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Sources of organic aerosols in Europe: a modeling study using CAMx with modified volatility basis set scheme

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Table S1 Description of semi-volatile organic compounds (SVOC) and intermediate-volatility organic compounds (IVOC). The same calculations were adopted for IVOC emissions in BASE and NEW. GV: Gasoline Vehicles; DV: Diesel Vehicles; BB: Biomass Burning; OthA: Other anthropogenic sources.

Species	Source	Calculations		References	Descriptions
		BASE	NEW		
SVOC	GV	= POA_GV	= 3 * POA_GV	Shrivastava et al., 2011; Tsimpidi et al., 2010; Ciarelli et al., 2017a	POA emissions of each source were calculated from TNO $PM_{2.5}$ emissions
	DV	= POA_DV	= 3 * POA_DV		
	BB	= POA_BB	= 3 * POA_BB		
	OthA	= POA_OthA	= 3 * POA_OthA		
IVOC	GV	= 25% * NMVOC_GV		Jathar et al., 2014	The portion of NMVOCs considered as IVOCs (25% for GV, 20% for DV) were removed from the NMVOC emissions
	DV	= 20% * NMVOC_DV		Jathar et al., 2014	
	BB	= 4.5 * POA_BB		Ciarelli et al., 2017	
	OthA	= 1.5 * POA_OthA		Robinson et al., 2007	

Table S2. Evaluation of the meteorological parameters (number of stations = 1094). Performance criteria for model results are from Emery et al., (2001). MB: mean bias; MGE: mean gross error; RMSE: root-mean-square error; IOA: index of agreement. DJF: December-January-February, MAM: March-April-May, JJA: June-July-August, SON: September-October-November.

Meteorological parameters*	Season	MB	*Criteria*	MGE	*Criteria*	RMSE	*Criteria*	IOA(-)	*Criteria*
Temperature (K)	DJF	-1.1		1.2		1.5		0.97	
	MAM	-0.8		0.9		1.2		0.97	
	JJA	-0.4	$\leq \pm 0.5$	0.7	≤ 2	1.0	-	0.98	≥ 0.8
	SON	-0.4		0.7		0.9		0.98	
	Annual	-0.7		0.8		1.0		0.98	
Wind speed (m s^{-1})	DJF	-0.1		1.2		1.6		0.84	
	MAM	-0.3		1.0		1.3		0.80	
	JJA	-0.3	$\leq \pm 0.5$	0.9	-	1.2	≤ 2	0.80	≥ 0.6
	SON	0.0		1.0		1.4		0.86	
	Annual	-0.2		1.0		1.3		0.83	
Wind direction (°)	DJF	8.9		17.7		28.6		0.70	
	MAM	5.4		16.1		24.4		0.73	
	JJA	11.4	$\leq \pm 10$	18.6	≤ 30	29.3	-	0.74	-
	SON	10.1		20.2		30.8		0.72	
	Annual	9.0		16.5		26.2		0.64	
Humidity (g kg^{-1})	DJF	-0.1		0.2		0.3		0.98	
	MAM	0.0		0.3		0.4		0.96	
	JJA	-0.1	$\leq \pm 1$	0.4	≤ 2	0.6	-	0.94	≥ 0.6
	SON	-0.2		0.3		0.4		0.97	
	Annual	-0.1		0.3		0.4		0.96	
Precipitation (mm)	DJF	-0.3		0.4		0.7		0.33	
	MAM	-0.3		0.3		1.0		-0.06	
	JJA	-0.4	-	0.5		1.3	-	0.03	-
	SON	-0.4		0.4		0.7		0.42	
	Annual	-0.4		0.4		0.8		0.09	

*The units in the brackets are only for MB, MGE and RMSE. IOA is unitless.

Table S3. Evaluation of the model performance for the chemical species. MB: mean bias; MGE: mean gross error; RMSE: root-mean-square error; MFB: mean fractional bias; MFE: mean fractional error; IOA: index of agreement. DJF: December-January-February, MAM: March-April-May, JJA: June-July-August, SON: September-October-November.

