

**Supplementary material:**

**Modelling of organic aerosols over Europe (2002-2007) using a volatility basis set (VBS) framework: application of different assumptions regarding the formation of secondary organic aerosol**

R. Bergström, H.A.C. Denier van der Gon, A.S.H. Prévôt,  
K.E. Yttri and D. Simpson

## Appendix S1 Extra Tables

Table S1: Filter measurements of Total Carbon (TC) in PM<sub>10</sub>. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002–2007. Units  $\mu\text{g}(\text{C})\text{m}^{-3}$ . Correlation coefficients given in parentheses. Data illustrated in Fig. 7.

	N	Observed	NPNA	PAP	PAPA	PAA
CARBOSOL (September 2002–September 2004):						
Schauinsland	104	2.69	1.96 (-0.05)	2.06 (0.26)	2.27 (0.41)	2.52 (0.56)
Puy de Dome	86	1.74	1.69 (0.07)	1.73 (0.27)	1.88 (0.38)	2.05 (0.53)
EMEP EC/OC (July 2002–June 2003):						
Virolahti	51	2.44	1.60 (0.76)	2.06 (0.80)	2.15 (0.80)	2.41 (0.81)
Aspvreten	48	2.41	1.51 (0.69)	2.02 (0.70)	2.14 (0.71)	2.37 (0.74)
Birkenes(2002–2004)	267	1.19	1.19 (0.71)	1.36 (0.74)	1.43 (0.76)	1.56 (0.79)
Penicuik	50	2.04	1.47 (0.70)	1.51 (0.67)	1.61 (0.66)	1.67 (0.65)
Kollumerwaard	50	3.22	2.18 (0.57)	2.32 (0.48)	2.48 (0.47)	2.66 (0.47)
Mace Head	50	1.39	0.81 (0.92)	0.93 (0.91)	1.00 (0.89)	1.05 (0.85)
Langenbrügge	50	4.93	1.87 (0.63)	2.14 (0.62)	2.31 (0.60)	2.55 (0.59)
Gent	52	5.92	3.70 (0.68)	3.42 (0.73)	3.58 (0.74)	3.71 (0.72)
Kosetice	38	5.59	2.30 (0.52)	2.40 (0.54)	2.54 (0.54)	2.64 (0.54)
Stara Lesna	52	5.12	2.06 (0.62)	2.54 (0.80)	2.72 (0.80)	3.03 (0.79)
Illmitz	50	6.51	2.18 (0.60)	2.50 (0.61)	2.70 (0.57)	3.02 (0.50)
Ispra	45	9.62	2.59 (0.15)	2.55 (-0.01)	2.91 (-0.11)	3.15 (-0.17)
San Pietro Capofiume	50	7.35	2.28 (0.51)	2.48 (0.34)	2.78 (0.26)	3.20 (0.16)
Braganca	50	4.89	1.27 (0.21)	1.48 (0.17)	1.57 (0.16)	1.67 (0.15)
EMEP intensive PM measurement period summer 2006:						
Birkenes	30	1.15	0.87 (0.69)	1.03 (0.68)	1.15 (0.73)	1.32 (0.78)
Harwell <sup>(a)</sup>	17	1.33	1.57 (0.63)	1.61 (0.69)	1.85 (0.71)	1.96 (0.77)
Melpitz	31	4.38	1.39 (0.80)	1.55 (0.81)	1.81 (0.82)	2.38 (0.82)
Kosetice	21	2.81	1.38 (0.38)	1.56 (0.44)	1.85 (0.44)	2.55 (0.38)
Montelibretti	31	5.43	1.59 (0.78)	1.73 (0.73)	2.16 (0.74)	3.16 (0.73)
Montseny <sup>(b)</sup>	2	3.04	2.36 (-)	2.90 (-)	3.51 (-)	4.64 (-)
EMEP intensive PM measurement period winter 2007:						
Birkenes	34	0.66	0.73 (0.84)	0.75 (0.89)	0.75 (0.89)	0.75 (0.89)
Melpitz	33	2.36	1.21 (0.65)	1.12 (0.74)	1.13 (0.74)	1.13 (0.74)
Kosetice	29	2.39	1.64 (0.59)	1.47 (0.65)	1.47 (0.65)	1.47 (0.65)
Montelibretti	31	16.8	1.94 (0.75)	1.77 (0.61)	1.82 (0.57)	1.82 (0.56)

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TableS1–Continued

	N	Observed	NPNA	PAP	PAPA	PAA
Montseny <sup>(b)</sup>	4	1.54	3.10 (0.88)	2.83 (0.88)	2.96 (0.87)	2.97 (0.87)
<b>All Data</b>	1356	3.57	1.69 (0.46)	1.83 (0.39)	1.97 (0.39)	2.18 (0.38)
mean absolute error			2.21	2.16	2.08	1.99
<b>Summer Data</b> (May–Oct)	662	3.20	1.53 (0.65)	1.78 (0.57)	2.00 (0.60)	2.40 (0.63)
mean absolute error			1.78	1.70	1.56	1.37
<b>Winter Data</b> (Nov–Apr)	694	3.92	1.84 (0.38)	1.88 (0.33)	1.95 (0.33)	1.97 (0.32)
mean absolute error			2.61	2.60	2.59	2.59

Notes: N is number of samples. (a) Hourly observation data were averaged to daily means for calculation of  $r$  values. (b) Very few data, so  $r$ -values not calculated (N=2) or need to be interpreted with caution (N=4).

Table S2: Filter measurements of Total Carbon (TC) in PM<sub>2.5</sub>. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002–2007. Units  $\mu\text{g}(\text{C})\text{m}^{-3}$ . Correlation coefficients given in parentheses. Data illustrated in Fig. S10.

	N	Observed	VBS-NPNA	VBS-PAP	VBS-PAPA	VBS-PAA
CARBOSOL (July 2002–June 2004):						
K-Puszt	77	7.31	1.99 (0.70)	2.23 (0.56)	2.45 (0.51)	2.74 (0.35)
Aveiro	103	6.33	1.96 (0.33)	1.85 (0.16)	1.97 (0.14)	2.13 (0.10)
EMEP intensive PM measurement period summer 2006:						
Birkenes	30	0.92	0.84 (0.73)	1.00 (0.72)	1.12 (0.77)	1.29 (0.81)
Melpitz	31	4.10	1.23 (0.62)	1.39 (0.64)	1.66 (0.60)	2.23 (0.74)
Illmitz	29	2.72	1.59 (0.46)	1.67 (0.58)	2.00 (0.59)	2.89 (0.58)
Payerne	12	3.24	1.25 (0.59)	1.40 (0.54)	1.69 (0.58)	1.92 (0.64)
Ispra	23	4.14	2.06 (0.65)	2.23 (0.64)	2.79 (0.64)	3.46 (0.61)
Montelibretti	31	4.48	1.51 (0.77)	1.66 (0.71)	2.08 (0.69)	3.09 (0.70)
Montseny <sup>(a)</sup>	1	1.85	2.00 (-)	2.61 (-)	3.29 (-)	3.91 (-)
EMEP intensive PM measurement period winter 2007:						
Birkenes	30	0.46	0.67 (0.64)	0.65 (0.73)	0.65 (0.73)	0.65 (0.73)
Melpitz	33	1.29	1.13 (0.65)	1.04 (0.73)	1.04 (0.73)	1.04 (0.73)
Payerne	21	6.57	1.80 (0.66)	1.60 (0.69)	1.60 (0.69)	1.60 (0.69)
Ispra	28	20.6	2.53 (0.42)	2.03 (0.31)	2.03 (0.32)	2.03 (0.31)
Montelibretti	32	18.3	1.86 (0.71)	1.69 (0.56)	1.73 (0.52)	1.74 (0.52)
<b>All Data</b>	481	6.40	1.67 (0.51)	1.67 (0.32)	1.84 (0.26)	2.13 (0.16)
mean absolute error			4.83	4.86	4.72	4.57
<b>SummerData</b> (May–Oct)	243	3.76	1.59 (0.49)	1.76 (0.49)	2.06 (0.51)	2.61 (0.53)
mean absolute error			2.23	2.13	1.91	1.62
<b>WinterData</b> (Nov–Apr)	238	9.11	1.74 (0.62)	1.58 (0.53)	1.62 (0.51)	1.64 (0.50)
mean absolute error			7.49	7.64	7.59	7.58

Notes: N is number of samples. For a few stations hourly observation data were averaged to daily means for calculation of  $r$ -values. (a) Very few data, so  $r$ -values not calculated.



Table S3: Filter measurements of Organic Carbon (OC) in PM<sub>10</sub>. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002–2007. Unit:  $\mu\text{g}(\text{C})\text{m}^{-3}$ . Correlation coefficients given in parentheses.

