Supplementary material:

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

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

A1 Extra Tables

Table A1: Measurement sites and campaigns used in this study.

	Country	Latitude	Longitude	Measurements	Notes
Schauinsland	Germany	47.91	7.91	EC, OC, S-A†, $PM_{2.5}$	(a)
Puy de Dome	France	45.77	2.95	EC, OC, S-A, $PM_{2.5}$	(a)
K-puszta	Hungary	46.97	19.58	EC, OC, S-A, $PM_{2.5}$	(a)
Aveiro	Portugal	40.58	-8.64	EC, OC, S-A, $PM_{2.5}$	(a)
Virolahti	Finland	60.53	27.69	EC, OC, PM_{10}	(b)
Aspvreten	Sweden	58.80	17.38	EC, OC, PM_{10}	(b)
Birkenes	Norway	58.38	8.25	EC, OC, PM_{10}, $PM_{2.5}$ (2006,2007)	(c), (d)
Penicuik	United Kingdom	55.86	-3.21	EC, OC, PM_{10}	(b)
Kollumerwaard	the Netherlands	53.33	6.28	EC, OC, PM_{10}	(b)
Gent	Belgium	51.05	3.72	EC, OC, PM_{10}	(b) (e)
Mace Head	Ireland	53.33	-9.90	EC, OC, PM_{10}	(b)
Langenbrügge	Germany	52.80	10.76	EC, OC, PM_{10}	(b)
Kosetice	Czech Republic	49.58	15.08	EC, OC, PM_{10}	(b), (c)
Stara Lesna	Slovakia	49.15	20.28	EC, OC, PM_{10}	(b)
Illmitz	Austria	47.77	16.77	EC, OC, PM_{10}, $PM_{2.5}$ (S, 2006)	(b), (c), (f 2006)
Ispra	Italy	45.80	8.63	EC, OC, PM_{10} (2002-2003), $PM_{2.5}$ (2006,2007)	(b), (c)
Braganca	Portugal	41.82	-6.77	EC, OC, PM_{10}	(b)
San Pietro Capofiume	Italy	44.48	11.33	EC, OC, PM_{10}	(b), (e)
Harwell	United Kingdom	51.57	-1.32	EC, OC, PM_{10} (S)	(c), (f)
Melpitz	Germany	51.53	12.93	EC, OC, PM_{10}, $PM_{2.5}$	(c)
Payerne	Switzerland	46.81	6.94	EC, OC, $PM_{2.5}$, AMS	(c), (f)
Montelibretti	Italy	42.10	12.63	EC, OC, PM_{10}, $PM_{2.5}$	(c)
Montseny	Spain	41.77	2.35	EC(S), OC(S), TC(W), PM_{10}	(c)
Hurdal	Norway	60.37	11.07	EC, OC, S-A, PM_1	(g)
Oslo	Norway	59.93	10.73	EC, OC, S-A, PM_1	(g), (e)
Gothenburg	Sweden	57.72	11.97	EC, OC, S-A, PM_{10} (W), $PM_{2.5}$ (S)	(h), (e)
Råö	Sweden	57.39	11.91	EC, OC, S-A $PM_{2.5}$ (W)	(h)

Notes: † S-A indicates data for source-apportionment, see below; (S) indicates summer, (W) indicates winter; (a) CARBOSOL campaign, Oct. 2004-June 2006, used weekly filter measurements of EC, OC, cellulose, levoglucosan, and (for seasonally-pooled samples) ^{14}C, see Gelencsér et al. (2007), Pio et al. (2007); (b) EMEP EC/OC campaign, 1 July 2002-1 July 2003, 24h filter measurements of EC and OC, once per week, see Yttri et al. (2007); (c) EMEP PM intensive campaign June 2006(S) and 8 Jan.-4 Feb. 2007(W), many different measurements were performed in the campaign, see Yttri et al. (2008), Aas et al. (2012), here we use daily data from filter measurements of EC and OC and hourly AMS (OM) data from Payerne for the summer period; (d) For Birkenes filter measurement data for EC and OC in PM_{10} were available from EMEP for the full years 2002-2004. The data were either weekly measurements or alternatingly 6-days and 24h measurements; (e) Urban background station; (f) Hourly observation data were available, averaged here to daily means (except for the AMS data, that were averaged to hourly means); (g) - SORGA campaign, southern Norway, 19 June-15 July 2006(S) and 1-8 March 2007(W), included filter measurements of EC, OC, sugars, levoglucosan, and ^{14}C, (Yttri et al., 2011), here we use the PM_1 data and compare to the model $PM_{2.5}$ results; (h) - Göte-2005 campaign, southern Sweden, 11 Feb.-4 March 2005(W) and 13 June-4 July 2006(S), included measurements of EC, OC, sugars, levoglucosan, and ^{14}C, (Szidat et al., 2009).

Table A2: Filter measurements of Total Carbon (TC) in $PM_{2.5}$. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002-2007. Unit: μg(C) m^{-3}.

