Sources of organic aerosols in Europe: A modelling 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 in winter (February, number of stations = 1094) and summer (July, number of stations = 753). 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.

Meteorological parameters	MB			MGE			RMSE			IOA(-)		
	Feb.	Jul.	Criteria	Feb.	Jul.	Criteria	Feb.	Jul.	Criteria	Feb.	Jul.	Criteria
Temperature (°C)	-1.0	-0.1	≤ ±0.5	1.2	0.7	≤ ±2	1.7	0.9	-	1.0	1.0	≥ 0.8
Wind speed (m s^{-1})	-0.1	-0.3	≤ ±0.5	1.3	0.9	-	1.6	1.2	≤ 2	0.8	0.5	≥ 0.6
Wind direction (°)	5.2	15.8	≤ ±10	18.1	22.6	≤ 30	28.4	33.3	-	0.7	0.8	-
Humidity (g kg^{-1})	-0.1	0.0	≤ ±1	0.2	0.4	≤ 2	0.3	0.5	-	1.0	1.0	≥ 0.6
Precipitation (mm)	-0.3	-0.4	-	0.3	0.5	-	0.7	1.0	-	0.2	0.4	-

Table S3. Evaluation of the model performance for the chemical species in winter (February) and summer (July). MB: mean bias; MGE: mean gross error; RMSE: root-mean-square error; MFB: mean fractional bias; MFE: mean fractional error; IOA: index of agreement.

	Model	Number of Stations	MB[a]		MGE[a]		MFB (%)		MFE (%)		RMSE[a]		IOA(-)	
			Feb.	Jul.	Feb.	Jul.	Feb.	Jul.	Feb.	Jul.	Feb.	Jul.	Feb.	Jul.
$PM_{2.5}$	BASE	565	0.5	-1.3	8.9	2.8	9	-12	35	28	11.7	3.7	0.6	0.4
	NEW	565	2.9	-0.8	9.3	2.6	17	-7	36	26	11.9	3.6	0.6	0.5
O_3	NEW	608	2.0	0.9	6.1	4.7	8.8	4.1	23.0	13.5	8.5	6.3	0.7	0.7
NO_2	NEW	3036	-6.2	-2.9	8.1	5.1	-43	-36	58.6	63.2	10.9	7.5	0.6	0.5
SO_2	NEW	1979	6.7	3.9	7.7	4.5	77.0	65.5	98.5	98.2	17.9	10.2	0.1	0.1

[a] Units are ppb, except for $PM_{2.5}$ which is μg 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	≤ ±60%	≤ ±30%
	MFE	≤ 75%	≤ 50%
O_3	MFB	≤ ±30%	≤ ±15%
	MFE	≤ 45%	≤ 30%

Table S5: 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
The 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

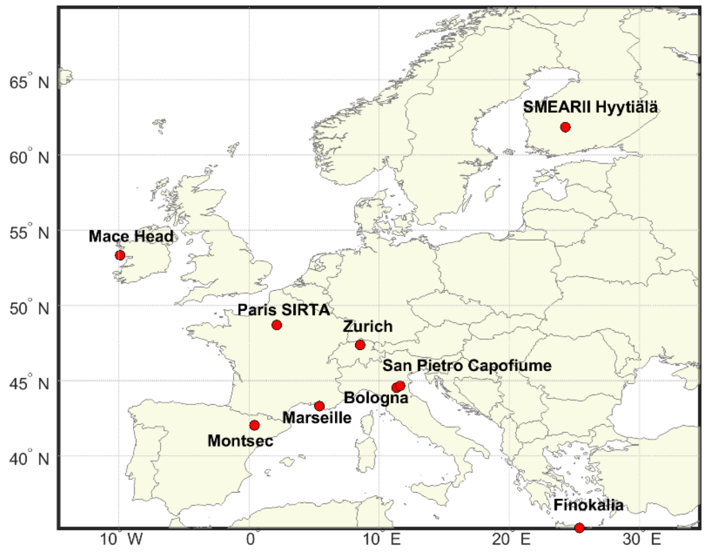

Figure S1: Model domain and spatial distribution of the ACSM/AMS stations.