Species	Number of stations	Model	Season	MB*	MGE*	RMSE*	MFB (%)	MFE (%)
$PM_{2.5}$	565	Base	DJF	1.9	6.8	9.0	18	37
			MAM	2.9	5.2	6.8	19	28
			JJA	-0.2	2.7	3.6	0	26
			SON	2.1	5.3	6.7	17	32
			Annual	1.6	4.4	5.7	13	27
		New	DJF	3.8	7.4	9.5	26	40
			MAM	4.5	6.0	7.7	26	32
			JJA	0.5	2.8	3.7	5	26
			SON	3.9	6.0	7.5	26	35
			Annual	3.1	5.0	6.2	21	30
O_3	608	New	DJF	1.3	5.4	7.6	6	23
			MAM	1.6	5.1	6.9	5	14
			JJA	1.0	4.6	6.3	4	13
			SON	2.7	5.2	7.2	11	21
			Annual	1.9	4.7	6.5	7	15
NO_2	3036	New	DJF	-4.8	7.2	9.8	-35	55
			MAM	-4.4	6.6	9.2	-42	62
			JJA	-2.7	5.1	7.5	-33	61
			SON	-3.3	6.3	8.5	-27	53
			Annual	-3.8	6.1	8.5	-35	57
SO_2	1979	New	DJF	6.9	7.8	18.6	80	102
			MAM	5.5	6.1	13.8	72	97
			JJA	4.4	4.9	11.2	71	99
			SON	7.4	8.0	18.2	90	108
			Annual	5.9	6.5	14.8	79	100

* Units are ppb, except for $PM_{2.5}$ which is $\mu g\ m^{-3}$.

Table S4. Performance criteria and goals for model results on $PM_{2.5}$ and ozone (Boylan and Russell, 2006; EPA, 2007).

Species	Metric	Criteria	Goal
$PM_{2.5}$	MFB	$\leq \pm 60\%$	$\leq \pm 30\%$
	MFE	$\leq$ 75%	$\leq$ 50%
O_3	MFB	$\leq \pm 30\%$	$\leq \pm 15\%$
	MFE	$\leq$ 45%	$\leq$ 30%

Table S5. Seasonal statistical analysis of daily average organic aerosols at nine ACSM/AMS stations. MB: mean bias; MGE: mean gross error; RMSE: root-mean-square error; MFB: mean fractional bias; MFE: mean fractional error. Spring: March-April-May, summer: June-July-August, autumn: September-October-November, winter: December-January-February.

Season	Site	MB (μg m^{-3})		MGE (μg m^{-3})		RMSE (μg m^{-3})		MFB (%)		MFE (%)	
		BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW
					OA						
spring	MHD	-0.1	0.1	0.7	0.8	1.6	1.7	-86	-74	117	114
	MRS	-9.4	-7.6	9.4	7.6	9.7	8.0	-163	-116	163	116
	ZRH	-1.7	0.8	3.5	3.7	4.7	5.0	-34	8	64	53
	SMEARII	0.0	0.7	0.4	0.8	0.6	1.2	-10	58	54	70
summer	MHD	0.0	0.0	0.3	0.3	0.5	0.6	-91	-83	119	115
	MSA	-2.4	-2.0	2.6	2.3	3.2	2.8	-89	-73	97	83
	ZRH	-1.0	0.1	3.2	3.3	4.8	5.0	-21	3	65	59
autumn	BLQ	-8.4	0.5	8.7	5.5	10.8	7.0	-62	10	67	35
	FKL	-1.1	-0.1	1.5	1.4	2.2	1.9	-53	-17	70	58
	MHD	-0.1	-0.1	0.2	0.2	0.5	0.5	-90	-83	116	113
	MSA	-1.4	-0.6	1.9	1.7	2.4	2.2	-42	-12	72	59
	SIRTA	-7.6	-4.3	7.7	5.4	11.0	8.5	-108	-43	111	63
	SPC	-2.7	5.7	5.3	8.0	6.9	9.6	-14	52	59	68
	ZRH	-0.6	2.2	3.6	4.4	4.7	6.1	-16	17	54	53
winter	BLQ	-21.4	-16.2	21.4	16.4	23.4	19.0	-149	-98	149	99
	MHD	0.0	0.1	0.3	0.3	0.6	0.6	-4	7	91	89
	MRS	-7.8	-5.3	7.8	5.8	11.0	9.3	-109	-55	111	65
	MSA	0.1	0.5	0.6	0.9	0.9	1.3	-10	10	90	92
	SIRTA	-3.6	-1.7	3.6	2.8	5.5	4.4	-110	-31	117	71
	ZRH	-4.1	-0.5	4.6	3.7	5.9	4.9	-68	-3	82	55
					HOA						
spring	MRS	-1.4	-0.8	1.4	1.2	2.1	1.7	-107	-33	137	99
	ZRH	-0.3	0.4	0.4	0.7	0.6	0.9	-41	52	81	81
summer	MSA	-0.3	-0.2	0.3	0.2	0.3	0.3	-157	-102	158	111
	ZRH	-0.2	0.3	0.3	0.5	0.4	0.6	-29	61	80	83
autumn	BLQ	-1.6	0.2	1.7	1.4	2.6	1.9	-66	35	82	65
	MSA	-0.2	-0.2	0.2	0.2	0.3	0.2	-147	-84	149	92
	SIRTA	-0.7	0.1	0.8	0.8	1.4	1.3	-76	21	98	73
	SPC	-2.3	-1.1	2.3	1.4	2.7	1.8	-129	-42	130	58
	ZRH	-0.3	0.6	0.5	0.9	0.7	1.1	-26	65	81	86
winter	BLQ	-3.4	-2.2	3.4	2.6	4.9	4.1	-134	-53	137	82
	MRS	-1.2	-0.5	1.3	1.1	2.1	1.8	-88	-3	116	83
	SIRTA	-0.3	0.1	0.3	0.3	0.5	0.5	-70	30	101	80
	ZRH	-0.1	1.0	0.4	1.2	0.6	1.5	3	92	80	105
					BBOA						
spring	MRS	-3.8	-3.0	3.8	3.0	4.6	3.9	-164	-104	164	104
	ZRH	-0.4	0.4	0.6	0.8	0.8	1.2	-57	37	85	74
summer	ZRH	-0.4	-0.2	0.4	0.3	0.6	0.5	-108	-28	120	75
autumn	BLQ	-4.0	1.0	4.0	2.9	5.1	3.5	-83	23	86	46
	SIRTA	-2.0	0.0	2.1	1.9	3.4	2.8	-70	22	103	76
	SPC	-1.0	4.1	2.9	5.3	4.3	6.5	35	104	115	118
	ZRH	-0.8	0.2	0.8	0.8	1.2	1.2	-74	18	94	67