	N	Observed	VBS-NPNA	VBS-PAP	VBS-PAPA	VBS-PAA
CARBOSOL (October 2002 - June 2004):						
K-Puszta	77	7.31	2.13	2.37	2.59	2.88
Aveiro	103	6.33	2.02	1.91	2.03	2.19
summer 2006:						
Birkenes	30	0.92	0.88	1.04	1.16	1.32
Melpitz	31	4.10	1.33	1.49	1.76	2.33
Illmitz	29	2.72	1.73	1.81	2.14	3.03
Payerne	12	3.24	1.31	1.47	1.75	1.99
Ispra	23	4.14	2.21	2.38	2.94	3.62
Montseny	1	(1.85)	(2.08)	(2.69)	(3.36)	(3.98)
Montelibretti	31	4.48	1.62	1.77	2.19	3.19
winter 2007:						
Birkenes	30	0.46	0.69	0.67	0.67	0.67
Melpitz	33	1.29	1.22	1.13	1.13	1.13
Payerne	21	6.57	1.87	1.66	1.67	1.67
Ispra	28	20.6	2.50	2.00	2.01	2.01
Montelibretti	32	18.3	1.94	1.76	1.81	1.81
All Data	481	6.40	1.75	1.75	1.92	2.21
correlation coeff. (r)			0.47	0.28	0.23	0.13
mean absolute error			4.77	4.80	4.66	4.53
Summer Data	243	3.76	1.69	1.86	2.15	2.71
correlation coeff. (r)			0.49	0.49	0.51	0.53
mean absolute error			2.15	2.06	1.84	1.59
Winter Data	238	9.11	1.81	1.64	1.69	1.70
correlation coeff. (r)			0.58	0.46	0.44	0.43
mean absolute error			7.44	7.59	7.55	7.54

Notes: For a few stations hourly observation data were available. Here these were averaged to daily means.

Table A3: Filter measurements of Organic Carbon (OC) in PM_{10}. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002-2007. Unit: μg(C) m^{-3}.

	N	Observed	VBS-NPNA	VBS-PAP	VBS-PAPA	VBS-PAA
CARBOSOL (October 2002 - June 2004):						
Schauinsland	104	2.40	1.54	1.64	1.84	2.09
Puy de Dome	86	1.52	1.36	1.41	1.55	1.72
EMEP EC/OC (July 2002 - June 2003):						
Virolahti	51	2.08	1.27	1.73	1.82	2.08
Aspvreten	48	2.12	1.23	1.74	1.86	2.08
Birkenes(2002-2004)	267	1.07	1.00	1.18	1.25	1.38
Penicuik	50	1.53	1.04	1.08	1.17	1.24
Kollumerwaard	50	2.59	1.56	1.71	1.87	2.05
Gent	52	4.12	2.33	2.05	2.21	2.34
Mace Head	50	1.20	0.69	0.81	0.88	0.93
Langenbrügge	50	4.30	1.39	1.67	1.83	2.07
Kosetice	38	4.54	1.65	1.75	1.90	1.99
Stara Lesna	52	4.32	1.60	2.07	2.26	2.57
Illmitz	50	5.51	1.65	1.97	2.17	2.49
Ispra	45	7.79	1.79	1.75	2.11	2.35
Braganca	50	4.10	1.08	1.30	1.38	1.49
San Pietro Capofiume	50	5.91	1.61	1.80	2.11	2.52
EMEP intensive PM measurement period summer 2006:						
Birkenes	30	1.03	0.77	0.93	1.05	1.22
Harwell	17	0.83	1.05	1.09	1.33	1.44
Melpitz	31	2.55	1.05	1.21	1.48	2.05
Kosetice	21	2.47	1.09	1.27	1.55	2.25
Montseny	11	2.19	1.46	1.72	2.23	3.60
Montelibretti	31	4.13	1.19	1.34	1.76	2.76
EMEP intensive PM measurement period winter 2007:						
Birkenes	30	0.52	0.62	0.59	0.59	0.60
Melpitz	33	1.38	0.86	0.78	0.78	0.78
Kosetice	29	0.37	1.17	1.00	1.01	1.01
Montelibretti	31	15.5	1.33	1.16	1.20	1.21
All Data	1357	2.93	1.28	1.42	1.57	1.78
correlation coeff. (r)			0.39	0.32	0.32	0.32
mean absolute error			1.91	1.85	1.78	1.70
Summer Data (May-Oct)	671	2.63	1.20	1.45	1.68	2.08
correlation coeff. (r)			0.63	0.56	0.60	0.63
mean absolute error			1.51	1.43	1.29	1.13
Winter Data (Nov-Apr)	686	3.22	1.36	1.40	1.47	1.49
correlation coeff. (r)			0.31	0.24	0.24	0.23
mean absolute error			2.30	2.27	2.25	2.25

Notes: For one station (Harwell) hourly observation data were available. Here these were averaged to daily means.

Table A4: Filter measurements of Elemental Carbon (EC) in PM_{10}. Comparison of model results to data from field campaigns in 2002-2007. Unit: μg(C) m^{-3}.

	N	Observed	Model
CARBOSOL (October 2002 - June 2004):			
Schauinsland	104	0.29	0.58
Puy de Dome	86	0.22	0.44
EMEP EC/OC (July 2002 - June 2003):			
Virolahti	51	0.36	0.43
Aspvreten	48	0.29	0.40
Birkenes(2002-2004)	256	0.12	0.28
Penicuik	50	0.51	0.50
Kollumerwaard	50	0.63	0.73
Gent	52	1.80	1.48
Mace Head	50	0.20	0.17
Langenbrügge	50	0.63	0.60
Kosetice	38	1.05	0.80
Stara Lesna	52	0.80	0.60
Illmitz	50	1.00	0.71
Ispra	45	1.83	0.96
Braganca	50	0.79	0.23
San Pietro Capofiume	50	1.44	0.84
EMEP intensive PM measurement period summer 2006:			
Birkenes	30	0.12	0.14
Harwell	17	0.50	0.52
Melpitz	31	1.82	0.44
Kosetice	21	0.33	0.41
Montseny	11	0.27	0.70
Montelibretti	31	1.30	0.50
EMEP intensive PM measurement period winter 2007:			
Birkenes	30	0.060	0.076
Melpitz	33	0.99	0.43
Kosetice	29	2.02	0.58
Montelibretti	31	1.30	0.69
All Data	1346	0.65	0.52
correlation coeff. (r)			0.54
mean absolute error			0.39
Summer Data (May-Oct)	667	0.56	0.42
correlation coeff. (r)			0.59
mean absolute error			0.31
Winter Data (Nov-Apr)	679	0.74	0.62
correlation coeff. (r)			0.51
mean absolute error			0.47

Notes: For one station (Harwell) hourly observation data were available. Here these were averaged to daily means.