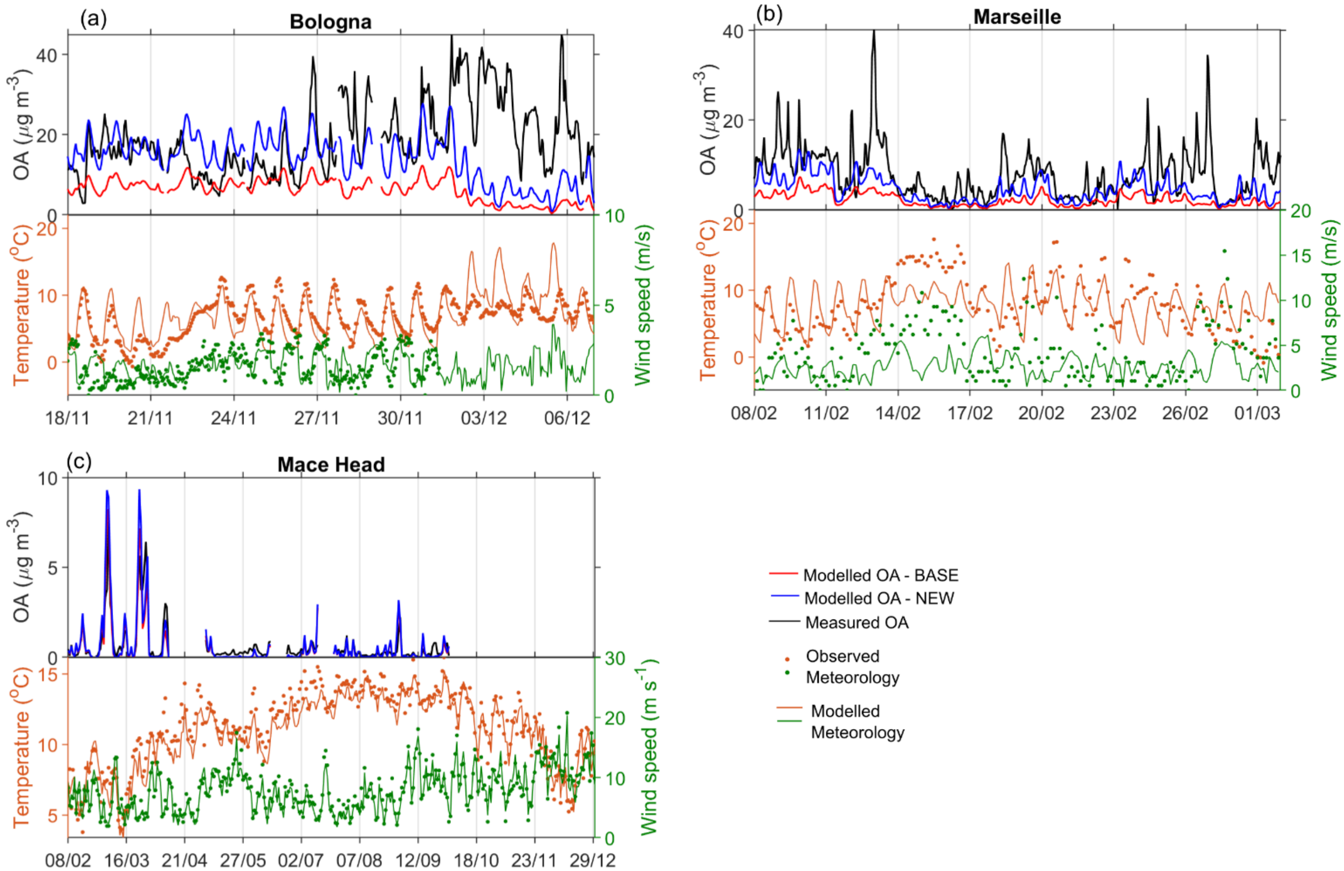

Figure S2: Temporal variations of modelled and measured organic aerosol concentrations together with some meteorological parameters available at Bologna, Marseille and Mace Head.