Season	Site	MB (μg m^{-3})		MGE (μg m^{-3})		RMSE (μg m^{-3})		MFB (%)		MFE (%)	
		BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW	BASE	NEW
winter	BLQ	-4.8	-1.6	4.8	3.2	6.0	4.2	-121	-29	121	59
	MRS	-3.6	-2.6	3.7	3.1	6.9	6.4	-118	-37	126	80
	SIRTA	-1.0	0.5	1.2	1.5	2.1	2.1	-46	46	100	85
	ZRH	-0.5	1.3	0.8	1.7	1.2	2.2	-21	71	82	96
					OOA						
spring	MRS	-3.9	-3.5	3.9	3.5	3.9	3.6	-158	-135	158	135
	ZRH	-0.3	0.6	2.9	3.0	3.8	4.0	-15	6	69	64
	SMEARII	-0.2	0.0	0.4	0.4	0.6	0.6	-57	-23	81	65
summer	MSA	-1.1	-0.9	1.5	1.3	1.9	1.7	-62	-48	82	72
	ZRH	0.4	0.8	2.5	2.5	3.5	3.6	5	16	64	62
autumn	BLQ	-0.8	1.4	2.3	2.9	2.9	3.5	-2	33	49	54
	FKL	-1.3	-0.7	1.6	1.3	2.3	1.9	-62	-37	75	61
	MSA	-1.0	-0.5	1.3	1.1	1.7	1.5	-43	-23	61	49
	SIRTA	-3.2	-2.6	3.2	2.9	4.8	4.4	-97	-72	104	86
	SPC	1.1	3.2	2.9	4.0	3.3	4.6	54	82	84	93
	ZRH	1.5	2.4	3.0	3.4	4.2	4.9	17	31	58	60
winter	BLQ	-9.3	-8.5	9.3	8.5	10.4	9.8	-144	-126	144	126
	MRS	-2.5	-1.8	2.6	2.2	3.4	2.9	-71	-43	84	65
	SIRTA	-1.3	-1.3	1.4	1.3	2.0	2.0	-138	-127	142	133
	ZRH	-3.8	-3.1	4.2	3.8	5.8	5.4	-78	-55	98	86

Table S6: Relative contributions (%) of different sources to the organic aerosol (OA) concentration on a country scale. DJF: December – January – February; JJA: June – July – August.

