sup>2</sup> European Commission, Joint Research Centre (JRC), 21027, Ispra, Italy

retired with Active Senior Agreement

10

Country	City	,	lon	lon	lat	lat
-	-		min	max	min	max
Belgium	Brussels	BRU	4.05	4.65	50.55	51.15
Germany	Berlin	BER	12.76	14.06	51.87	53.17
Italy	Rome	ROM	12.09	12.89	41.5	42.3
Spain	Madrid	MAD	-4.15	-3.25	39.96	40.86
Sweden	Stockholm	STO	17.62	18.52	58.88	59.78
Romania	Bucharest	BUC	25.90	26.30	44.23	44.63
Malopolska region		MAL	18.0	23.0	48.7	51.5
Po Valley		POV	6.5	14.0	43.5	47.0

#### Table S1 List of local domains selected, with their corresponding area for emission reductions.

Table S2 Overview GNFR sectors
--------------------------------

GNFR sector
A PublicPower
B Industry
C OtherStationaryComb
D Fugitive
E Solvents
F RoadTransport
G Shipping
H Aviation
I OffRoad
J Waste
K AgriLiveStock
L AgriOther
M Other

#### 15

#### Table S3. Overview total emissions per emission inventory for the different locations, mg/m2/day

PPM	EDGAR	EMEP-GNFR	CAMS221	CAMS42C
Berlin	1.82	1.38	1.64	1.93
Brussels	4.38	4.34	5.19	4.94
Bucharest	7.63	3.06	8.04	8.16
Madrid	3.24	2.41	3.06	3.47
Malopolska	3.49	2.41	2.68	3.21
Po Valley	1.65	1.45	1.40	1.51
Rome	2.63	2.48	2.44	2.52
Stockholm	3.33	3.09	1.69	2.19

VOC	EDGAR	EMEP-GNFR	CAMS221	CAMS42C
Berlin	15.65	7.67	8.55	8.78
Brussels	40.50	15.29	16.13	14.93
Bucharest	26.35	15.73	28.15	21.40
Madrid	21.28	18.97	20.31	19.00
Malopolska	12.11	6.71	6.35	7.06
Po Valley	6.65	6.72	6.08	5.74
Rome	21.05	16.66	17.69	16.02
Stockholm	18.13	7.63	9.69	9.46

NOx	EDGAR	EMEP-GNFR	CAMS221	CAMS42C
Berlin	9.76	8.50	7.72	8.24
Brussels	31.57	27.21	31.05	28.16
Bucharest	13.94	17.01	19.93	18.28
Madrid	16.15	16.59	19.44	15.05
Malopolska	7.36	7.79	8.20	8.04
Po Valley	5.23	5.62	5.48	5.16
Rome	12.01	11.54	12.26	10.11
Stockholm	10.71	7.41	7.01	6.03

SOx	EDGAR	EMEP-GNFR	CAMS221	CAMS42C	
Berlin	3.82	1.48	1.65	1.57	
Brussels	7.23	2.56	3.07	2.75	
Bucharest	9.51	7.49	5.95	6.00	
Madrid	6.54	2.43	2.56	2.69	
Malopolska	8.72	8.71	8.69	8.86	
Po Valley	1.31	0.73	0.82	0.81	
Rome	2.25	0.74	0.94	0.79	
Stockholm	11.43	0.67	1.03	0.80	

NH3	EDGAR	EMEP-GNFR	CAMS221	CAMS42C	
Berlin	4.02	2.26	3.77	3.25	
Brussels	7.69	3.34	5.36	5.11	
Bucharest	3.34	4.40	6.89	6.69	
Madrid	3.24	1.91	3.13	3.60	
Malopolska	3.52	1.27	1.79	2.05	
Po Valley	4.58	3.16	3.24	3.17	
Rome	3.65	2.41	2.52	1.88	
Stockholm	1.67	0.61	1.06	0.60	

Table S4(a) Overview of Base Case emissions (mg/m2/day) for NOx and NH3x, together with the ratio in the emissions between these two pollutants. (b) Similar as to (a) but for potency at P95 in  $\mu$ g/m3.