	N	Observed	NPNA	PAP	PAPA	PAA
CARBOSOL (September 2002–September 2004):						
Schauinsland	104	2.40	1.54 (0.02)	1.64 (0.39)	1.84 (0.53)	2.09 (0.63)
Puy de Dome	86	1.52	1.36 (0.08)	1.41 (0.33)	1.55 (0.44)	1.72 (0.58)
EMEP EC/OC (July 2002–June 2003):						
Virolahti	51	2.08	1.27 (0.80)	1.73 (0.82)	1.82 (0.82)	2.08 (0.83)
Aspvreten	48	2.12	1.23 (0.72)	1.74 (0.74)	1.86 (0.76)	2.08 (0.79)
Birkenes(2002–2004)	267	1.07	1.00 (0.71)	1.18 (0.75)	1.25 (0.77)	1.38 (0.80)
Penicuik	50	1.53	1.04 (0.72)	1.08 (0.68)	1.17 (0.67)	1.24 (0.66)
Kollumerwaard	50	2.59	1.56 (0.51)	1.71 (0.42)	1.87 (0.42)	2.05 (0.44)
Mace Head	50	1.20	0.69 (0.92)	0.81 (0.90)	0.88 (0.87)	0.93 (0.81)
Langenbrügge	50	4.30	1.39 (0.62)	1.67 (0.60)	1.83 (0.58)	2.07 (0.57)
Gent	52	4.12	2.33 (0.68)	2.05 (0.76)	2.21 (0.76)	2.34 (0.73)
Kosetice	38	4.54	1.65 (0.52)	1.75 (0.54)	1.90 (0.53)	1.99 (0.53)
Stara Lesna	52	4.32	1.60 (0.67)	2.07 (0.82)	2.26 (0.82)	2.57 (0.80)
Illmitz	50	5.51	1.65 (0.58)	1.97 (0.58)	2.17 (0.53)	2.49 (0.44)
Ispra	45	7.79	1.79 (0.13)	1.75 (-0.08)	2.11 (-0.18)	2.35 (-0.24)
San Pietro Capofiume	50	5.91	1.61 (0.49)	1.80 (0.29)	2.11 (0.21)	2.52 (0.12)
Braganca	50	4.10	1.08 (0.19)	1.30 (0.15)	1.38 (0.15)	1.49 (0.14)
EMEP intensive PM measurement period summer 2006:						
Birkenes	30	1.03	0.77 (0.62)	0.93 (0.64)	1.05 (0.71)	1.22 (0.76)
Harwell <sup>(a)</sup>	17	0.83	1.05 (0.83)	1.09 (0.84)	1.33 (0.84)	1.44 (0.81)
Melpitz	31	2.55	1.05 (0.87)	1.21 (0.86)	1.48 (0.84)	2.05 (0.86)
Kosetice	21	2.47	1.09 (0.23)	1.27 (0.32)	1.55 (0.34)	2.25 (0.27)
Montelibretti	31	4.13	1.19 (0.66)	1.34 (0.60)	1.76 (0.62)	2.76 (0.63)
Montseny	11	2.19	1.46 (0.28)	1.72 (0.23)	2.23 (0.23)	3.60 (0.37)
EMEP intensive PM measurement period winter 2007:						
Birkenes	30	0.52	0.62 (0.41)	0.59 (0.56)	0.59 (0.56)	0.60 (0.56)
Melpitz	33	1.38	0.86 (0.47)	0.78 (0.53)	0.78 (0.52)	0.78 (0.52)
Kosetice	30	1.96	1.17 (0.52)	0.99 (0.56)	1.00 (0.56)	1.00 (0.56)
Montelibretti	31	15.5	1.33 (0.68)	1.16 (0.24)	1.20 (0.20)	1.21 (0.19)

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	N	Observed	NPNA	PAP	PAPA	PAA
<b>All Data</b>	1358	2.96	1.28 (0.39)	1.42 (0.32)	1.57 (0.32)	1.78 (0.31)
mean absolute error			1.91	1.86	1.78	1.70
<b>Summer Data</b> (May–Oct)	671	2.63	1.20 (0.63)	1.45 (0.56)	1.68 (0.60)	2.08 (0.63)
mean absolute error			1.51	1.43	1.29	1.13
<b>Winter Data</b> (Nov–Apr)	687	3.28	1.36 (0.31)	1.40 (0.24)	1.47 (0.24)	1.49 (0.23)
mean absolute error			2.30	2.29	2.26	2.27

Notes: N is number of samples. (a) Hourly observation data were averaged to daily means for calculation of  $r$  values.

Table S4: Filter measurements of Organic Carbon (OC) in PM<sub>2.5</sub>. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002–2007. Unit:  $\mu\text{g}(\text{C})\text{m}^{-3}$ .

	N	Observed	VBS-NPNA	VBS-PAP	VBS-PAPA	VBS-PAA
CARBOSOL (July 2002–June 2004):						
K-Pusztta	77	6.17	1.48 (0.66)	1.72 (0.46)	1.94 (0.42)	2.24 (0.26)
Aveiro	103	5.33	1.49 (0.31)	1.38 (0.12)	1.49 (0.10)	1.65 (0.06)
EMEP intensive PM measurement period summer 2006:						
Birkenes	30	0.84	0.75 (0.66)	0.91 (0.69)	1.03 (0.76)	1.20 (0.81)
Melpitz	31	1.20	0.98 (0.51)	1.14 (0.53)	1.40 (0.47)	1.97 (0.63)
Illmitz	29	2.35	1.28 (0.51)	1.36 (0.62)	1.69 (0.64)	2.58 (0.61)
Payerne	12	2.67	1.01 (0.62)	1.16 (0.58)	1.45 (0.60)	1.68 (0.66)
Ispra	22	3.34	1.45 (0.64)	1.62 (0.61)	2.16 (0.63)	2.84 (0.60)
Montelibretti	31	3.33	1.14 (0.68)	1.29 (0.60)	1.71 (0.58)	2.71 (0.61)
Montseny <sup>(a)</sup>	1	1.77	1.34 (-)	1.95 (-)	2.63 (-)	3.25 (-)
EMEP intensive PM measurement period winter 2007:						
Birkenes	30	0.40	0.62 (0.41)	0.59 (0.55)	0.59 (0.55)	0.59 (0.54)
Birkenes	34	0.47	0.66 (0.80)	0.68 (0.87)	0.68 (0.87)	0.68 (0.87)
Melpitz	33	0.69	0.83 (0.47)	0.74 (0.58)	0.74 (0.59)	0.74 (0.58)
Payerne	21	5.15	1.32 (0.59)	1.11 (0.55)	1.12 (0.54)	1.12 (0.54)
Ispra	28	16.3	1.58 (0.45)	1.08 (0.19)	1.08 (0.20)	1.08 (0.20)
Montelibretti	32	17.2	1.28 (0.63)	1.11 (0.19)	1.15 (0.16)	1.16 (0.15)
<b>All Data</b>	480	5.24	1.24 (0.39)	1.25 (0.15)	1.42 (0.10)	1.70 (0.01)
mean absolute error			4.12	4.14	4.03	3.95
<b>SummerData</b> (May–Oct)	242	2.83	1.24 (0.51)	1.41 (0.51)	1.70 (0.53)	2.25 (0.51)
mean absolute error			1.69	1.59	1.41	1.28
<b>WinterData</b> (Nov–Apr)	238	7.69	1.25 (0.53)	1.09 (0.33)	1.13 (0.31)	1.14 (0.30)
mean absolute error			6.59	6.73	6.69	6.67

Notes: N is number of samples. For a few stations hourly observation data were averaged to daily means for the calculation of  $r$ -values. (a) Only one sample, so  $r$ -values not calculated.

Table S5: Source apportionment studies, summer. Comparison of model results to observation-derived values for TC and different source categories of OC and EC (units  $\mu\text{g}(\text{C})\text{m}^{-3}$ ). The observed values are based on a statistical approach (Latin-hypercube sampling) and given as 5-95th (CARBOSOL) or 10-90th (SORGA and Göte) percentiles where this range is available from the publications.

		Observed	NPNA	PAP	PAPA	PAA
Hurdal (SORGA) (Rural, PM <sub>1</sub> , model PM <sub>2.5</sub> )	TC	1.67	1.22	1.27	1.41	1.78
	OC <sub>wood</sub>	0.06-0.09	0.20	0.049	0.051	0.054
	OC <sub>onf</sub>	1.14-1.27	0.66	0.81	0.82	1.22
	OC <sub>f</sub>	0.11-0.26	0.19	0.24	0.37	0.33
	OC <sub>POA</sub>	0.04-0.14	0.16	0.037	0.038	0.041
	OC <sub>fASOA</sub>	0.01-0.18	0.032	0.20	0.33	0.29
	EC <sub>wood</sub>	0.02-0.04		0.045		
	EC <sub>f</sub>	0.11-0.24		0.13		
Oslo (SORGA) (Urban, PM <sub>1</sub> , model, PM <sub>2.5</sub> )	TC	2.23	1.53	1.41	1.56	1.95
	OC <sub>wood</sub>	0.23-0.38	0.27	0.067	0.069	0.073
	OC <sub>onf</sub>	0.79-1.04	0.66	0.80	0.82	1.24
	OC <sub>f</sub>	0.46-0.74	0.31	0.26	0.39	0.36
	OC <sub>POA</sub>	0.07-0.35	0.28	0.050	0.052	0.055
	OC <sub>fASOA</sub>	0.16-0.65	0.034	0.21	0.34	0.30
	EC <sub>wood</sub>	0.09-0.16		0.089		
	EC <sub>f</sub>	0.14-0.40		0.19		
Gothenburg (Göte) (Urban, PM <sub>2.5</sub> )	TC	2.7±0.4	2.10	2.12	2.34	2.78
	OC <sub>wood</sub>	0.13-0.28	0.13	0.026	0.026	0.028
	OC <sub>onf</sub>	0.99-1.36	0.62	0.85	0.86	1.36
	OC <sub>f</sub>	0.65-1.02	0.59	0.50	0.69	0.64
	EC <sub>wood</sub>	0.02-0.07		0.025		
	EC <sub>f</sub>	0.31-0.47		0.72		
Aveiro (CARBOSOL) (Rural, PM <sub>2.5</sub> )	TC	4.0	2.32	2.40	2.59	2.94
	OC <sub>wood</sub>	0.23-0.44	0.76	0.17	0.17	0.18
	OC <sub>onf</sub>	~2.1-2.7	0.72	1.39	1.40	1.79
	OC <sub>f</sub>	~0.7	0.35	0.35	0.51	0.47
	OC <sub>POA</sub>	0.19-0.55	0.29	0.056	0.057	0.060
	OC <sub>fASOA</sub>	0.07-0.77	0.057	0.29	0.46	0.41
	EC <sub>wood</sub>	0.02-0.20		0.10		
	EC <sub>f</sub>	0.32-0.65		0.40		
K-Pusztá (CARBOSOL) (Rural, PM <sub>2.5</sub> )	TC	5.0	1.78	2.50	2.87	3.63
	OC <sub>wood</sub>	0.27-0.52	0.34	0.12	0.13	0.13
	OC <sub>onf</sub>	~2.7-4.0	0.77	1.55	1.59	2.42
	OC <sub>f</sub>	~0.62	0.33	0.49	0.81	0.73
	OC <sub>POA</sub>	0.15-0.49	0.26	0.063	0.067	0.071
	OC <sub>fASOA</sub>	0.05-0.77	0.072	0.43	0.75	0.66
	EC <sub>wood</sub>	0.02-0.24		0.060		
	EC <sub>f</sub>	0.25-0.55		0.28		

Notes: Observed single values preceded by ~ are "best estimates" from the respective publication and OC<sub>onf</sub> ranges preceded by ~ are estimated 5-95th percentiles based on the corresponding values for OC<sub>BSOA</sub> and OC<sub>PBAP</sub> from the references. For SORGA and Göte the comparisons are for the actual time periods for which measurements were performed. For CARBOSOL model results for the complete months given in Gelencsér et al. (2007) were used, due to lack of information of exact measurement periods.