Country	Gasoline vehicles		Diesel vehicles		Biomass burning		Other anthropogenic		Biogenic	
	DJF	JJA	DJF	JJA	DJF	JJA	DJF	JJA	DJF	JJA
Albania	2.0	6.3	1.8	3.1	71.8	34.6	9.2	21.4	15.2	34.7
Austria	1.6	4.8	3.9	3.7	59.9	15.9	10.0	18.7	24.6	57.0
Belarus	0.9	2.2	2.1	1.7	65.7	21.1	8.4	11.5	22.8	63.5
Belgium	1.4	3.9	7.3	6.7	51.0	16.7	14.4	28.7	25.9	44.0
Bosnia and Herzegovina	1.3	5.5	1.4	3.2	79.8	35.4	6.4	19.8	11.2	36.1
Bulgaria	1.4	4.0	1.4	2.3	77.9	39.2	7.9	17.5	11.3	37.0
Croatia	1.9	6.1	2.4	3.7	70.8	28.8	7.6	21.1	17.2	40.3
Cyprus	1.3	2.5	2.5	1.5	35.6	13.5	6.1	8.9	54.6	73.6
Czech Republic	1.1	3.3	4.7	4.4	54.9	16.1	6.7	15.4	32.6	60.7
Denmark	0.6	3.5	2.3	2.9	64.2	20.1	8.2	25.7	24.6	47.9
Estonia	0.4	2.0	1.1	1.3	78.5	19.1	4.3	11.1	15.8	66.5
Finland	0.4	1.4	1.8	0.9	57.0	7.8	7.6	7.4	33.2	82.5
France	1.4	4.6	4.5	4.9	62.7	22.3	10.8	29.4	20.6	38.8
Germany	1.3	3.8	4.2	3.6	46.6	12.5	10.3	19.4	37.5	60.8
Greece	2.3	5.1	1.4	1.8	58.3	23.2	9.9	16.9	28.1	53.0
Hungary	1.5	4.7	2.4	3.8	74.5	30.7	8.1	20.3	13.6	40.5
Ireland	0.6	1.6	3.2	2.8	16.0	5.5	5.8	11.2	74.4	79.0
Italy	4.4	10.9	4.4	4.5	70.0	25.2	10.5	29.3	10.7	30.1
Latvia	0.4	2.2	1.4	1.7	80.1	23.3	4.3	12.2	13.9	60.6
Lithuania	0.6	2.6	2.8	2.5	69.3	22.2	7.5	14.9	19.8	57.8
Luxembourg	1.6	3.9	12.9	11.6	50.3	15.5	13.3	23.6	21.9	45.5
Northern Macedonia	1.5	4.9	1.2	2.5	73.8	36.9	7.6	18.2	15.8	37.5
Malta	5.4	11.7	3.2	6.1	57.3	20.2	19.6	45.7	14.5	16.3
Netherlands	1.6	3.9	7.1	5.5	44.4	11.6	14.5	28.3	32.4	50.7
Norway	0.4	1.4	1.2	1.1	59.3	9.8	9.8	11.3	29.3	76.5
Poland	0.9	3.1	5.4	4.6	56.9	17.0	8.9	17.1	27.8	58.2
Portugal	0.9	2.2	2.4	2.8	42.4	17.8	8.9	17.9	45.4	59.3
Republic of Moldova	1.4	3.7	1.3	2.2	78.7	44.3	9.5	19.5	9.1	30.3
Romania	1.1	3.7	1.0	2.2	82.2	42.5	6.7	17.2	9.1	34.3
Russia	0.7	1.4	1.7	1.0	62.9	13.9	8.8	8.0	25.8	75.7
Serbia and Montenegro	1.4	5.1	1.3	3.0	81.9	44.0	7.0	19.0	8.3	28.9
Slovakia	0.9	3.8	2.3	4.0	74.1	27.5	5.3	16.8	17.4	47.9
Slovenia	1.3	5.5	2.3	3.7	76.2	26.5	5.2	17.6	14.9	46.6
Spain	0.9	2.0	2.4	2.0	42.0	18.9	8.0	17.2	46.6	59.9
Sweden	0.5	1.6	1.5	1.0	34.3	7.0	8.8	9.7	55.0	80.6
Switzerland	2.9	6.9	4.6	3.9	56.4	17.5	15.8	23.4	20.3	48.3
Turkey	1.1	2.3	1.2	1.3	60.7	21.2	9.0	11.9	28.0	63.3
Ukraine	1.2	3.1	1.6	2.0	71.7	33.8	11.2	17.4	14.3	43.8
United Kingdom	0.8	2.0	3.7	3.0	28.1	7.5	11.5	20.7	56.0	66.7