(a)								
Emissions m	g/m2/day				Ratio emis	sions NOx/NH	13	
		EMEP-				EMEP-		
NO <sub>x</sub>	EDGAR	GNFR	CAMS221	CAMS42C	EDGAR	GNFR	CAMS221	CAMS42C
BER	9.764	8.501	7.717	8.236	2.43	3.76	2.05	2.53
BRU	31.572	27.208	31.046	28.161	4.10	8.16	5.79	5.52
BUC	13.944	17.014	19.926	18.279	4.18	3.86	2.89	2.73
MAD	16.149	16.586	19.437	15.053	4.99	8.70	6.21	4.18
MAL	7.36	7.79	8.195	8.039	2.09	6.14	4.57	3.93
POV	5.229	5.617	5.482	5.155	1.14	1.78	1.69	1.63
ROM	12.012	11.544	12.257	10.11	3.29	4.80	4.86	5.39
STO	10.709	7.411	7.01	6.027	6.42	12.22	6.62	10.03

Emissions mg/m2/day								
		EMEP-						
NH <sub>3</sub>	EDGAR	GNFR	CAMS221	CAMS42C				
BER	4.023	2.261	3.766	3.253				
BRU	7.694	3.335	5.362	5.106				
BUC	3.336	4.404	6.886	6.69				
MAD	3.238	1.907	3.13	3.598				
MAL	3.522	1.269	1.792	2.046				
POV	4.583	3.156	3.235	3.172				
ROM	3.652	2.406	2.524	1.875				
STO	1.668	0.6067	1.059	0.6006				

(b)								
Potency P95	(µg/m³/ton)				Ratio Poter	ncy NOx/NH3	5	
		EMEP-				EMEP-		
NOx	EDGAR	GNFR	CAMS221	CAMS42C	EDGAR	GNFR	CAMS221	CAMS42C
BER	-0.0018	-0.0011	-0.0024	-0.0018	0.11	0.07	0.19	0.14
BRU	0.0013	0.0013	0.0015	0.0012	-0.05	-0.02	-0.03	-0.02
BUC	-0.0067	-0.0047	-0.0019	-0.0017	0.07	0.12	0.05	0.05
MAD	0.0002	-0.0002	-0.0002	-0.0013	-0.01	0.01	0.01	0.07
MAL	-0.001	-0.0011	-0.0004	-0.001	0.20	0.09	0.05	0.10
POV	-0.0064	-0.0047	-0.0046	-0.0059	1.64	0.67	0.79	0.78
ROM	-0.022	-0.0076	-0.0151	-0.0089	0.27	0.10	0.17	0.10
STO	-0.0011	-0.0011	-0.0006	-0.0005	0.01	0.02	0.01	0.01
	•				•			

Potency P9	5 (µg/m³/ton)			
		EMEP-		
NH <sub>3</sub>	EDGAR	GNFR	CAMS221	CAMS42C
BER	-0.0162	-0.0168	-0.0125	-0.013
BRU	-0.0253	-0.0738	-0.0473	-0.0507
BUC	-0.095	-0.0384	-0.0398	-0.0375
MAD	-0.0229	-0.0227	-0.0193	-0.0174
MAL	-0.0051	-0.0116	-0.0082	-0.0098
POV	-0.0039	-0.007	-0.0058	-0.0076
ROM	-0.0803	-0.0773	-0.0884	-0.0928
STO	-0.0779	-0.0644	-0.0515	-0.049











PM25 total emissions EMEP-GNFR Berlin mg/m2 54°N 53°30'N 53°N 52°30'N 52°N 51°30'N 51°N 12°E 12°30'E 13°E 13°30'E 14°E 14°30'E 15°E



(c)



40





(d)





PM25 total emissions CAMS221 Brussels mg/m2





PM25 total emissions EMEP-GNFR Brussels mg/m2

51°40'N

51°20'N

51°N

51°20'N 50°40'N 50°20'N 50°N 4°E 4°20'E 4°40'E 5°E

45



(e)