Table S6: Source apportionment studies, winter. For notation, see Table S5.

		Observed	NPNA	PAP	PAPA	PAA
Hurdal (SORGA) (Rural, PM <sub>1</sub> , model PM <sub>2.5</sub> )	TC	1.05	2.43	2.53	2.53	2.54
	OC <sub>wood</sub>	0.29-0.44	1.03	0.94	0.94	0.94
	OC <sub>onf</sub>	0.05-0.27	0.68	0.75	0.75	0.76
	OC <sub>f</sub>	0.20-0.36	0.14	0.25	0.25	0.25
	OC <sub>POA</sub>	0.01-0.13	0.11	0.19	0.19	0.19
	OC <sub>fASOA</sub>	0.09-0.34	0.022	0.053	0.057	0.055
	EC <sub>wood</sub>	0.11-0.18		0.44		
	EC <sub>f</sub>	0.02-0.17		0.14		
Oslo (SORGA) (Urban, PM <sub>1</sub> , model PM <sub>2.5</sub> )	TC	2.63	7.58	6.49	6.50	6.51
	OC <sub>wood</sub>	0.28-0.44	4.29	3.13	3.13	3.13
	OC <sub>onf</sub>	0.06-0.51	0.81	0.91	0.91	0.93
	OC <sub>f</sub>	0.53-1.01	0.52	0.50	0.50	0.50
	OC <sub>POA</sub>	0.05-0.42	0.48	0.41	0.41	0.41
	OC <sub>fASOA</sub>	0.17-0.94	0.044	0.086	0.089	0.088
	EC <sub>wood</sub>	0.28-0.44		1.59		
	EC <sub>f</sub>	0.10-0.57		0.36		
Gothenburg (Göte) (Urban, PM <sub>10</sub> )	TC	3.0±0.7	1.28	1.23	1.24	1.25
	OC <sub>wood</sub>	0.42-0.75	0.20	0.14	0.14	0.14
	OC <sub>onf</sub>	0.36-0.91	0.55	0.59	0.59	0.61
	OC <sub>f</sub>	0.67-1.01	0.22	0.18	0.20	0.19
	EC <sub>wood</sub>	0.06-0.12		0.075		
	EC <sub>f</sub>	0.63-0.97		0.23		
Råö (Göte) (Rural, PM <sub>2.5</sub> )	TC	1.8±0.1	1.38	1.40	1.43	1.44
	OC <sub>wood</sub>	0.34-0.77	0.22	0.18	0.18	0.18
	OC <sub>onf</sub>	0.07-0.55	0.59	0.65	0.65	0.68
	OC <sub>f</sub>	0.41-0.63	0.24	0.24	0.26	0.25
	EC <sub>wood</sub>	0.10-0.18		0.085		
	EC <sub>f</sub>	0.23-0.35		0.25		
Aveiro (CARBOSOL) (Rural, PM <sub>2.5</sub> )	TC	14.1	1.88	1.37	1.38	1.38
	OC <sub>wood</sub>	7.4-9.8	0.66	0.16	0.16	0.16
	OC <sub>onf</sub>	~0.2-3.5	0.55	0.61	0.61	0.62
	OC <sub>f</sub>	~2.4	0.19	0.11	0.12	0.12
	OC <sub>POA</sub>	0.09-1.1	0.17	0.056	0.056	0.056
	OC <sub>fASOA</sub>	0.2-2.8	0.017	0.053	0.069	0.060
	EC <sub>wood</sub>	0.56-1.69		0.22		
	EC <sub>f</sub>	0.14-1.41		0.26		
K-Pusztá (CARBOSOL) (Rural, PM <sub>2.5</sub> )	TC	10.7	2.44	2.14	2.19	2.21
	OC <sub>wood</sub>	3.6-5.9	0.73	0.26	0.27	0.26
	OC <sub>onf</sub>	~0.3-3.6	0.58	0.77	0.77	0.80
	OC <sub>f</sub>	~2.2	0.43	0.42	0.46	0.44
	OC <sub>POA</sub>	0.2-1.3	0.38	0.22	0.22	0.22
	OC <sub>fASOA</sub>	0.3-3.0	0.052	0.19	0.24	0.22
	EC <sub>wood</sub>	0.29-1.39		0.24		
	EC <sub>f</sub>	0.28-1.71		0.46		

## Appendix S2 Extra Figures

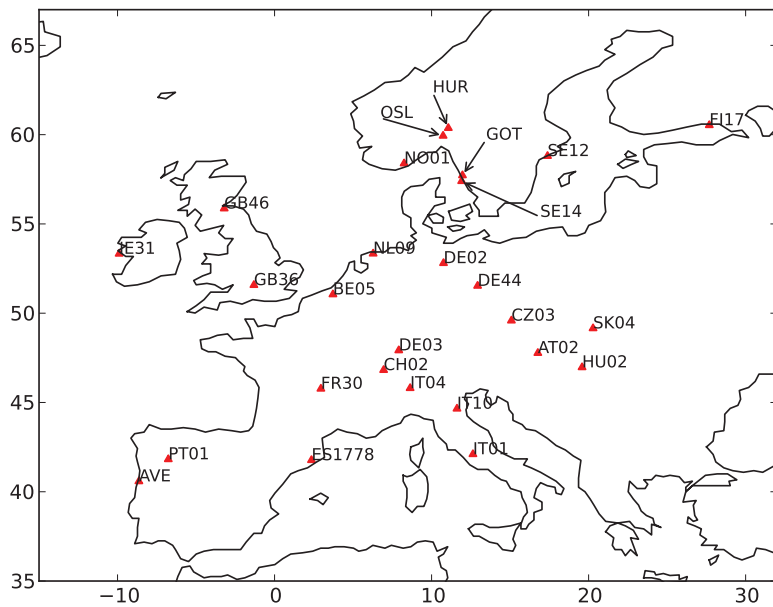


Fig. S1: Measurement sites used in this study. For more information see Table 2.

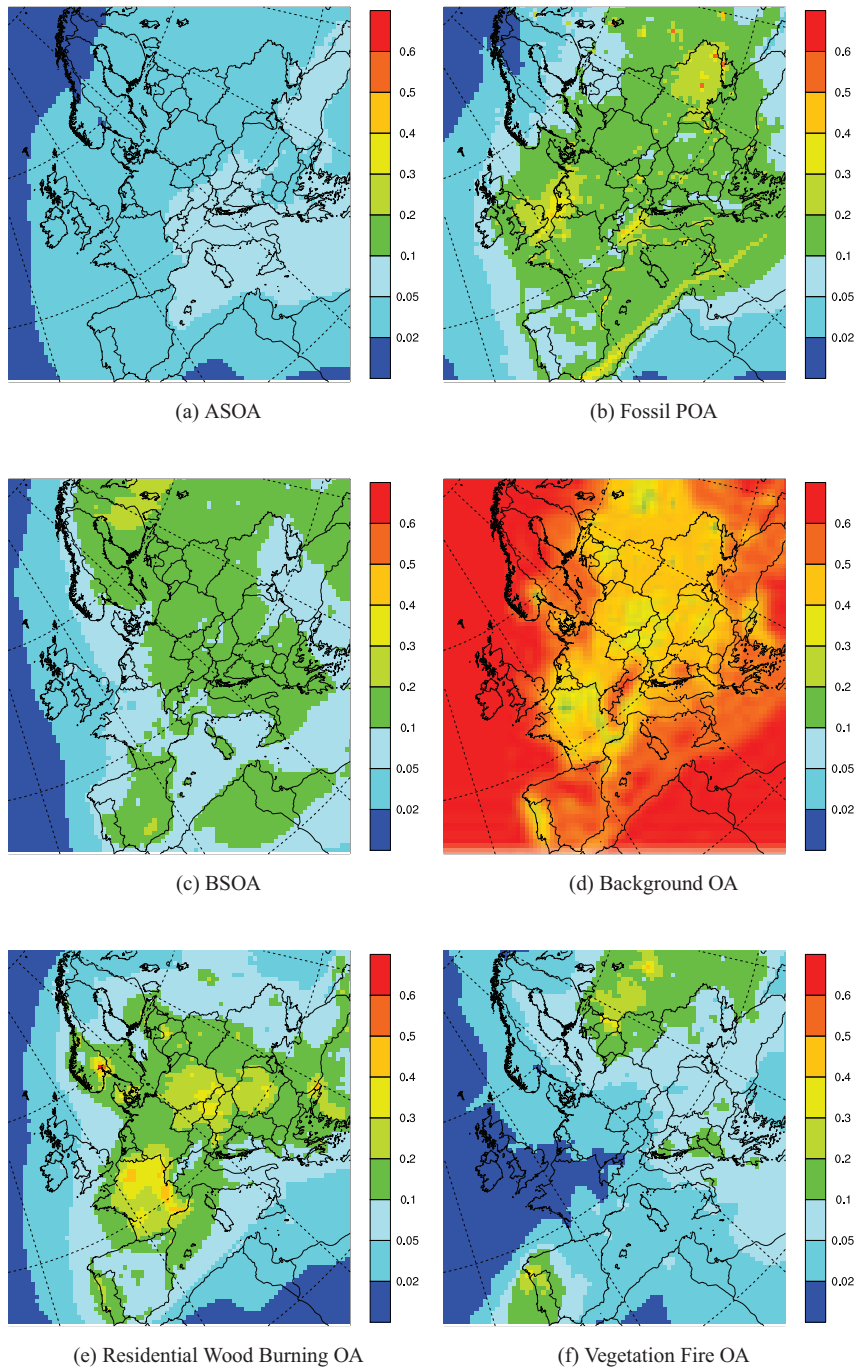


Fig. S2: Calculated relative contribution to total particulate OM in PM<sub>2.5</sub> from different sources, using the model version NPNA. Fraction of OM<sub>PM<sub>2.5</sub></sub> from (a) anthropogenic SOA (from AVOC), (b) fossil fuel primary OA (POA), (c) biogenic SOA (from BVOC), (d) background organic aerosol (from sources not explicitly included in the model), (e) residential wood combustion, (f) vegetation fires (open burning of biomass). Average for the 6-yr period 2002–2007.