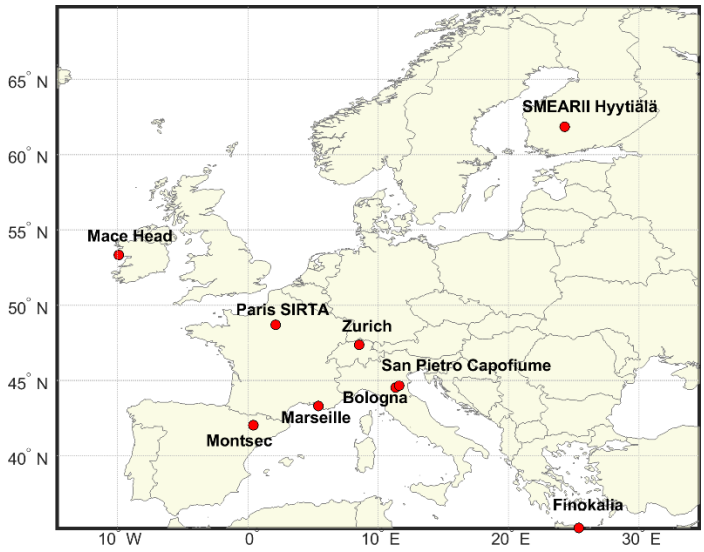

Figure S1: Model domain and spatial distribution of the ACSM/AMS stations.