Table A5: Filter measurements of Organic Carbon (OC) in $PM_{2.5}$. Comparison of model results (four different model versions, see text) to data from field campaigns in 2002-2007. Unit: μg(C) m^{-3}.

	N	Observed	VBS-NPNA	VBS-PAP	VBS-PAPA	VBS-PAA
CARBOSOL (October 2002 - June 2004):						
K-Puszta	77	6.17	1.48	1.72	1.94	2.24
Aveiro	103	5.33	1.49	1.38	1.49	1.65
summer 2006:						
Birkenes	30	0.84	0.75	0.91	1.03	1.20
Melpitz	31	1.20	0.98	1.14	1.40	1.97
Illmitz	29	2.35	1.28	1.36	1.69	2.58
Payerne	12	2.67	1.01	1.16	1.45	1.68
Ispra	22	3.34	1.45	1.62	2.16	2.84
Montseny	1	(1.77)	(1.34)	(1.95)	(2.63)	(3.25)
Montelibretti	31	3.33	1.14	1.29	1.71	2.71
winter 2007:						
Birkenes	30	0.40	0.62	0.59	0.59	0.59
Melpitz	33	0.69	0.83	0.74	0.74	0.74
Payerne	21	5.15	1.32	1.11	1.12	1.12
Ispra	28	16.3	1.58	1.08	1.08	1.08
Montelibretti	32	17.2	1.28	1.11	1.15	1.16
All Data	480	5.24	1.24	1.25	1.42	1.70
correlation coeff. (r)			0.39	0.15	0.10	0.01
mean absolute error			4.12	4.14	4.03	3.95
Summer Data	242	2.83	1.24	1.41	1.70	2.25
correlation coeff. (r)			0.51	0.51	0.53	0.51
mean absolute error			1.69	1.59	1.41	1.28
Winter Data	238	7.69	1.25	1.09	1.13	1.14
correlation coeff. (r)			0.53	0.33	0.31	0.30
mean absolute error			6.59	6.73	6.69	6.67

Notes: For a few stations hourly observation data were available. Here these were averaged to daily means.

Table A6: Filter measurements of Elemental Carbon (EC) in $PM_{2.5}$. Comparison of model results to data from field campaigns in 2002-2007. Unit: μg(C) m^{-3}.

	N	Observed	Model
CARBOSOL (October 2002 - June 2004):			
K-Puszta	77	1.15	0.64
Aveiro	103	1.00	0.53
summer 2006:			
Birkenes	30	0.09	0.13
Melpitz	31	2.90	0.35
Illmitz	28	0.39	0.45
Payerne	12	0.57	0.31
Ispra	23	0.77	0.72
Montseny	1	(0.084)	(0.735)
Montelibretti	31	1.15	0.48
winter 2007:			
Birkenes	30	0.051	0.075
Melpitz	33	0.59	0.39
Payerne	21	1.42	0.55
Ispra	28	4.29	0.93
Montelibretti	32	1.10	0.66
All Data	480	1.17	0.50
correlation coeff. (r)			0.38
mean absolute error			0.74
Summer Data (May-Oct)	242	0.93	0.45
correlation coeff. (r)			0.10
mean absolute error			0.58
Winter Data (Nov-Apr)	238	1.41	0.56
correlation coeff. (r)			0.50
mean absolute error			0.89

Notes: For a few stations hourly observation data were available. Here these were averaged to daily means.

A2 Extra Figures

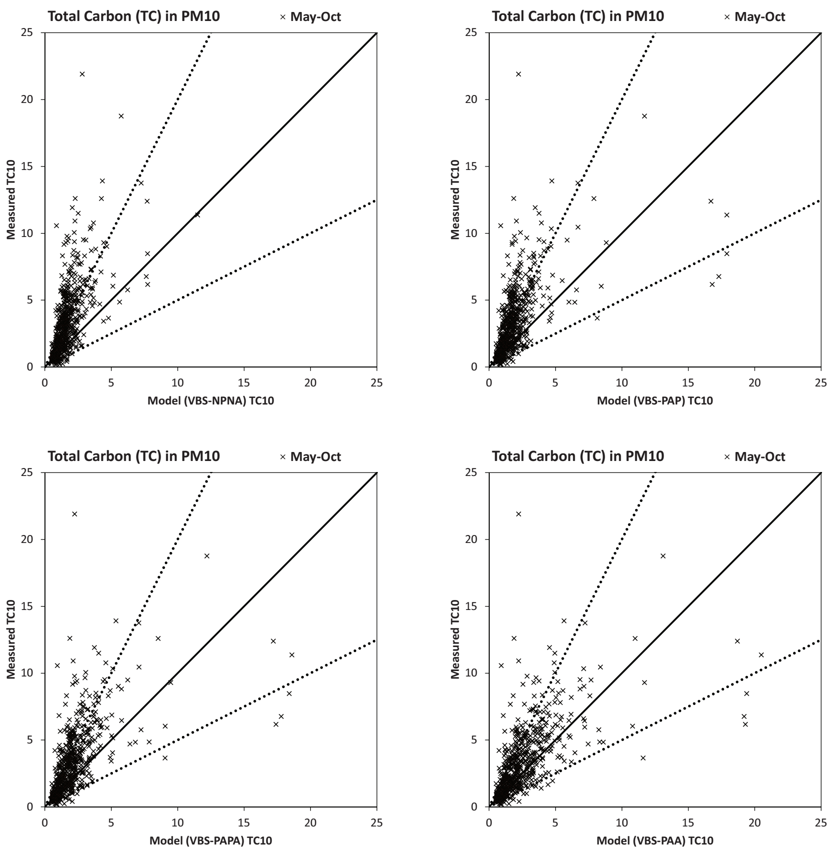


Fig. A1: Total Carbon (TC) in PM_{10}. Measured and modelled concentrations, May-Oct (summer) data (Model versions: VBS-NPNA[top left], VBS-PAP[top right], VBS-PAPA[bottom left] and VBS-PAA[bottom right]). Statistics, see Table 4. Units are μg(C) m^{-3}.