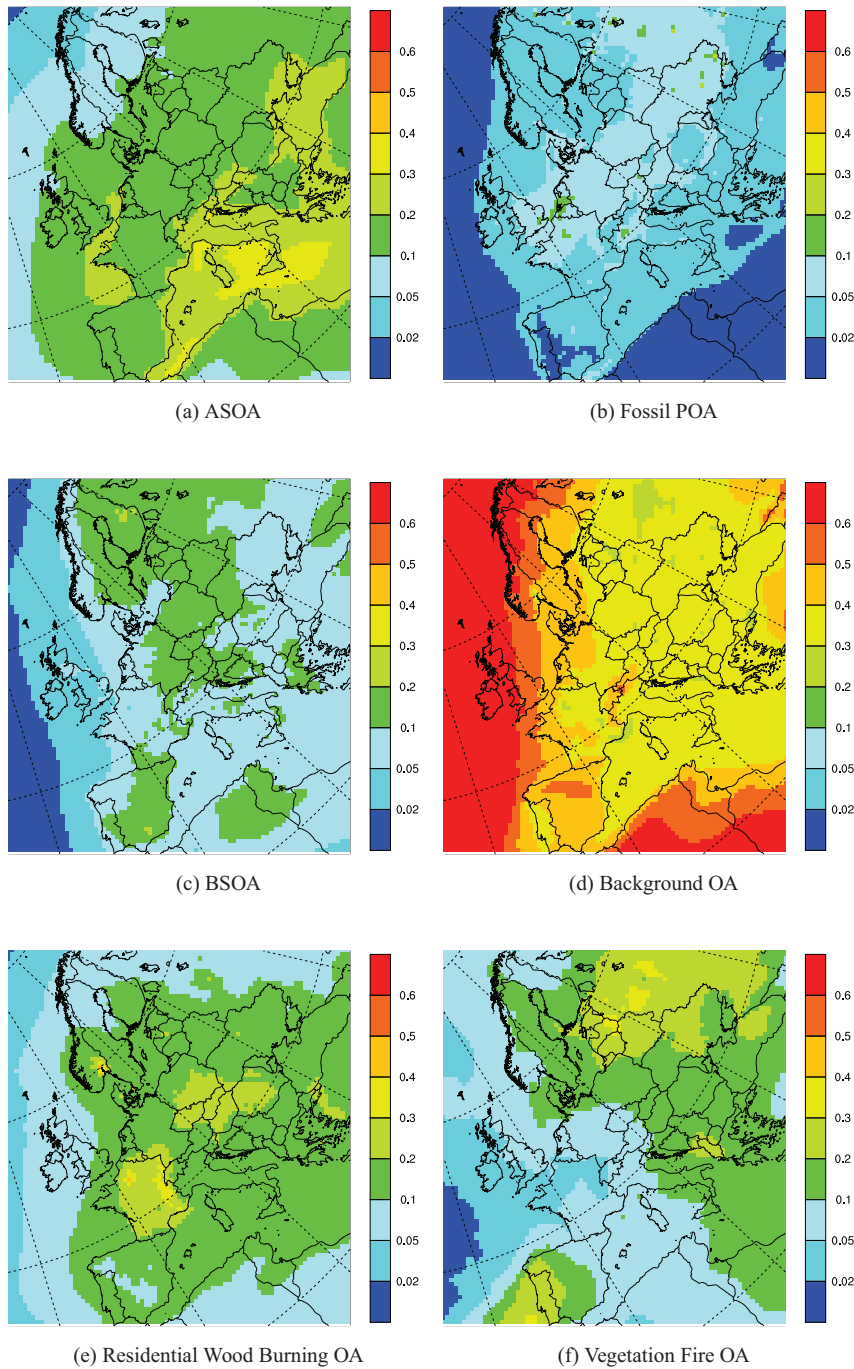


Fig. S3: Calculated relative contribution to total particulate OM in  $PM_{2.5}$  from different sources, using the model version PAP. Fraction of  $OM_{PM_{2.5}}$  from (a) anthropogenic SOA (from AVOC and fossil fuel S/IVOC), (b) fossil fuel primary OA (POA), (c) biogenic SOA (from BVOC), (d) background organic aerosol (from sources not explicitly included in the model), (e) residential wood combustion (primary +  $SOA^{SI}$ ), (f) vegetation fires (primary +  $SOA^{SI}$ ). Average for the 6-yr period 2002–2007.



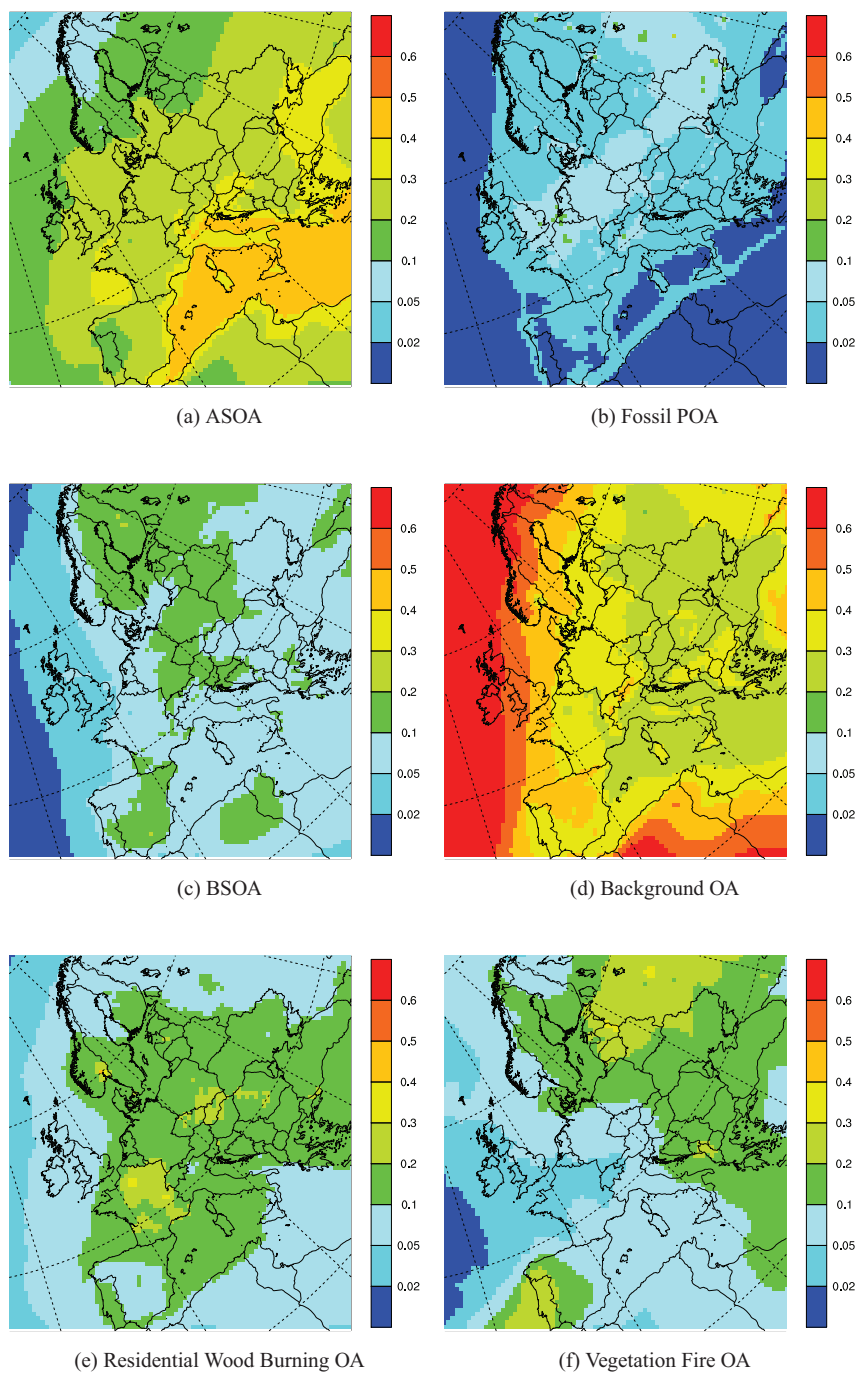


Fig. S4: Calculated relative contribution to total particulate OM in  $PM_{2.5}$  from different sources, using the model version PAPA. Fraction of  $OM_{PM_{2.5}}$  from (a) anthropogenic SOA (from AVOC and fossil fuel S/IVOC), (b) fossil fuel primary OA (POA), (c) biogenic SOA (from BVOC), (d) background organic aerosol (from sources not explicitly included in the model), (e) residential wood combustion (primary +  $SOA^{SI}$ ), (f) vegetation fires (primary +  $SOA^{SI}$ ). Average for the 6-yr period 2002–2007.