(g)

(h)





(i)

(k)























(m)

(o)





(t)







(s)







49°30'N

49°N

18°E

19°E

20°E

21°E

500 1000 1500 2000 2500 3000 3500 4000 4500 5000

22°E

23°E

(ab)



49°30'N

49°N

18°E

20°E

21°E

500 1000 1500 2000 2500 3000 3500 4000 4500 5000

19°E

22°E

23°E

(aa)







Figure S1: PM25 geographical distribution (mg/m2/year) for CAMS221, CAMS422C, EDGAR and EMEP-GNFR for Berlin, (a-d) Brussels (e-h), Bucharest (i-l), Madrid (m-p), Rome (q-t), Stockholm (u-x), Malopolska (y-ab) and Po Valley (ac-af). The red rectangle represents the area for which the emissions were reduced, as indicated in Table S1. For the Po Valley and Malopolska the emissions are reduced over the entire domain.



Figure S2: Yearly average relative potential for 50% reduction of (a) NOx, (b) SOx, (c) NH3 and (d) all precursors together (ALL: VOC, SOx, NH3, PPM) on relative PM10 concentration change. The values given represent 95 Percentile values (P95), showing the highest 5% values in the domain for the BaseCase. EDGAR (red), EMEP-GNFR (light blue), CAMS221 (blue) and CAMS42C (green), for the eight locations (Berlin, Brussels, Bucharest, Madrid, Malopolska region, Po Valley region, Rome and Stockholm.



100

Figure S3: Yearly average absolute potential for 50% reduction of (a) NOx, (b) SOx, (c) NH3 and (d) all precursors together (ALL: VOC, SOx, NH3, PPM) on relative PM10 concentration change. The values given represent 95 Percentile values (P95), showing the highest 5% values in the domain for the BaseCase. EDGAR (red), EMEP-GNFR (light blue), CAMS221(blue) and CAMS42C (green), for the eight locations (Berlin, Brussels, Bucharest, Madrid, Malopolska region, Po Valley region, Rome and Stockholm.

When NOx emissions are reduced by 50% (Fig. S2 and S3), we see in general a decrease in calculated PM10 concentrations, but for Brussels we see a slight increase. The reason for this is that over urbanized areas, lower

NO2 concentrations at constant or similar VOC concentrations lead to an increase of ozone (O3) values. O3 is a

105 reactive oxidant in natural and polluted atmosphere and increasing levels of O3 concentrations lead to an enhancement of the atmospheric oxidizing capacity, which might lead to an increase in the Secondary Organic Aerosol (SOA) formation and Organic Aerosol (OA) components (Thunis et al., 2021a and references therein). It appears that reducing PPM emissions by 50% shows the largest effective measure to reduce PM10 concentrations when compared to the aerosol precursors.



115 Figure S4: Yearly average absolute potency for 50% reduction of (a) PPM, (b) NOx, (c) SOx, (d) NH3 on PM10 concentration change. The values given represent 95 Percentile values, showing the highest 5% values in the domain for the BaseCase. EDGAR (red), EMEP-GNFR (light blue), CAMS221 (blue) and CAMS42C (green), for the eight locations (Berlin [BER], Brussels [BRU], Bucharest [BUC], Madrid [MAD], Malopolska region [MAL], Po Valley region [POV], Rome [ROM] and Stockholm [STO]).

The very low values in the potencies for Malopolska and Po Valley can be explained by the fact that we divide by the emissions that are much larger, because they cover an entire region rather than a city. We see that the most effective reduction of the secondary aerosol part is obtained by reducing SOx emissions,

because the potencies for SOx are in general higher than for NOx.



Figure S5: Median relative potential for PM10 for the eight locations, Berlin [BER], Brussels [BRU], Bucharest [BUC], Madrid [MAD], Malopolska region [MAL], Po Valley region [POV], Rome [ROM] and Stockholm [STO]). The median is based on EDGAR, CAMS221 and EMEP-GNFR. The values given represent 95 Percentile values, showing the highest 5% values in the domain for the BaseCase.