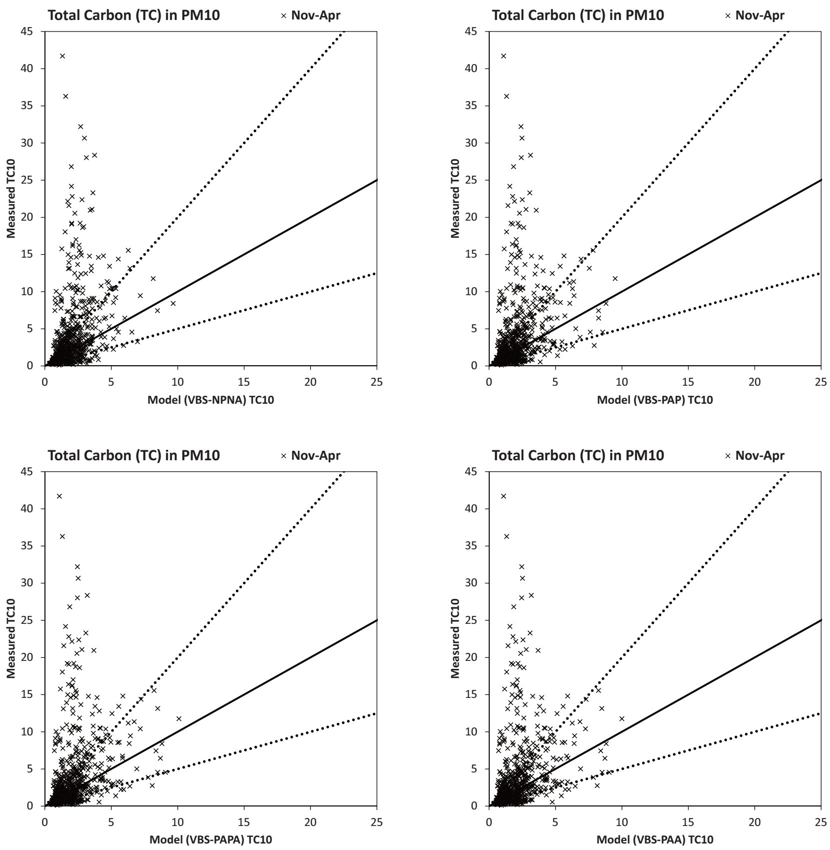
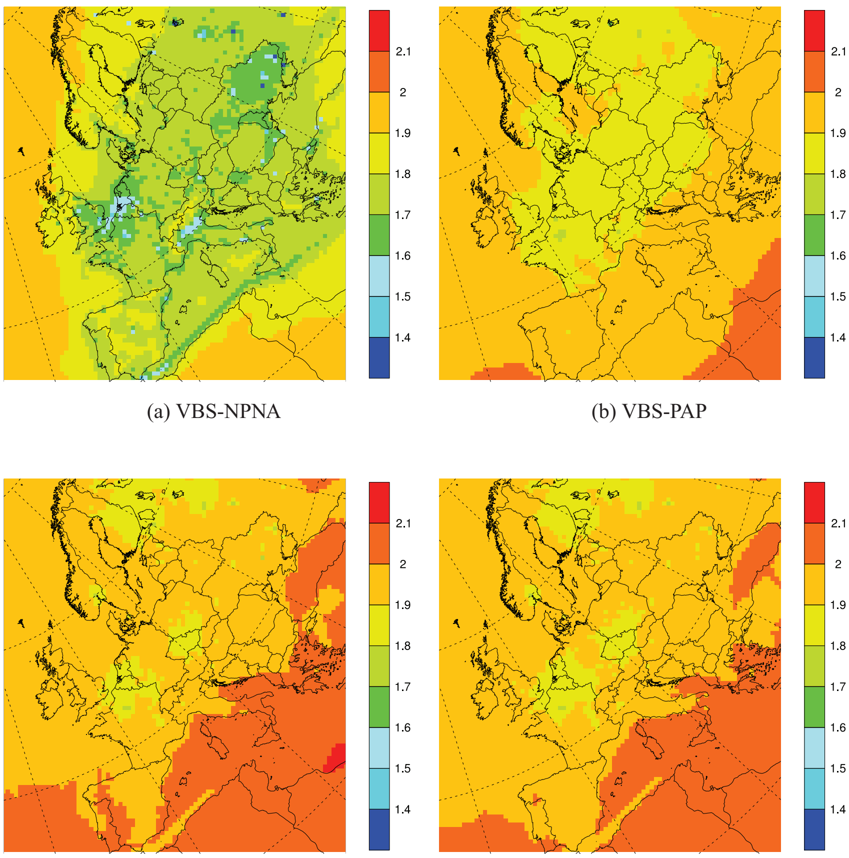