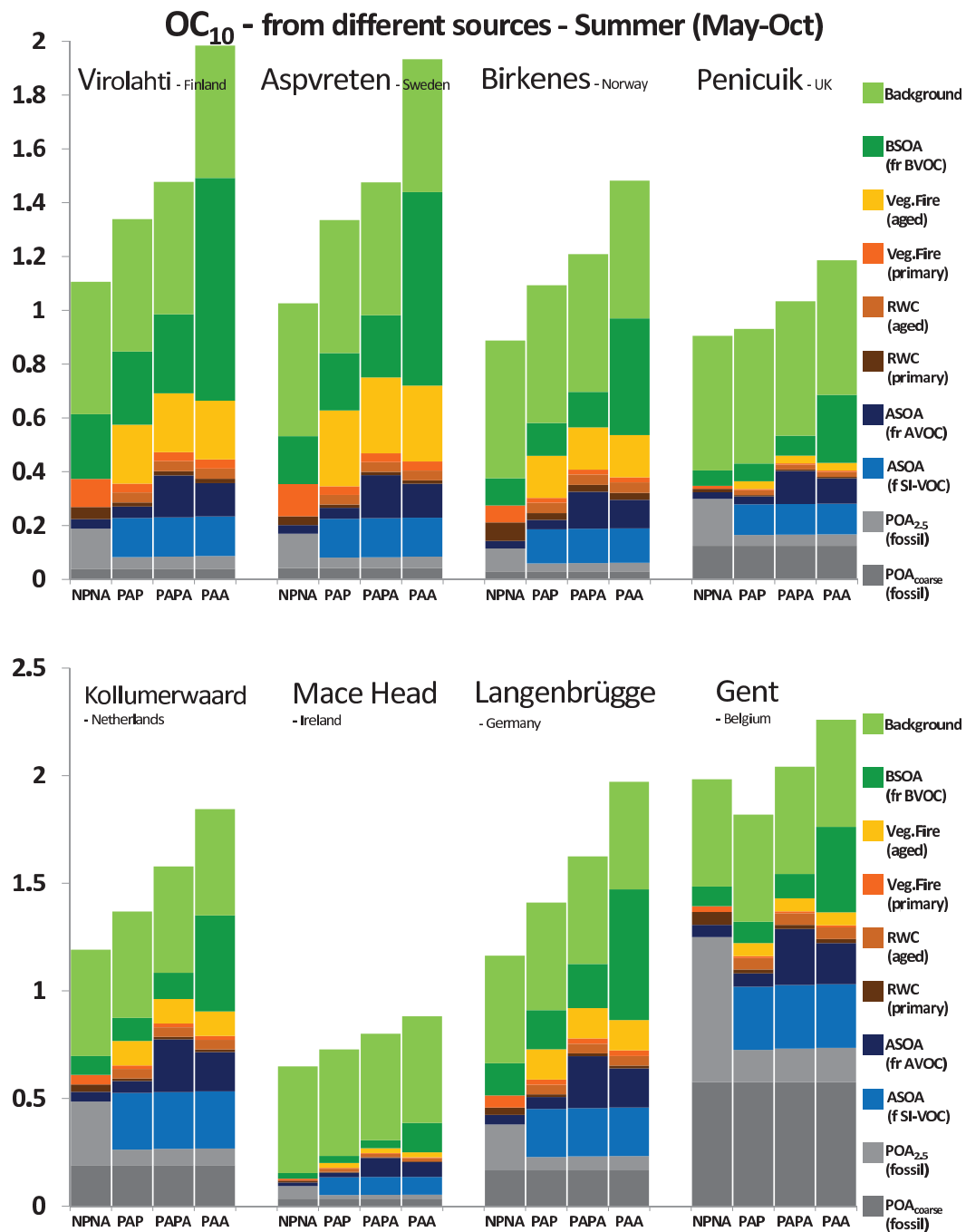


Fig. S5: (a) Modelled contribution from different sources to OC<sub>PM10</sub> during the months May–October (summer half-years) 2002–2004, at selected sites, arranged from north to south. Continued with Fig. S5(b). Colours/Notation: see legend and Fig. 5(a).

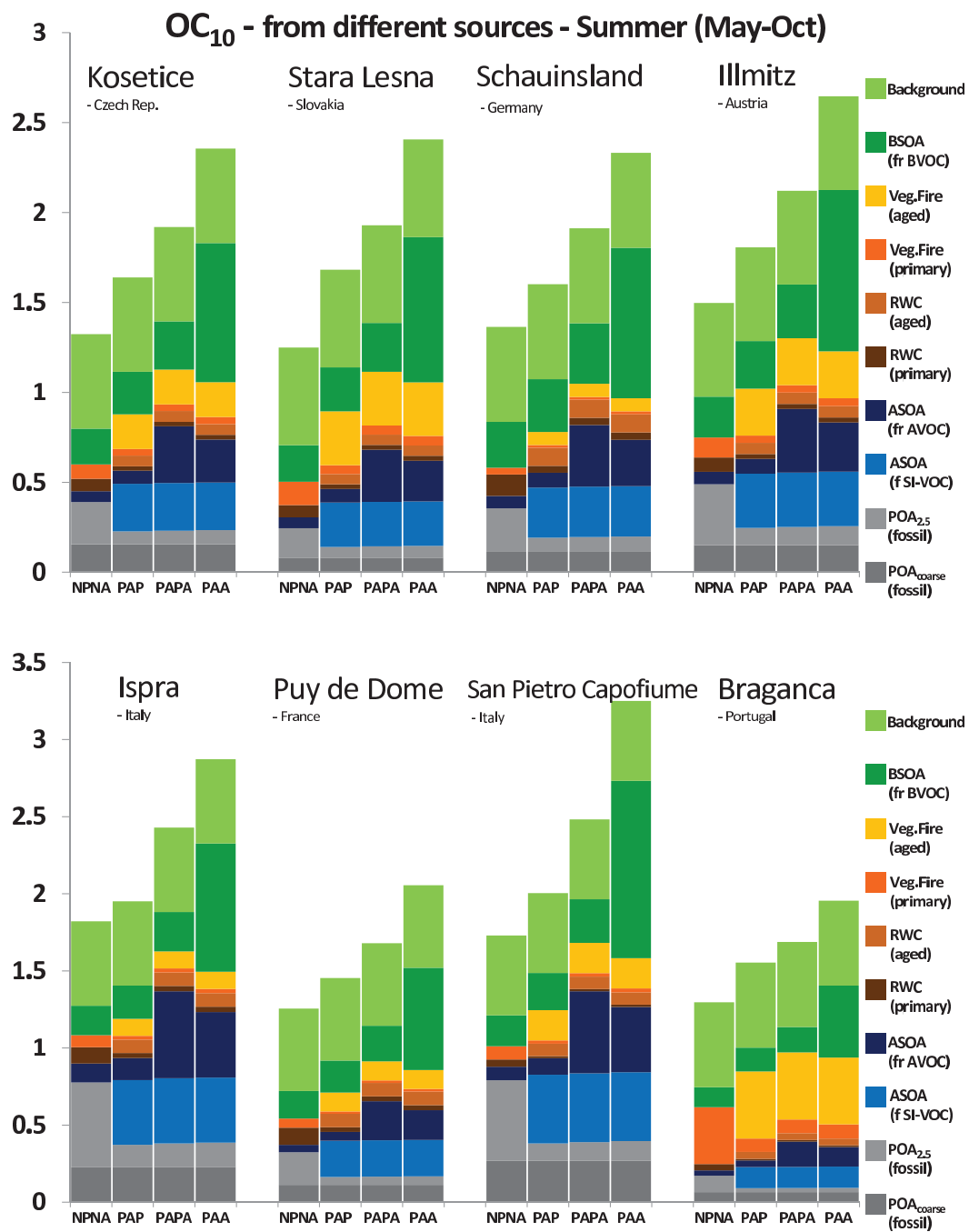


Fig. S5: (b), continued from Fig. S5(a).

