

Supplementary material:

Light absorbing carbon in Europe. Measurement and modelling, with a focus on residential wood combustion emissions

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Table S1: Comparison of modelled to observed NO₂. Data are from different periods at different stations (daily measurements were available from three stations (Aspvreten, Birkenes and Vavihill); for Harwell, Hyytiälä and Melpitz hourly or half-hourly data were available, here converted to daily [d] and hourly [h] concentrations); N=number of days [or hours] with measurements, Observed=Measured average NO₂-concentration; Observed, Model, Bias and MAE (Mean Absolute Error) are given in µg N/m³. Relative Bias and MAE values are given in parenthesis (relative to the observed mean).

Station	N	Observed	Model	Bias	MAE	r
Aspvreten [d] (SE)	714	0.72	1.03	0.31 (43%)	0.44 (60%)	0.61
Birkenes [d] (NO)	1806	0.41	0.78	0.37 (89%)	0.44 (107%)	0.64
Harwell [d] (GB)	2131	3.41	3.12	-0.29 (-9%)	1.09 (32%)	0.83
Harwell [h]	49796	3.40	3.11	-0.30 (-9%)	1.70 (50%)	0.68
Hyytiälä [d] (FI)	1377	0.74	0.76	0.02 (2%)	0.36 (48%)	0.72
Hyytiälä [h]	32659	0.74	0.76	0.02 (3%)	0.45 (61%)	0.63
Melpitz [d] (DE)	2157	3.23	3.38	0.15 (5%)	1.01 (31%)	0.71
Melpitz [h]	50768	3.24	3.39	0.15 (5%)	1.39 (43%)	0.60
Vavihill [d] (SE)	141	1.33	1.63	0.30 (22%)	0.57 (43%)	0.74

Table S2a: Comparison of modelled EC, with three different assumptions regarding the EC hygroscopicity and atmospheric ageing, to observed EC. N=number of measurements, Obsvd=Measured average EC-concentration, Model (r)=Modelled average EC concentration for the same time periods (r=correlation coefficient), MAE=Mean Absolute Error. Observed, Model, and MAE are given in $\mu\text{g}/\text{m}^3$. Relative MAE values are given in parentheses (relative to the observed mean). Data from the years 2005-2010 but data coverage differs greatly between the stations (see Table 1).

Station	N	Obsvd	FRESH No ageing (all EC externally mixed)		STD Ageing anthropogenic emissions		AGED Hygroscopic emissions	
			Model (r)	MAE	Model (r)	MAE	Model (r)	MAE
Aspvreten (SE)	357	0.25	0.31 (0.67)	0.13(52 %)	0.22 (0.63)	0.11(43 %)	0.21 (0.61)	0.11(44 %)
Birkenes EC _{PM10} (NO)	537	0.13	0.17 (0.68)	0.08(63 %)	0.11 (0.76)	0.06(44 %)	0.096 (0.75)	0.06(47 %)
Birkenes EC _{PM2.5} (NO)	534	0.11	0.16 (0.69)	0.07(69 %)	0.094 (0.71)	0.05(45 %)	0.084 (0.70)	0.05(48 %)
Harwell (GB)	672	0.52	0.50 (0.45)	0.33(65 %)	0.45 (0.45)	0.33(64 %)	0.43 (0.44)	0.33(65 %)
Hyytiälä (FI)	248	0.18	0.25 (0.67)	0.12(66 %)	0.16 (0.71)	0.07(41 %)	0.14 (0.70)	0.07(42 %)
Mace Head (IE)	9	0.11	0.13 (0.92)	0.04(35 %)	0.11 (0.91)	0.04(36 %)	0.11 (0.91)	0.04(37 %)
Melpitz EC _{PM10} (DE)	2157	1.71	0.66 (0.54)	1.09(64 %)	0.53 (0.55)	1.20(70 %)	0.50 (0.55)	1.22(72 %)
Melpitz EC _{PM2.5} (DE)	2100	1.43	0.58 (0.61)	0.89(62 %)	0.45 (0.64)	0.99(70 %)	0.42 (0.64)	1.02(71 %)
Overtoom (NL)	224	0.76	0.97 (0.50)	0.36(48 %)	0.89 (0.51)	0.31(41 %)	0.86 (0.52)	0.30(40 %)
Vavihill (SE)	143	0.19	0.44 (0.64)	0.25(128 %)	0.32 (0.53)	0.14(75 %)	0.30 (0.50)	0.13(67 %)

Table S2b: Comparison of modelled EC (with three different model versions) to observed BC_e. N=number of hours with measurements, Observed=Optical measurements converted to BC_e-concentrations (using the conversions factors in Table 2); Observed, Model, Bias and MAE (Mean Absolute Error) are given in $\mu\text{g}/\text{m}^3$. Relative Bias and MAE values are given in parentheses (relative to the observed mean). Data from the years 2005-2010 but data coverage differs greatly between the stations (see Table 1).