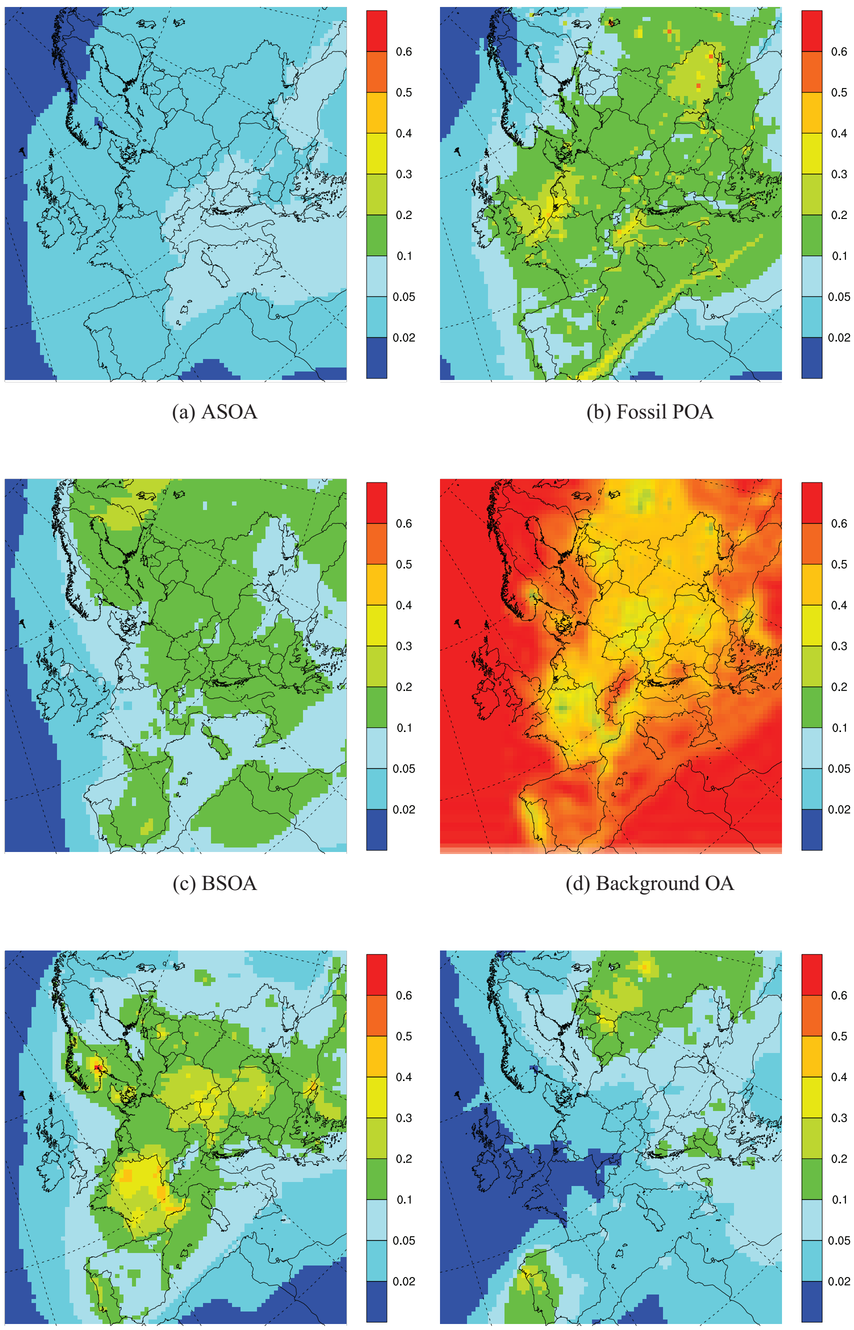