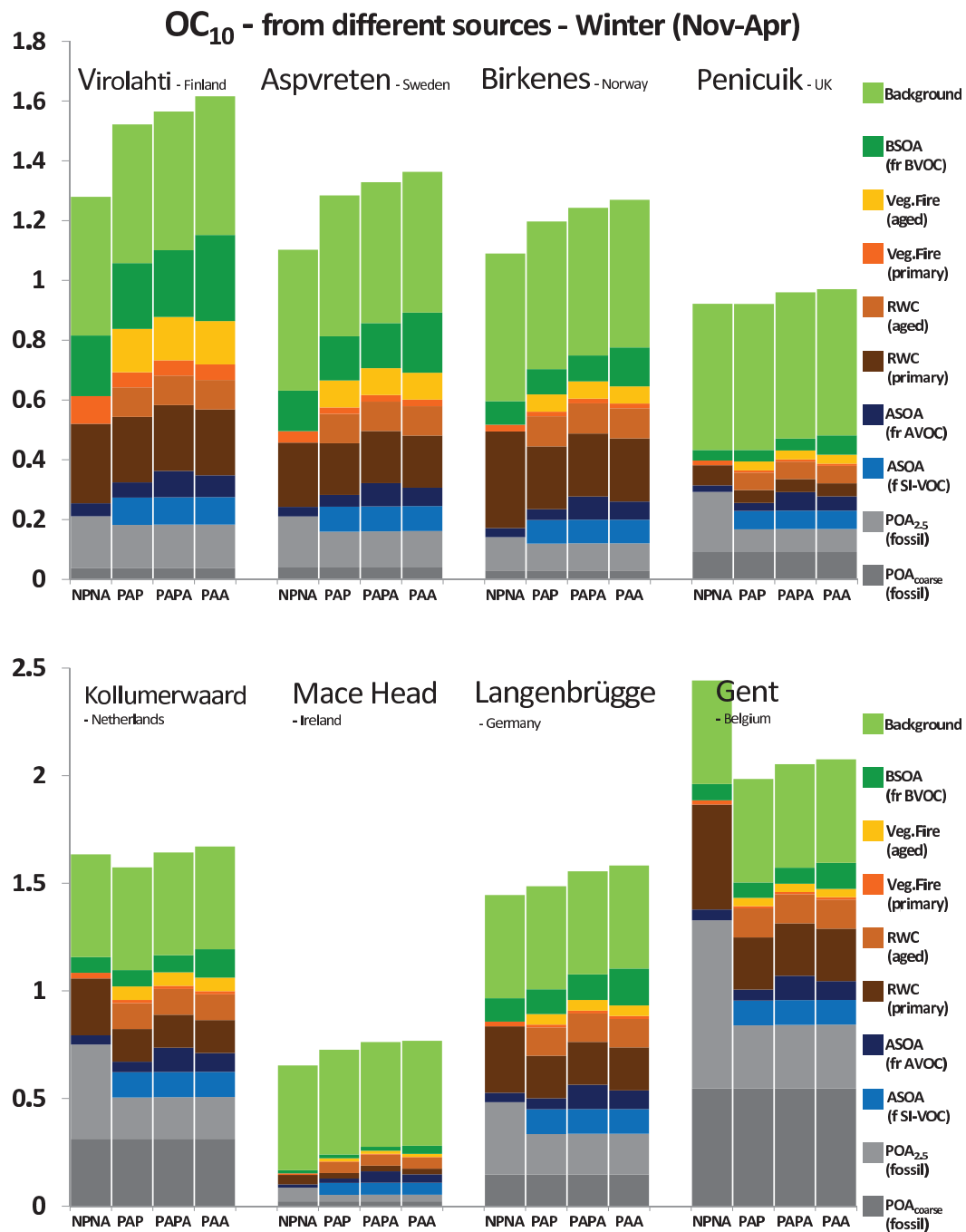


Fig. S6: (a) Modelled contribution from different sources to OC<sub>PM<sub>10</sub></sub> during the months Jan–April and November–December (winter half-years) 2002–2004, at selected sites, arranged from north to south. Continued with Fig. S6(b). Colours/Notation: see legend and Fig. 5(a).

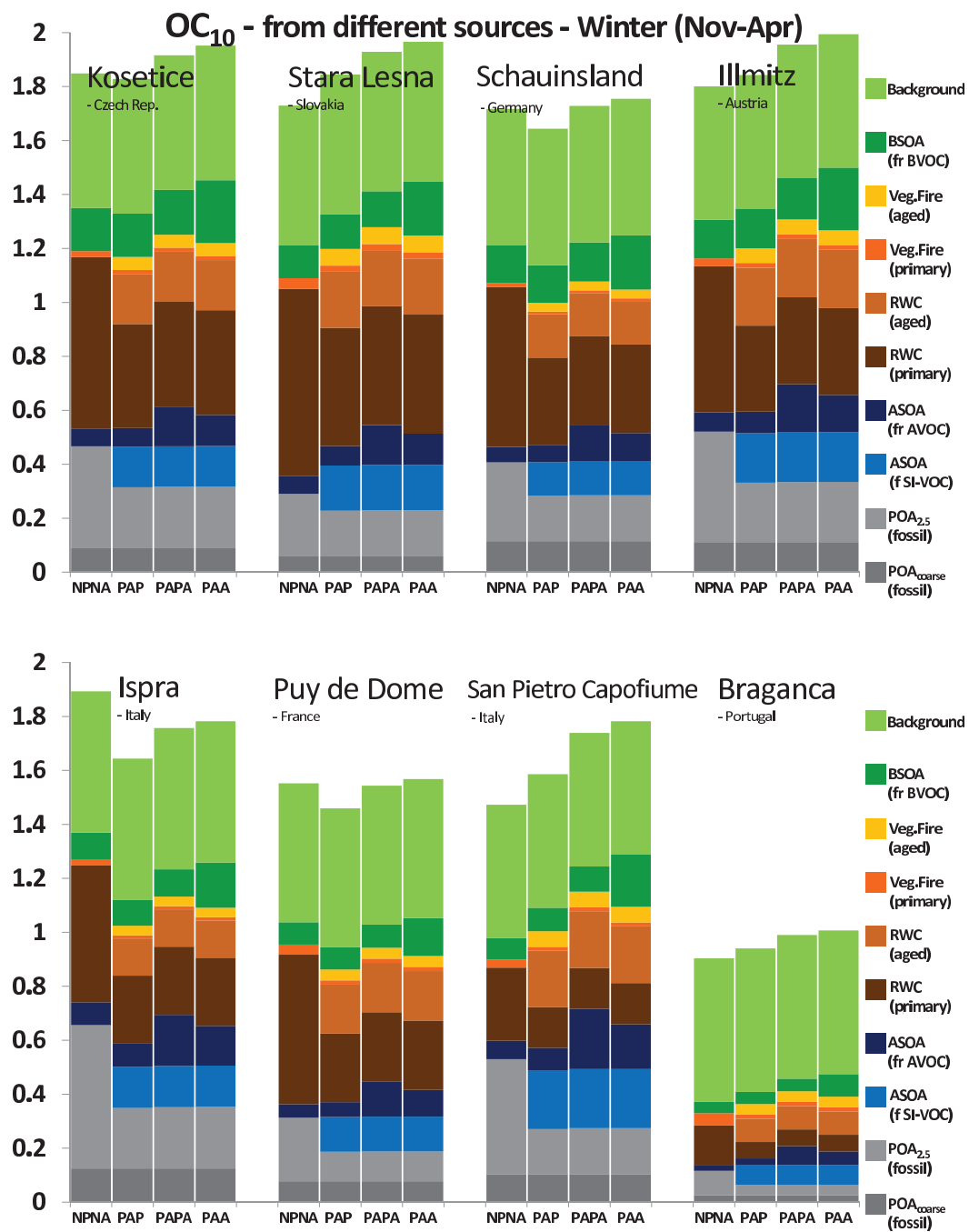


Fig. S6: (b), continued from Fig. S6(a).

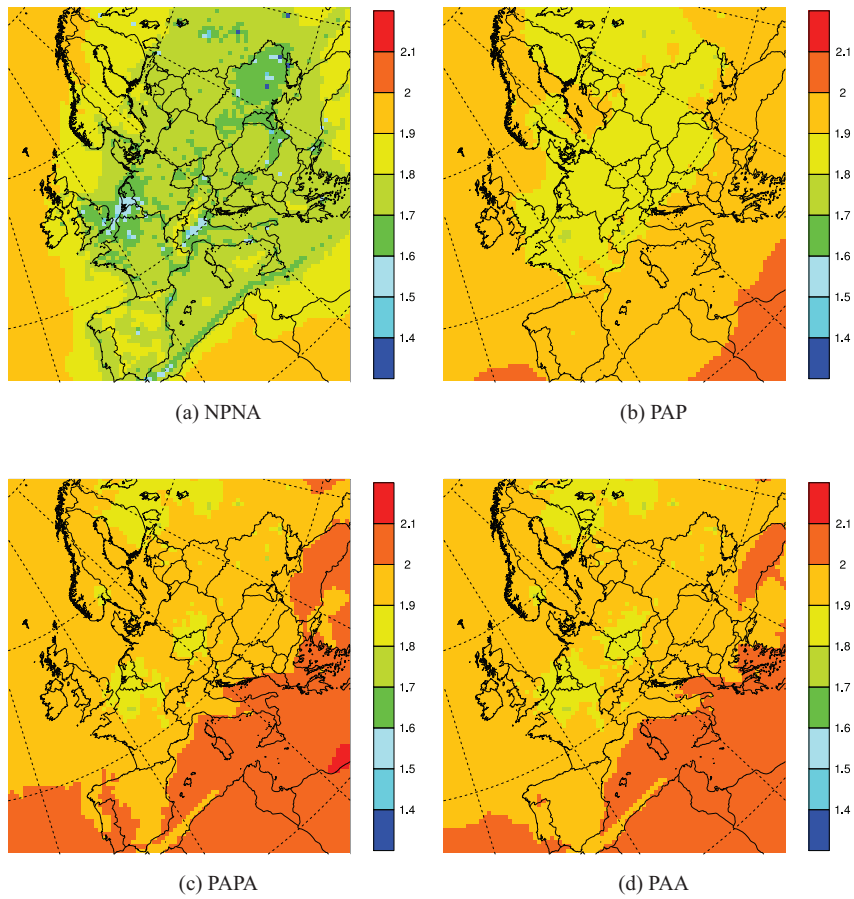


Fig. S7: Calculated OM/OC ratio in  $PM_{2.5}$  with four different model versions (see text). Average for the whole 6-yr period 2002–2007.