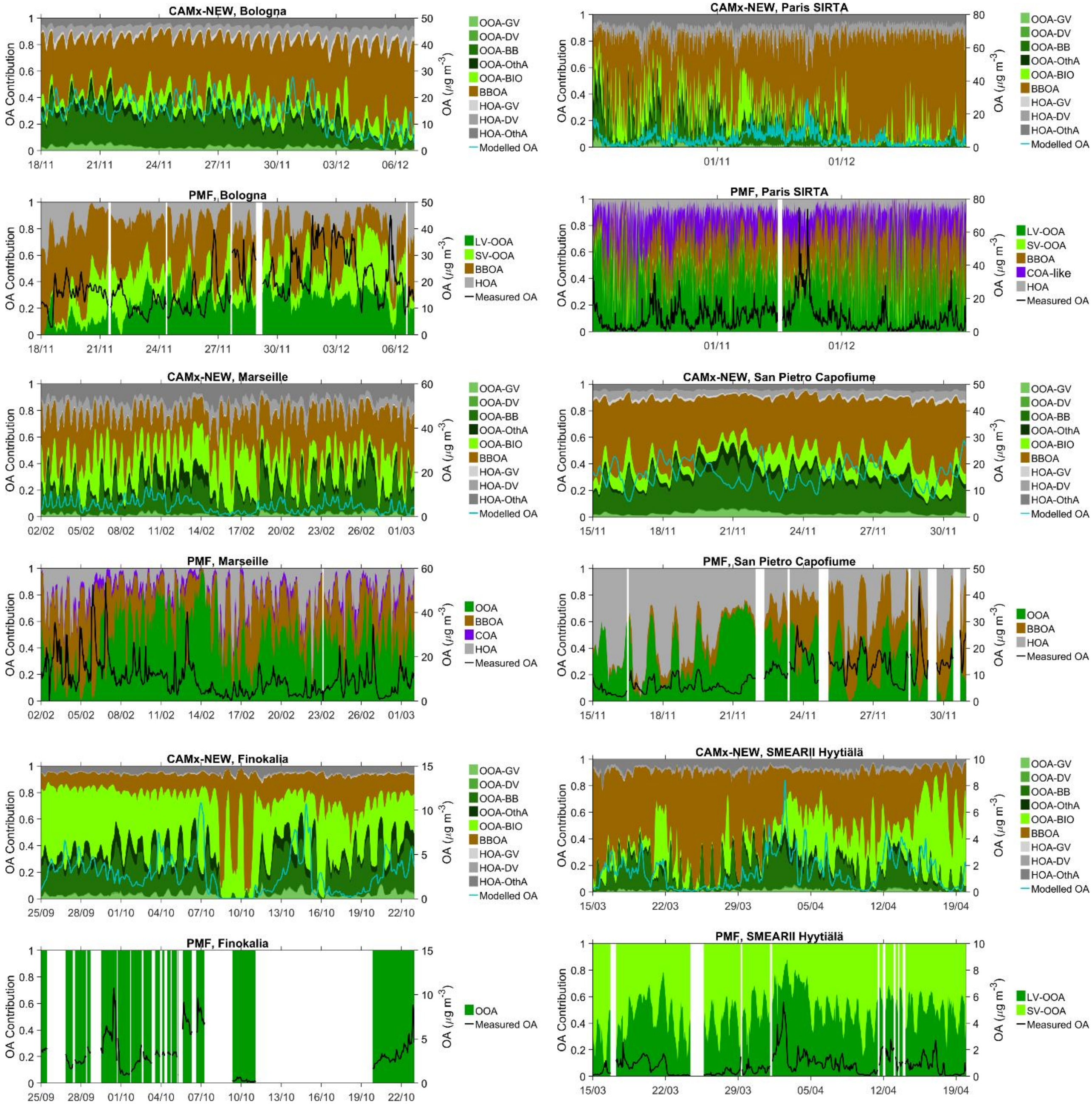

Figure S3: Comparison between modelled relative contribution of OA components and positive matrix factorization (PMF) analysis results. GV: Gasoline Vehicles; DV: Diesel Vehicles; BB: Biomass Burning; OthA: Other anthropogenic sources; BIO: Biogenic sources.