Fig. A2: Total Carbon (TC) in PM_{10}. Measured and modelled concentrations, Nov-Apr (winter) data (Model versions: VBS-NPNA[top left], VBS-PAP[top right], VBS-PAPA[bottom left] and VBS-PAA[bottom right]). Statistics, see Table 4. Units are μg(C) m^{-3}.



(a) VBS-NPNA (b) VBS-PAP (c) VBS-PAPA (d) VBS-PAA

Fig. A3: Calculated OM/OC ratio in $PM_{2.5}$ with four different model versions (see text). Average for the whole 6-year period 2002-2007.



(a) ASOA

(b) Fossil POA

(c) BSOA

(d) Background OA

(e) Residential Wood Burning OA

(f) Wildfire OA

Fig. A4: Calculated relative contribution to total particulate OM in $PM_{2.5}$ from different sources, using the model version VBS-NPNA. Fraction of $OM_{PM2.5}$ from (a) anthropogenic SOA, (b) fossil fuel primary OA (POA) and oxidised POA, (c) biogenic SOA, (d) background organic aerosol (from sources not explicitly included in the model), (e) residential biomass burning, (f) vegetation (wild) fires. Average for the 6-year period 2002-2007.

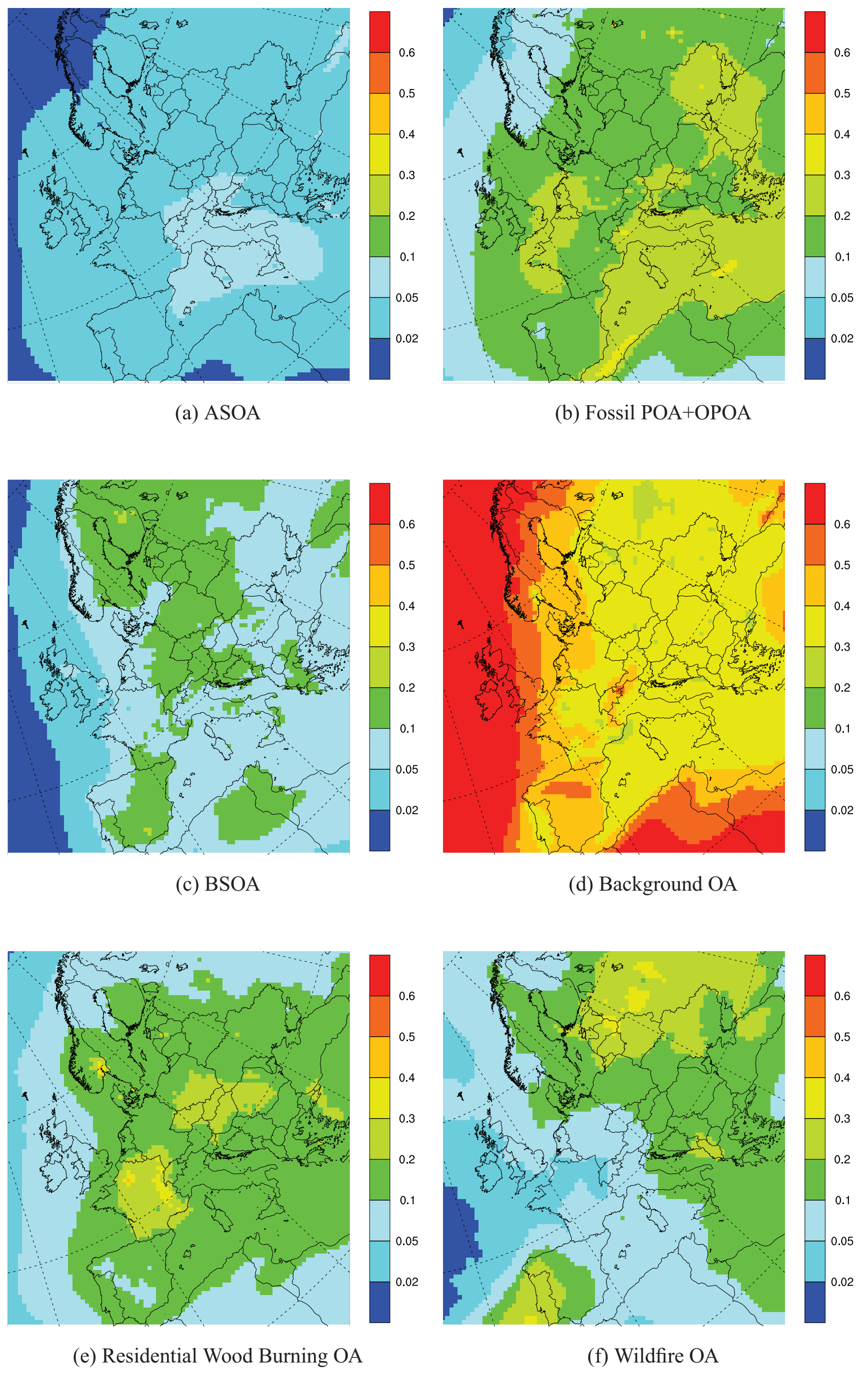
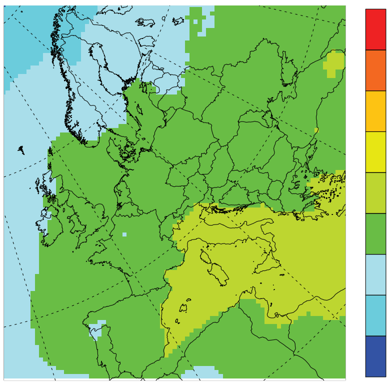
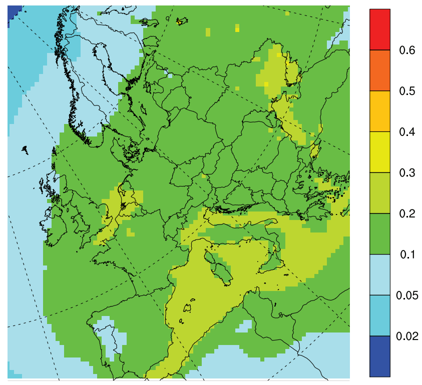


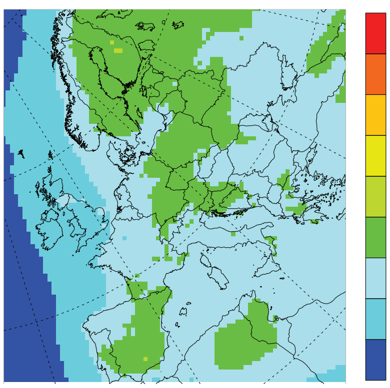
Fig. A5: Calculated relative contribution to total particulate OM in $PM_{2.5}$ from different sources, using the model version VBS-PAP. Fraction of $OM_{PM2.5}$ from (a) anthropogenic SOA, (b) fossil fuel primary OA (POA) and oxidised POA, (c) biogenic SOA, (d) background organic aerosol (from sources not explicitly included in the model), (e) residential biomass burning, (f) vegetation (wild) fires. Average for the 6-year period 2002-2007.