Figure S6: Yearly average relative potential for 50% reduction of (a) NOx, (b) VOC and (c) ALL (NOx + VOC) on relative O3 concentration change. The values given represent 95 Percentile values (P95), showing the highest 5% values in the domain for the BaseCase. EDGAR (red), EMEP-GNFR (light blue), CAMS221 (blue) and CAMS42C (green), for the eight locations (Berlin [BER], Brussels [BRU], Bucharest [BUC], Madrid [MAD], Malopolska region [MAL], Po Valley region [POV], Rome [ROM] and Stockholm [STO]).

Whether reductions in NOX or VOC emissions will lead to lower O3 concentrations depend on location and on the type of chemical regime, also better known as NOx - or VOC-limited regimes. This means that for NOx limited regimes (locations downwind of urban and suburban areas), lowering NOx concentrations at constant VOC levels or in combination with lowering VOCs results in lower O3 peak concentrations. So, decreasing the available NOx leads directly to a decrease in ozone. In VOC-limited areas (highly polluted urban areas), where VOCs are kept constant, but NOx emissions are reduced, lead to, opposite, higher O3 concentrations (Fig. S6a).

150 On the other hand, lowering VOCs and keeping NOx constant lead to reduced O3 values (Fig. S6b). When VOCs and NOx are decreased proportionately at the same time O3 increase (Fig. S6c).

The underlying reason for the increase when NOx emissions are reduced is that less O3 is removed by NO (NOx
155 -titration), therefore augmenting O3 values in VOC-limited zones, as mentioned earlier. Therefore, we see in general an increase in O3 values over the urban areas. The side-effect of reducing NOx emissions in urban areas

(e.g. via traffic) lead to higher O3 concentrations and possible exceedances in cities that are currently below the O3 limit values.

- 160 The negative RP and AP for POV (Fig. S6 and S7) can be explained by the fact that the domain is large when compared to cities (e.g. Brussels, Bucharest, Madrid) and that background concentrations might have an impact on the P95 values. In sub-urban and background areas O3 decrease when NOx emissions are reduced, as mentioned in the main text.
- **165** Fig. S6a shows that for Malopolska, the 50% NOx reduction can lead to an increase or decrease on O3 concentrations depending on the choice of emission inventory.



Figure S7: Yearly average absolute potential for 50% reduction of (a) NOx, (b) VOC and (c) ALL (NOx + VOC) on relative O3 concentration change. The values given represent 95 Percentile values (P95), showing the highest 5% values in the domain for the BaseCase. EDGAR (red), EMEP-GNFR (light blue), CAMS221 (blue) and CAMS42C (green), for the eight locations (Berlin [BER], Brussels [BRU], Bucharest [BUC], Madrid [MAD], Malopolska region [MAL], Po Valley region [POV], Rome [ROM] and Stockholm [STO]).



Figure S8: Yearly average potency for (a) 50% NOx and (b) 50% VOC reduction on O3 concentration change (µg/m3) per ton emission reduction by EDGAR (red), EMEP-GNFR (light blue), CAMS221 (blue) and CAMS42C (green), for the eight locations (Berlin [BER], Brussels [BRU], Bucharest [BUC], Madrid [MAD], Malopolska region [MAL], Po Valley region [POV], Rome [ROM] and Stockholm [STO]). The values given in (a) and (b) represent 95 Percentile values (P95), showing the highest 5% values in the domain for the BaseCase.



NOx

Figure S9: Median relative potential for O3 for the eight locations, Berlin [BER], Brussels [BRU], Bucharest [BUC], Madrid [MAD], Malopolska region [MAL], Po Valley region [POV], Rome [ROM] and Stockholm [STO]). The median is based on EDGAR, CAMS221 and EMEP-GNFR. The values given represent 95 Percentile values, showing the highest 5% values in the domain for the BaseCase.

VOC