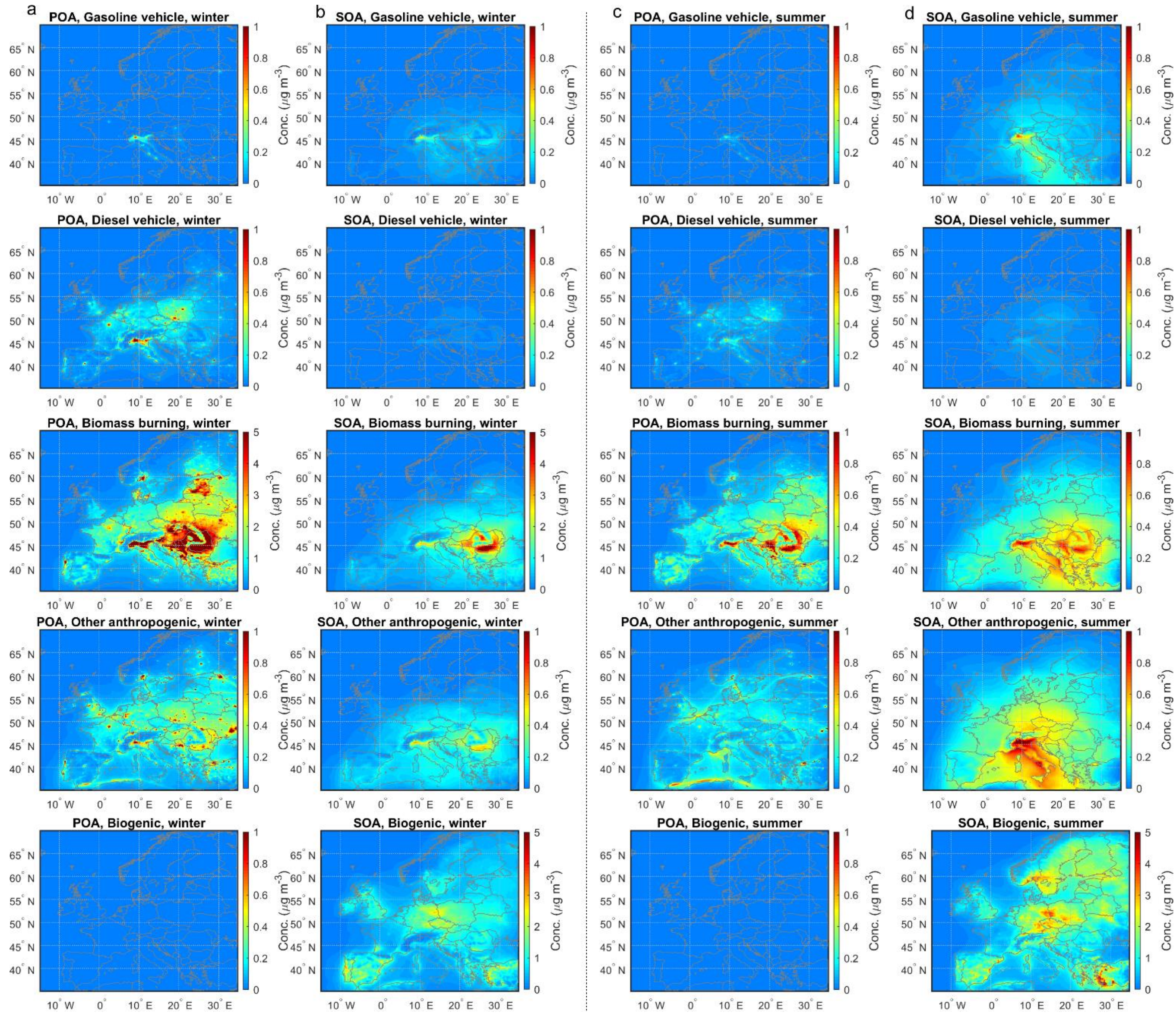

Figure S4: Spatial distributions of primary and secondary OA from different sources in winter (a, b) and summer (c, d). The winter and summer results are the averages of December – January – February and June – July – August, respectively. Note that different scales are used for biomass burning and biogenic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