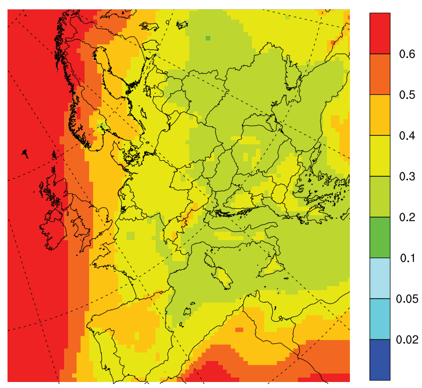
(a) ASOA



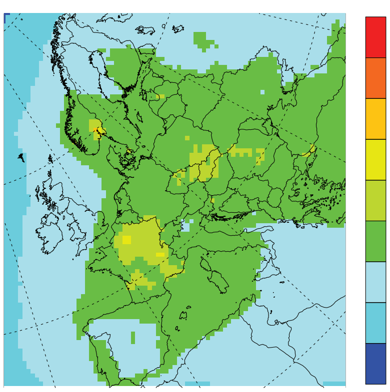
(b) Fossil POA+OPOA



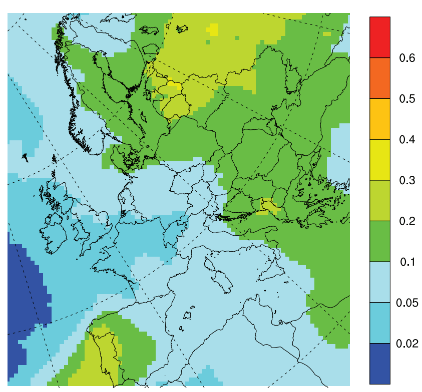
(c) BSOA



(d) Background OA

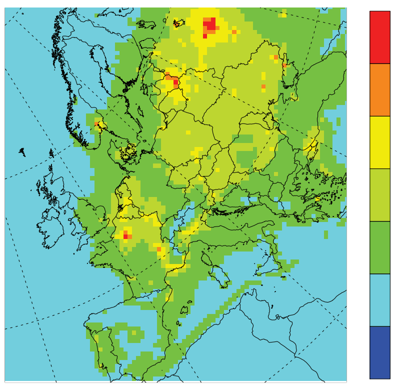


(e) Residential Wood Burning OA

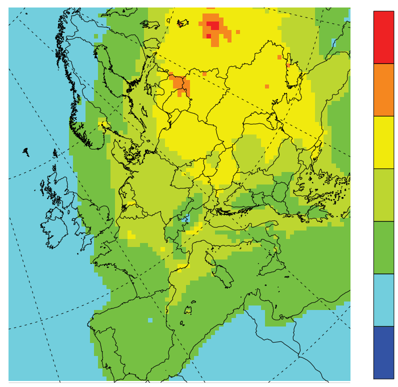


(f) Wildfire OA

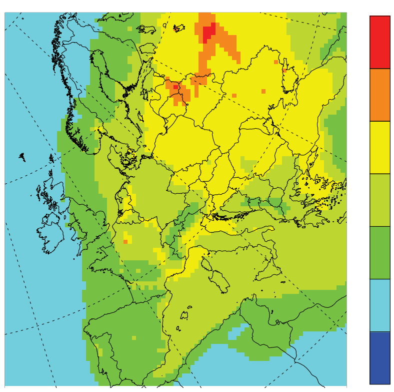
Fig. A6: Calculated relative contribution to total particulate OM in $PM_{2.5}$ from different sources, using the model version VBS-PAPA. Fraction of $OM_{PM2.5}$ from (a) anthropogenic SOA, (b) fossil fuel primary OA (POA) and oxidised POA, (c) biogenic SOA, (d) background organic aerosol (from sources not explicitly included in the model), (e) residential biomass burning, (f) vegetation (wild) fires. Average for the 6-year period 2002-2007.



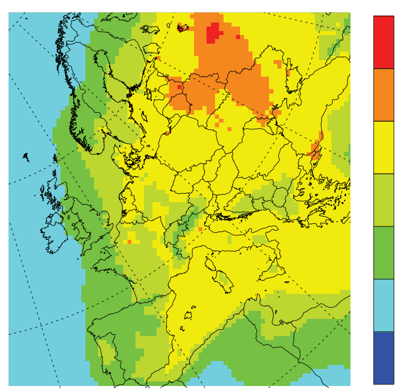
(a) OC_{PM10} VBS-NPNA



(b) OC_{PM10} VBS-PAP

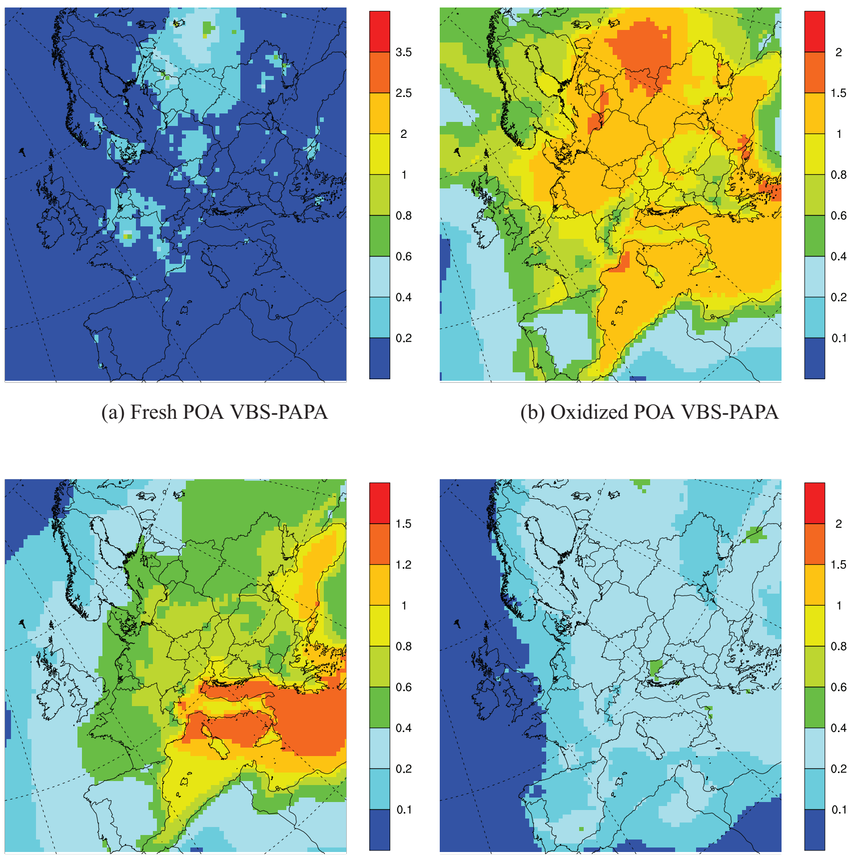


(c) OC_{PM10} VBS-PAPA



(d) OC_{PM10} VBS-PAA

Fig. A7: Calculated yearly average concentration of total organic carbon (OC) in PM_{10} for 2002, with the four different model versions included in this study (see text). Unit: μg(C) m^{-3}