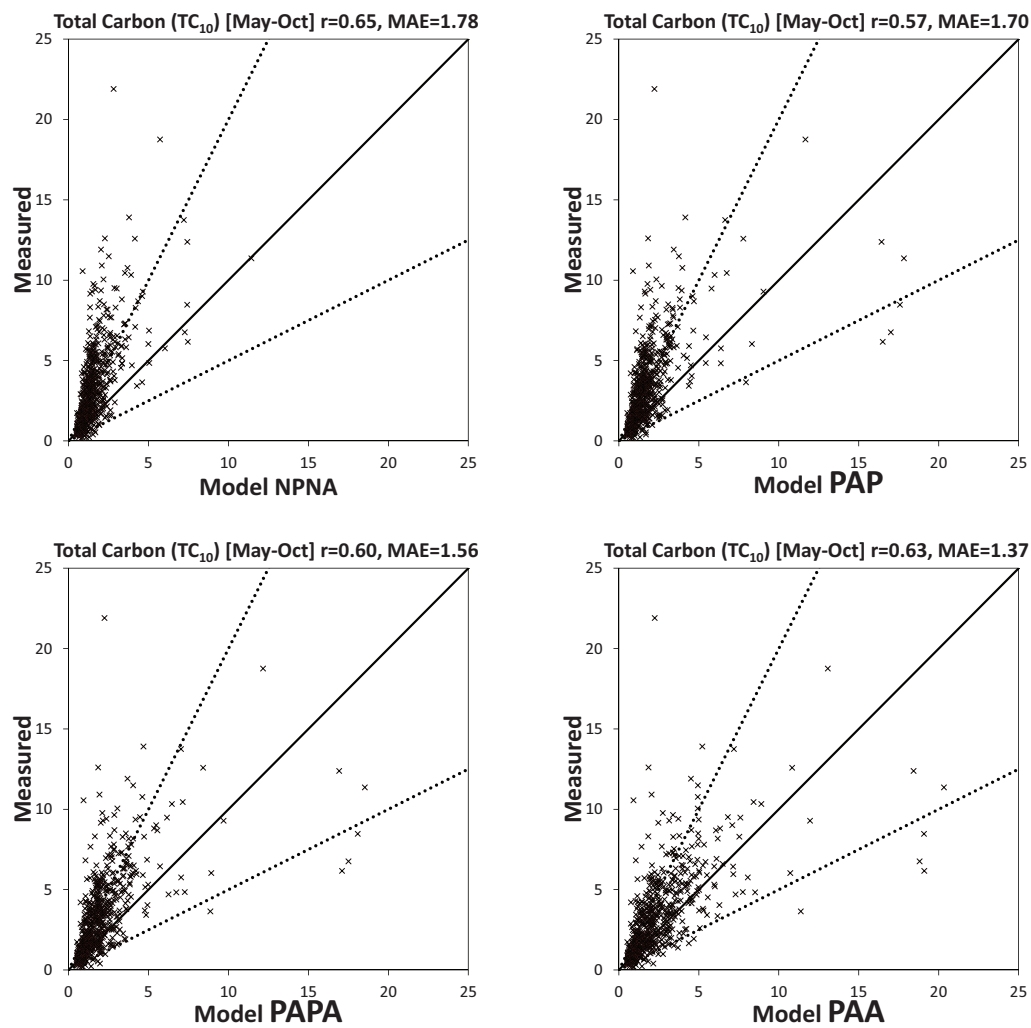


Fig. S8: Total Carbon (TC) in PM<sub>10</sub>. Data from summer half-years (May–October) from filter measurements and corresponding model concentrations. (Model versions: NPNA [top left], PAP [top right], PAPA [bottom left] and PAA [bottom right]). Statistics, see Table S1. Units are  $\mu\text{g}(\text{C})\text{m}^{-3}$ .

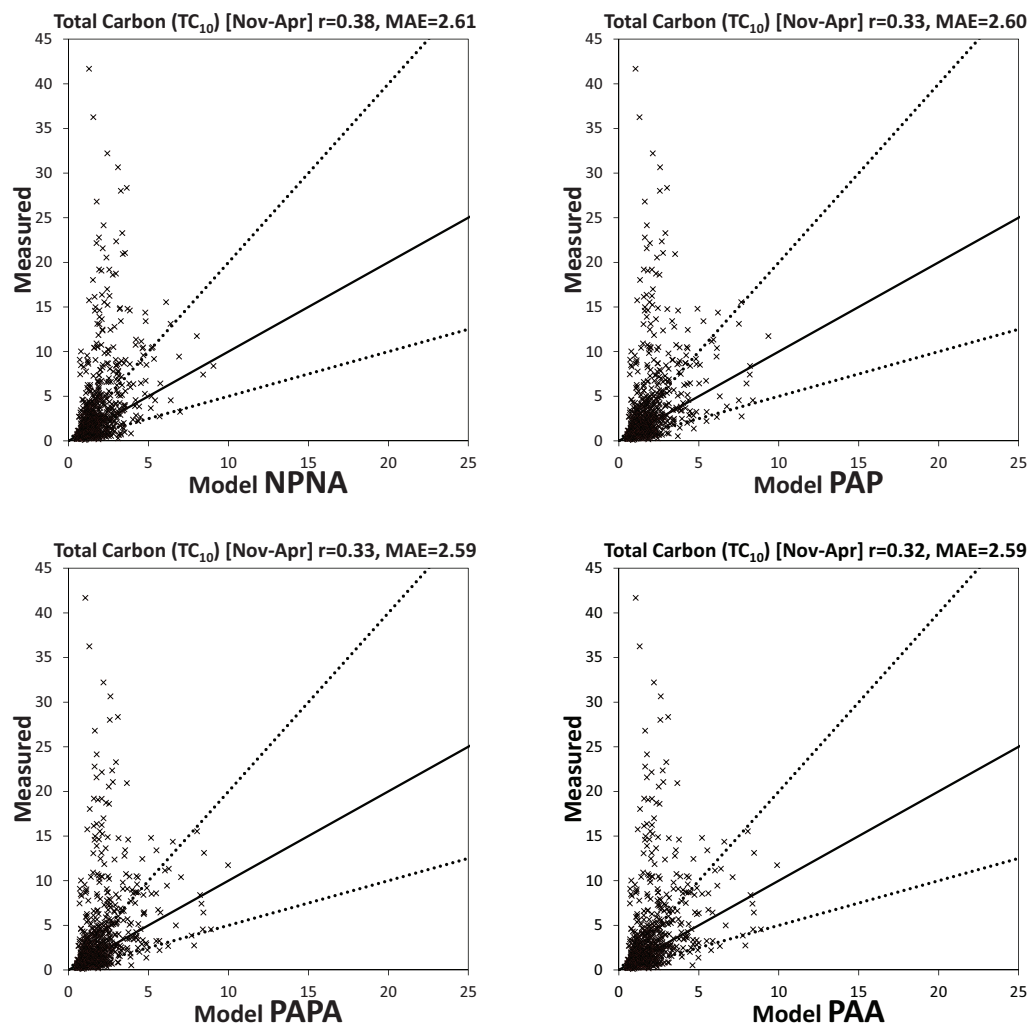


Fig. S9: Total Carbon (TC) in PM<sub>10</sub>. Data from winter half-years (November–April) from filter measurements and corresponding model concentrations. (Model versions: NPNA [top left], PAP [top right], PAPA [bottom left] and PAA [bottom right]). Statistics, see Table S1. Units are  $\mu\text{g}(\text{C})\text{m}^{-3}$ .



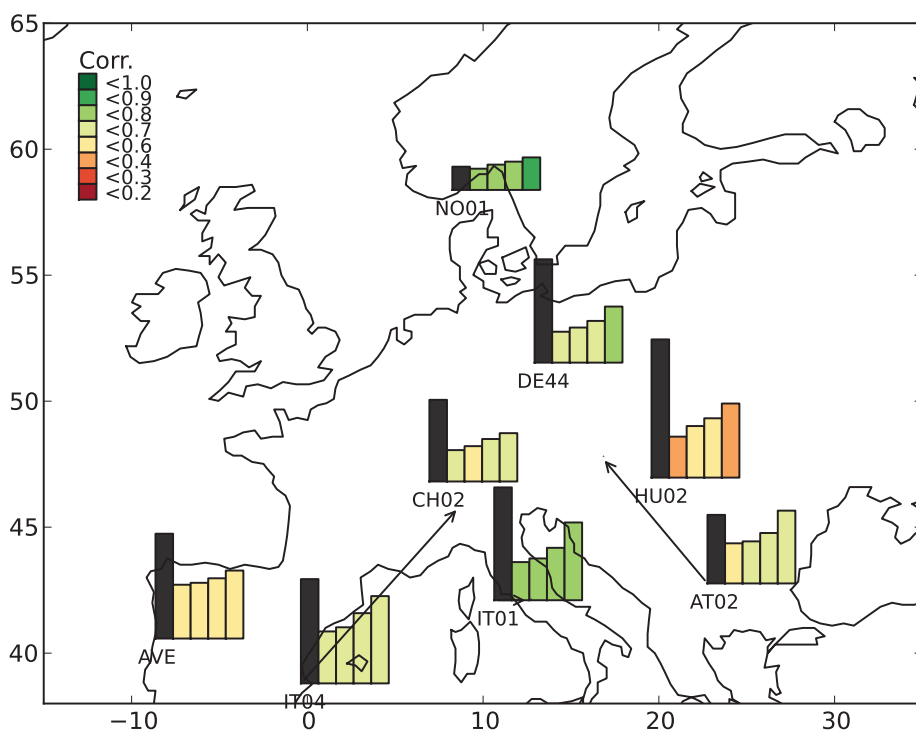


Fig. S10: Observed and modelled  $TC_{PM_{2.5}}$  during the summer half-year period (May–October) at different European sites from the CARBOSOL (2002–2004) campaign and EMEP intensive PM measurement period (2006). The leftmost bars show observed average concentrations (black for stations located at less than 600 m altitude, light gray for sites above 1000 m and medium gray for stations at 600–1000 m height) and the following four bars the corresponding model concentrations with the four different model versions (NPNA, PAP, PAPA and PAA). The colours of the model bars illustrate the correlation coefficients. Note that number of samples varies between stations (N=12–103) - see Table S4 for details.