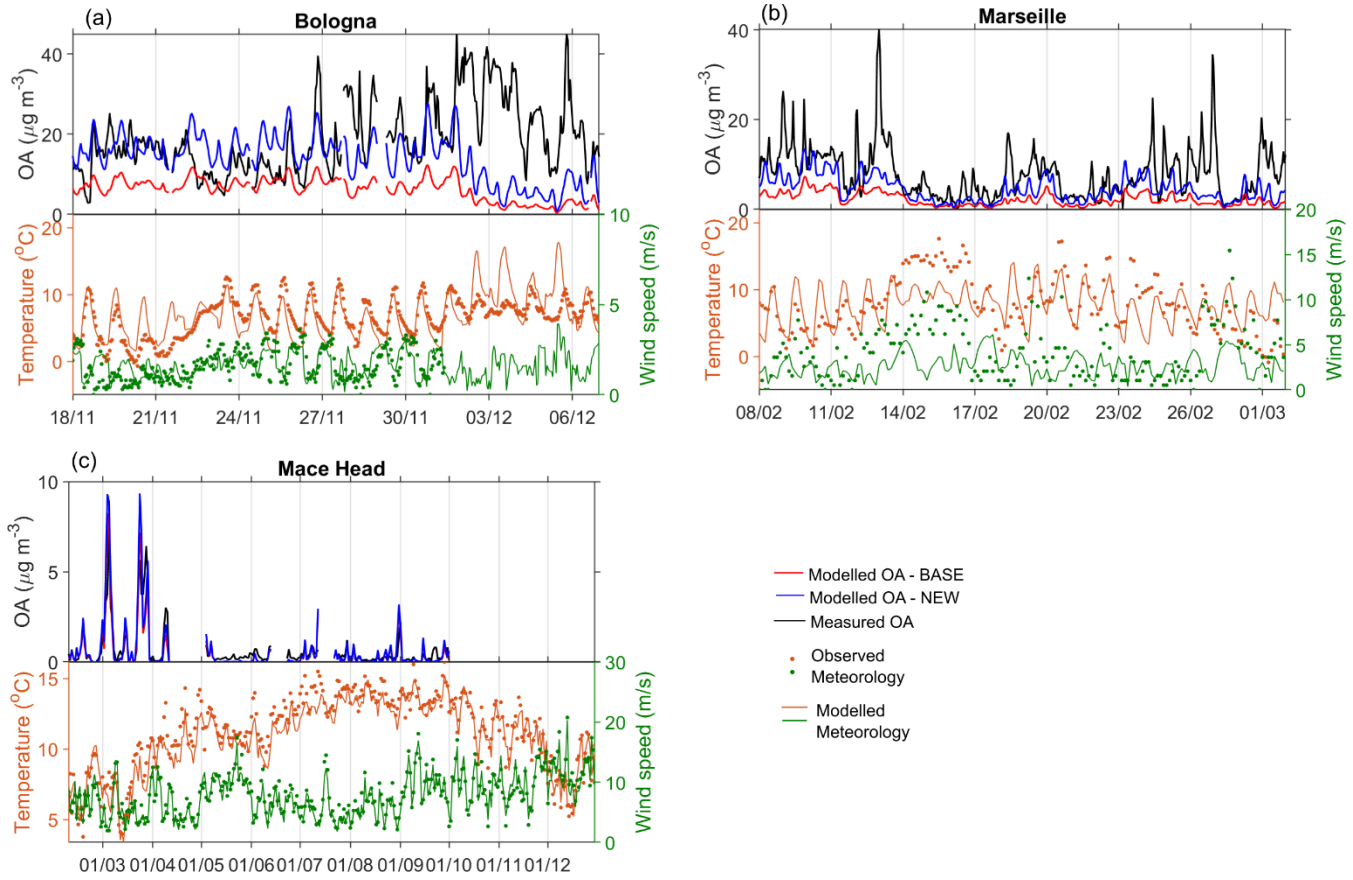

Figure S2: Temporal variations of modelled and measured organic aerosol concentrations together with some meteorological parameters available at Bologna, Marseille and Mace Head.