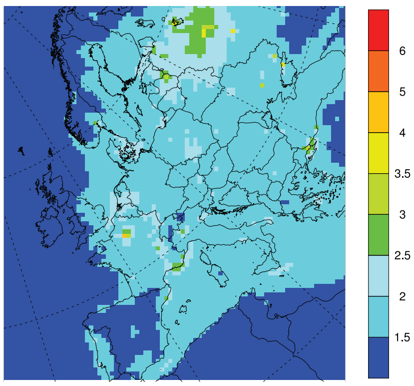
(a) Fresh POA VBS-PAPA

(b) Oxidized POA VBS-PAPA

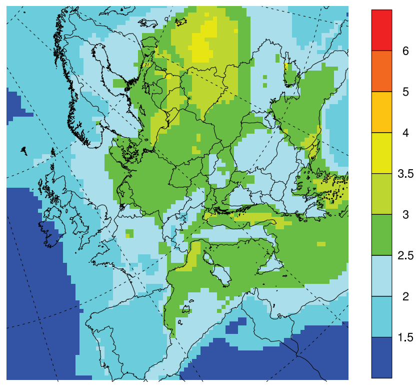
(c) ASOA VBS-PAPA

(d) BSOA VBS-PAPA

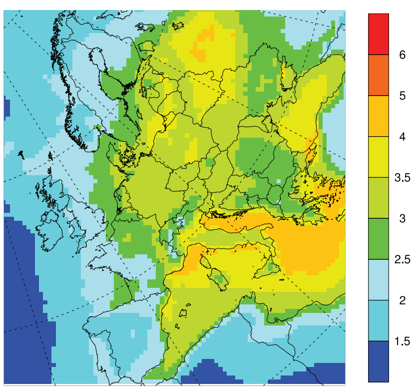
Fig. A8: Calculated concentrations of fresh and oxidized primary organic aerosol (POA) and anthropogenic and biogenic SOA ("traditional" ASOA and BSOA) in $PM_{2.5}$ for the month May with the EMEP VBS-PAPA model version. Note that in these maps POA includes *all* anthropogenic POA (including residential biomass burning) and POA from wildfires. Average for all May-months in the 6-year period 2002-2007. Unit: $\mu g\, m^{-3}$.



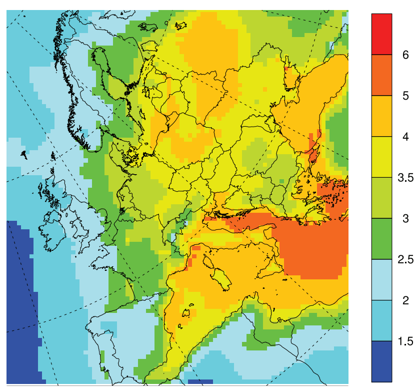
(a) $OM_{PM2.5}$ VBS-NPNA



(b) $OM_{PM2.5}$ VBS-PAP

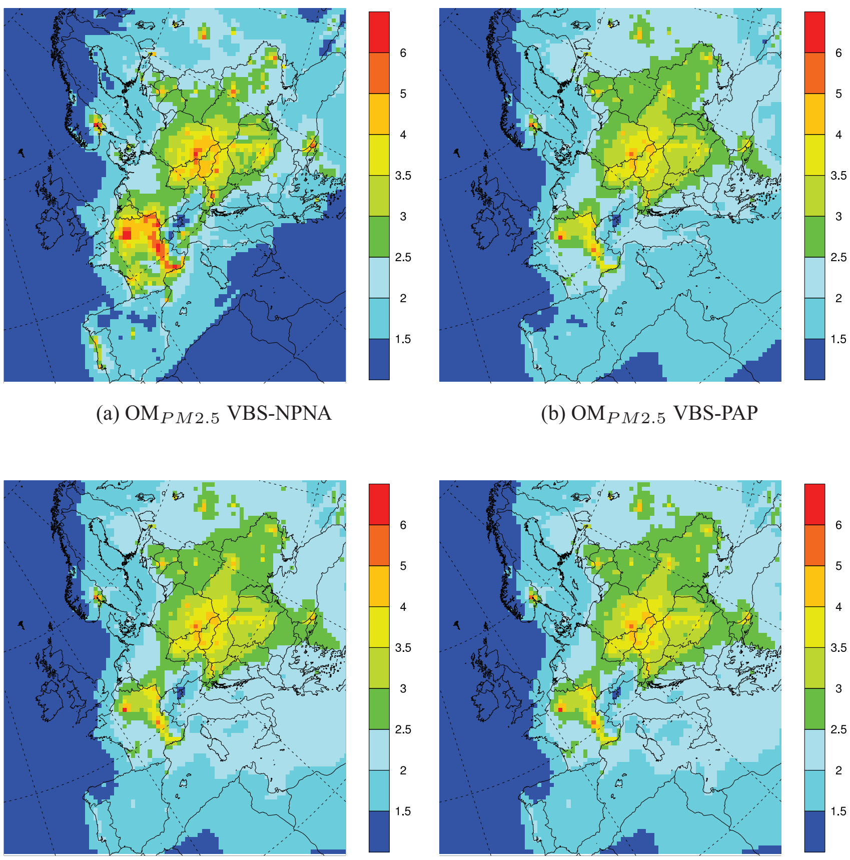


(c) $OM_{PM2.5}$ VBS-PAPA



(d) $OM_{PM2.5}$ VBS-PAA

Fig. A9: Calculated concentration of total organic aerosol in $PM_{2.5}$ for the month May with the four different model versions included in this study (see text). Average for all May-months in the 6-year period 2002-2007. Unit: μg m^{-3}.



(a) $OM_{PM2.5}$ VBS-NPNA

(b) $OM_{PM2.5}$ VBS-PAP

(c) $OM_{PM2.5}$ VBS-PAPA

(d) $OM_{PM2.5}$ VBS-PAA

Fig. A10: Calculated concentration of total organic aerosol 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-year period 2002-2007. Unit: μg m^{-3}.