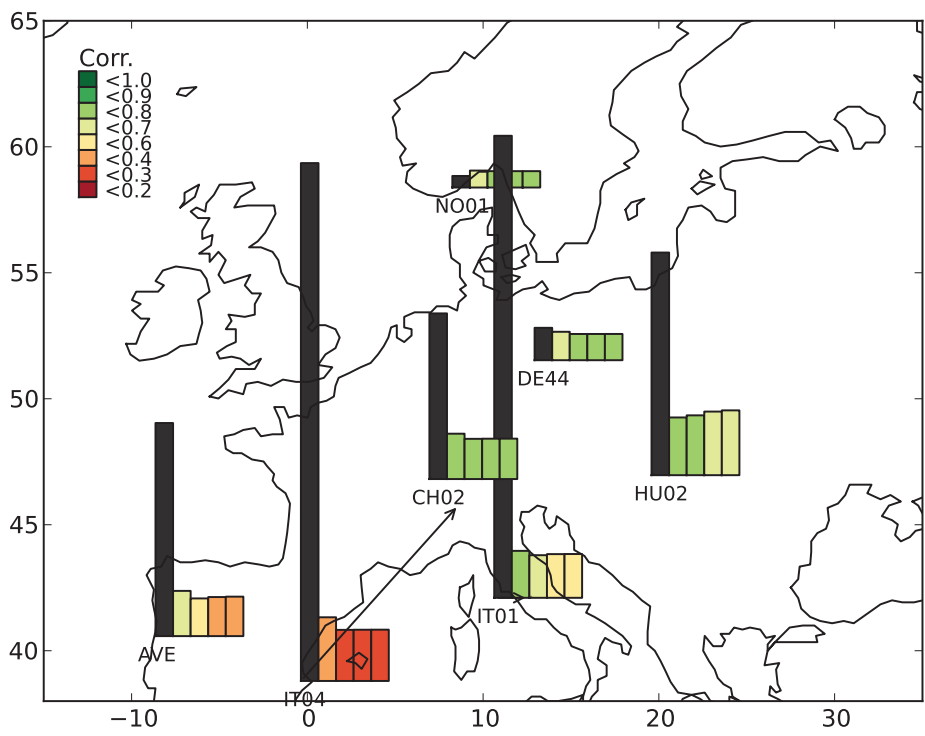


Fig. S11: Observed and modelled  $TC_{PM_{2.5}}$  during the winter half-year period (November–April) at different European sites from the CARBOSOL campaign (2002-2004) and EMEP intensive PM measurement period (2007). Details/Notation see Fig. S10.

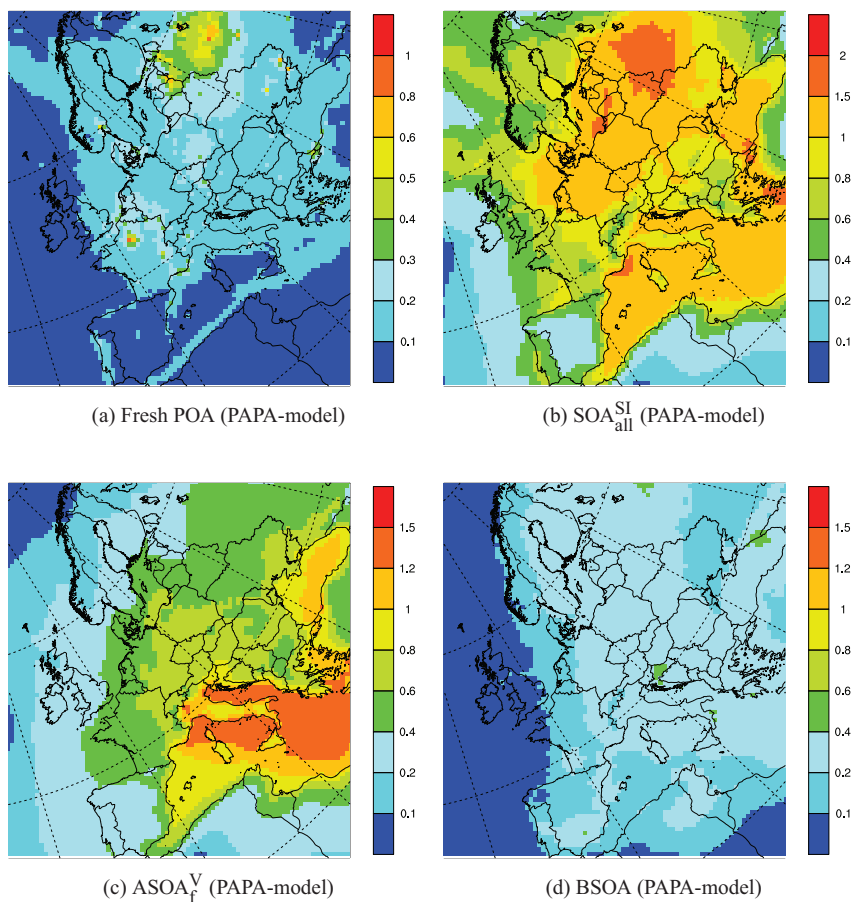


Fig. S12: Calculated concentrations of fresh primary organic aerosol (POA) and secondary organic aerosol (SOA) in  $PM_{2.5}$  for the month May with the PAPA model version. Note that in these maps POA includes both *all* anthropogenic POA (including residential wood combustion) and POA from vegetation fires.  $SOA_{all}^{SI}$  includes SOA formed from S/IVOC species emitted from anthropogenic sources as well as vegetation fires. Average for all May-months in the 6-yr period 2002–2007. Unit:  $\mu g m^{-3}$ .

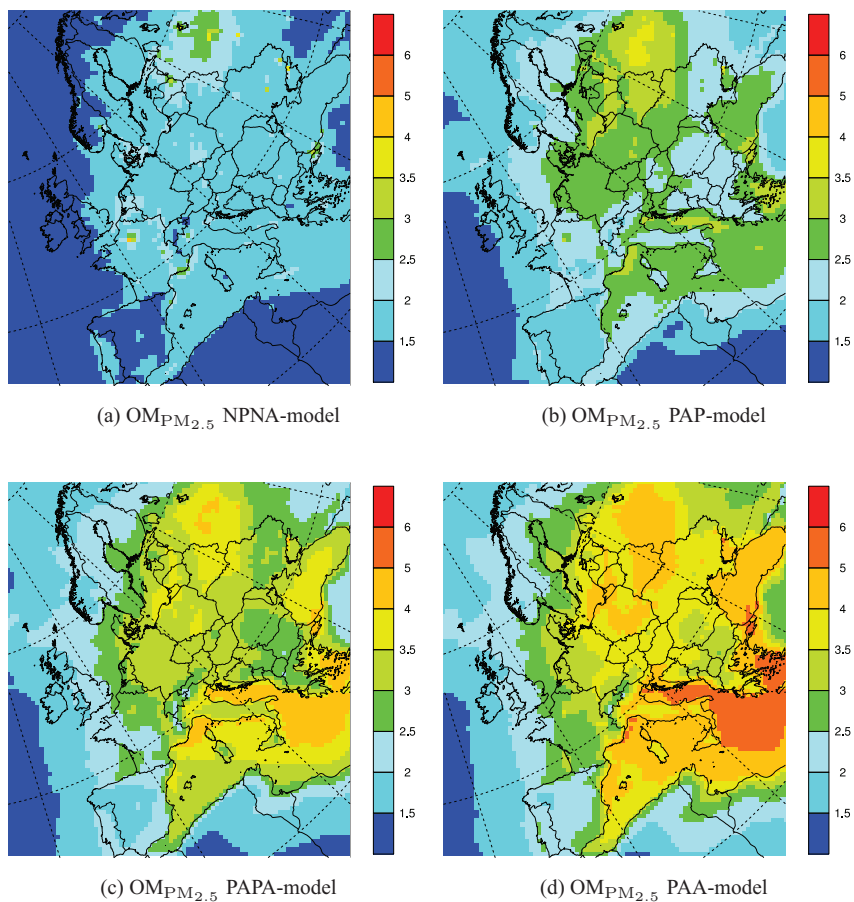


Fig. S13: Calculated total organic mass in  $PM_{2.5}$  ( $OM_{PM_{2.5}}$ ) for the month May with the four different model versions included in this study (see text). Average for all May-months in the 6-yr period 2002–2007. Unit:  $\mu\text{g m}^{-3}$ .

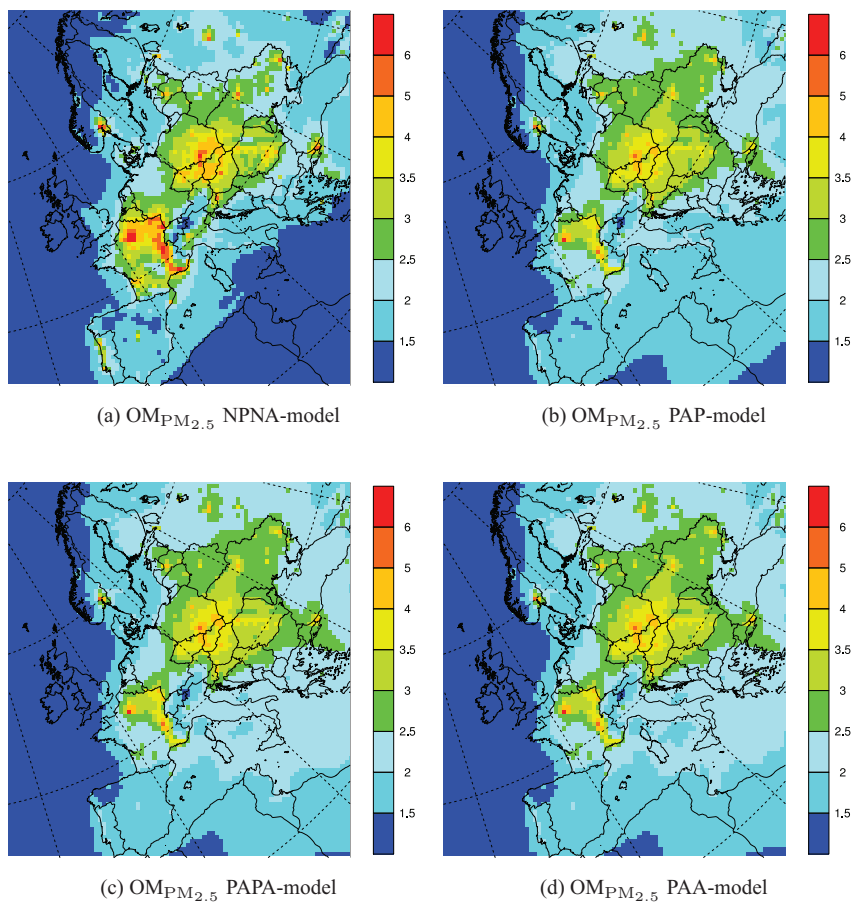


Fig. S14: Calculated total organic mass in  $PM_{2.5}$  for the month January with the four different model versions included in this study (see text). Average for all January-months in the 6-yr period 2002–2007. Unit:  $\mu\text{g m}^{-3}$ .