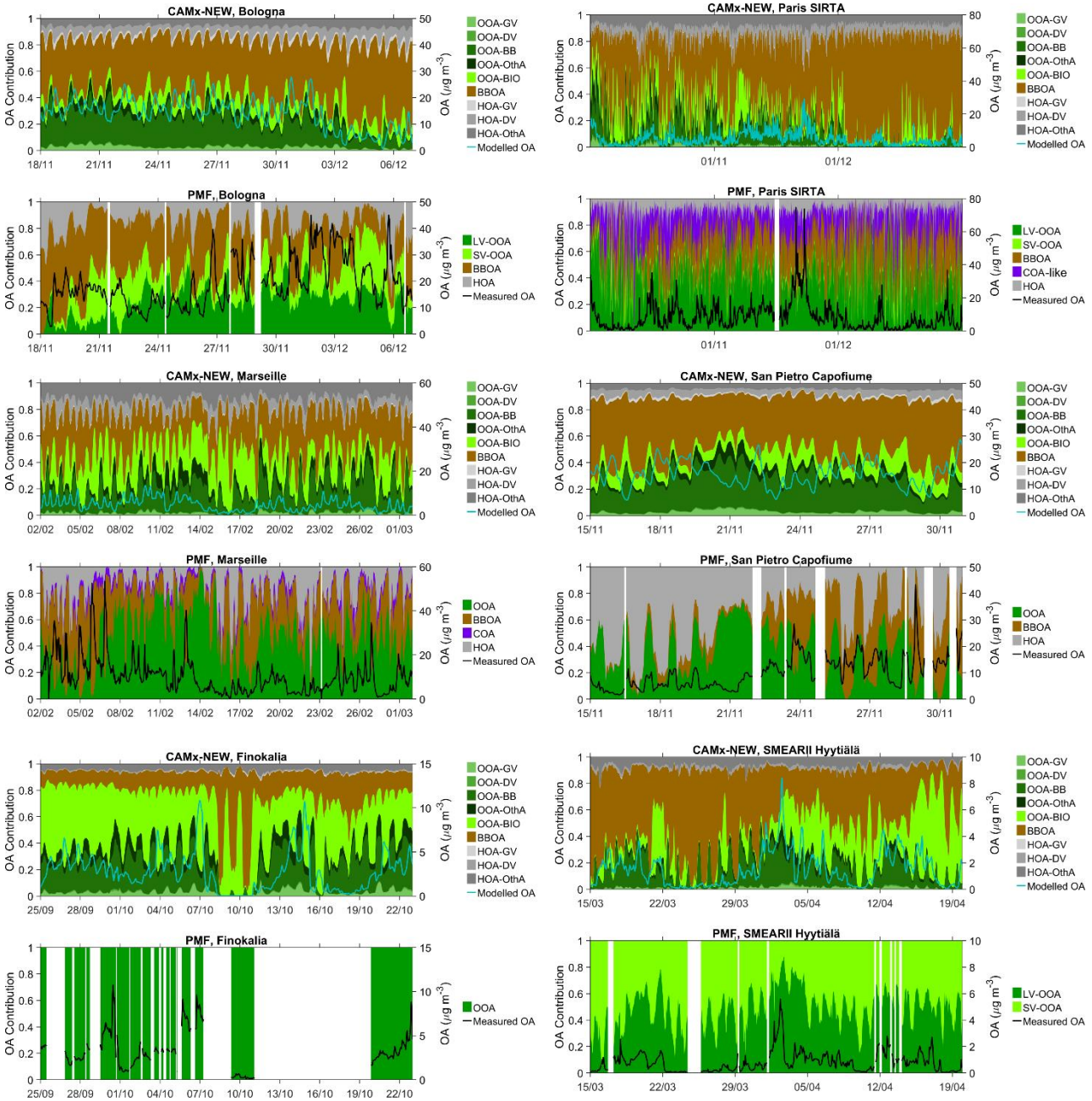

Figure S3: Comparison between modelled relative contribution of OA components and positive matrix factorization (PMF) analysis results. GV: Gasoline Vehicles; DV: Diesel Vehicles; BB: Biomass Burning; OthA: Other anthropogenic sources; BIO: Biogenic sources.