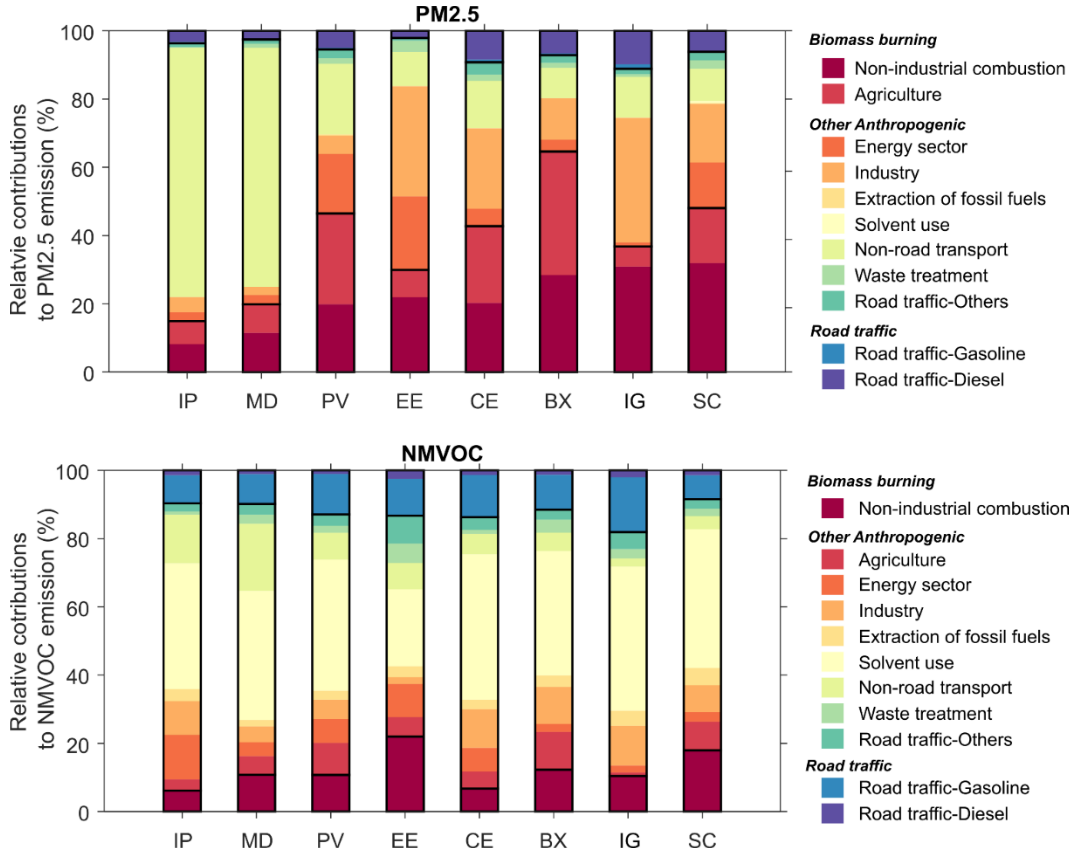

Figure S5: Relative contributions of different anthropogenic sources to total $PM_{2.5}$ and NMVOC emissions in 2011. The 8 sub-regions are the Iberian Peninsula (IP), the Mediterranean (MD), Po Valley (PV), eastern Europe (EE), central Europe (CE), Benelux (BX), Ireland and Great Britain (IG), and Scandinavia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