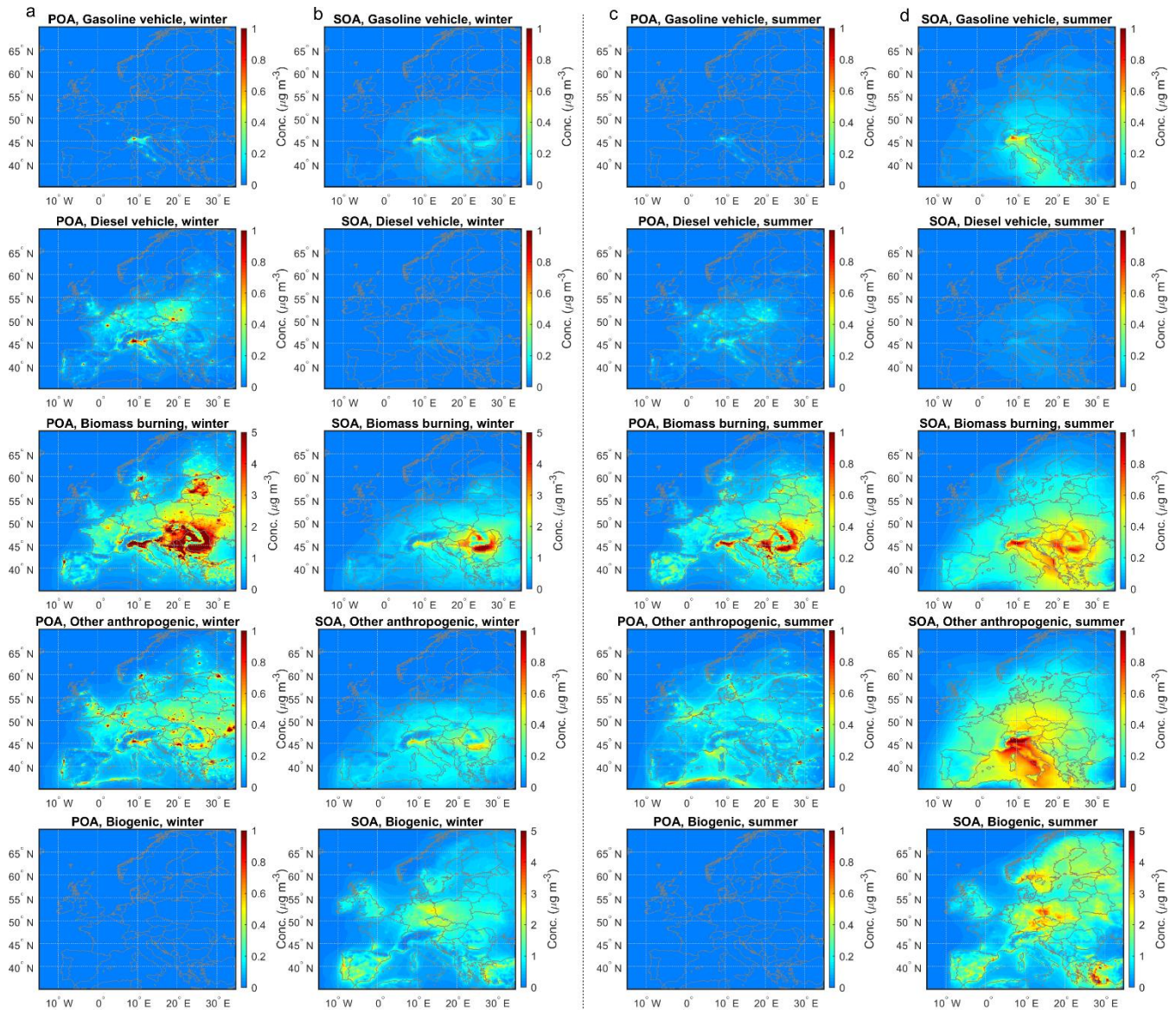

Figure S4: Spatial distributions of primary and secondary OA from different sources in winter (a, b) and summer (c, d). The winter and summer results are the averages of December – January – February and June – July – August, respectively. Note that different scales are used for biomass burning and biogenic source to facilitate visualization.

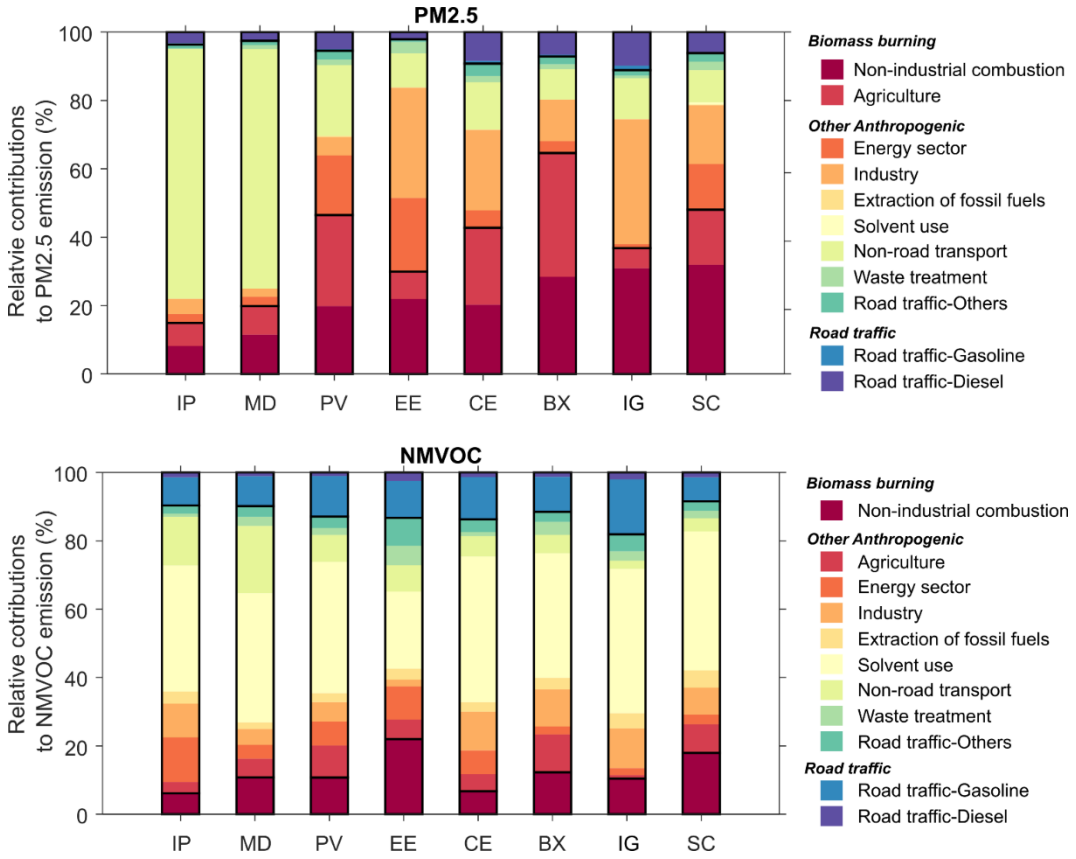

Figure S5: Relative contributions of different anthropogenic sources to total $PM_{2.5}$ and NMVOC emissions in 2011. The 8 sub-regions are the Iberian Peninsula (IP), the Mediterranean (MD), Po Valley (PV), eastern Europe (EE), central Europe (CE), Benelux (BX), Ireland and Great Britain (IG), and Scandinavia (SC).