Station	N	Observed	FRESH No ageing (all EC treated as externally mixed)		STD Ageing anthropogenic emissions		AGED Hygroscopic emissions	
			Model (r)	MAE	Model (r)	MAE	Model (r)	MAE
Aspvreten (SE)	18573	0.22	0.28 (0.24)	0.21 (93 %)	0.20 (0.17)	0.17 (78 %)	0.18 (0.16)	0.17 (78 %)
Birkenes (NO)	13364	0.096	0.15 (0.44)	0.093 (97 %)	0.078 (0.34)	0.065 (67 %)	0.068 (0.31)	0.067 (70 %)
Harwell (GB)	9593	0.82	0.57 (0.65)	0.39 (48 %)	0.48 (0.65)	0.42 (51 %)	0.45 (0.63)	0.43 (53 %)
Hyytiälä (FI)	51188	0.17	0.27 (0.68)	0.13 (77 %)	0.19 (0.65)	0.094 (55 %)	0.18 (0.65)	0.093 (54 %)
Mace Head (IE)	27590	0.14	0.15 (0.67)	0.079 (56 %)	0.11 (0.67)	0.070 (50 %)	0.11 (0.67)	0.072 (51 %)
Melpitz (DE)	29212	1.36	0.64 (0.68)	0.78 (57 %)	0.50 (0.66)	0.88 (65 %)	0.47 (0.65)	0.90 (67 %)
Vavihill (SE)	10506	0.16	0.46 (0.57)	0.31 (199 %)	0.35 (0.44)	0.22 (143 %)	0.33 (0.40)	0.21 (138 %)

Table S3a: Comparison of observed EC to modelled EC during winter half-year periods (defined as Jan-Apr+Nov-Dec) using two different emission inventories (EUCAARI and TNO new RWC) for residential wood combustion (RWC). Observed=Measured average EC-concentration (number of measurements); Observed, Model, Bias and MAE (Mean Absolute Error) are given in $\mu\text{g}/\text{m}^3$. Relative Bias and MAE values are given in parenthesis (relative to the observed mean). Data from the years 2005-2010 but data coverage differs greatly between the stations (see Table 1).

Station	EUCAARI emissions					TNO new RWC emissions (STD = Base case)			
	Observed	Model	Bias	MAE	r	Model	Bias	MAE	r
Aspvreten (SE)	0.32 (155)	0.26	-0.058 (-18 %)	0.15 (47 %)	0.50	0.29	-0.030(-10 %)	0.15 (47 %)	0.50
Birkenes EC _{PM10} (NO)	0.13 (257)	0.15	0.013 (10 %)	0.065 (49 %)	0.70	0.11	-0.023 (-17 %)	0.059 (44 %)	0.74
Birkenes EC _{PM2.5} (NO)	0.12 (255)	0.13	0.015 (12 %)	0.054 (46 %)	0.68	0.097	-0.021 (-18%)	0.049 (42 %)	0.69
Harwell (GB)	0.65 (343)	0.46	-0.19 (-30 %)	0.41 (63 %)	0.40	0.47	-0.18 (-28 %)	0.41 (63 %)	0.40
Hyytiälä (FI)	0.24 (129)	0.17	-0.071 (-30 %)	0.10 (43 %)	0.73	0.21	-0.028 (-12 %)	0.10 (44 %)	0.69
Mace Head (IE)	0.11 (4)	0.12	0.012 (11 %)	0.039 (36 %)	0.98	0.12	0.016 (15 %)	0.042 (39 %)	0.98
Melpitz EC _{PM10} (DE)	1.92 (1081)	0.60	-1.32 (-69 %)	1.36 (71 %)	0.54	0.65	-1.27 (-66 %)	1.31 (68 %)	0.53
Melpitz EC _{PM2.5} (DE)	1.73 (1074)	0.51	-1.22 (-71 %)	1.24 (72 %)	0.65	0.56	-1.17 (-68 %)	1.19 (69 %)	0.64
Overtoom (NL)	0.83 (104)	0.92	0.087 (10 %)	0.38 (45 %)	0.42	0.99	0.16 (19 %)	0.42 (50 %)	0.41
Vavihill (SE)	0.25 (65)	0.40	0.15 (61 %)	0.19 (76 %)	0.26	0.41	0.16 (63 %)	0.19 (76 %)	0.33

Table S3b: Comparison of observed BC_e (BC_e=EC equivalent BC, see text) to modelled EC during winter half-year periods (defined as Jan-Apr+Nov-Dec) using two different emission inventories (EUCAARI and TNO new RWC) for residential wood combustion. Observed=Optical measurements converted to BC_e [using the MAC in Table 2] (number of hours with measurements); Observed, Model, Bias and MAE (Mean Absolute Error) are given in $\mu\text{g}/\text{m}^3$. Relative Bias and MAE values are given in parentheses (relative to the observed mean). Data from the years 2005-2010 but data coverage differs greatly between the stations (see Table 1).

Station	EUCAARI emissions					TNO new RWC emissions (STD = Base case)			
	Observed	Model	Bias	MAE	r	Model	Bias	MAE	r
Aspvreten (SE)	0.22 (6509)	0.24	0.02 (11 %)	0.16 (73 %)	0.36	0.27	0.05 (22 %)	0.17 (79 %)	0.35
Birkenes (NO)	0.10 (6166)	0.13	0.03 (29 %)	0.09 (88 %)	0.42	0.09	-0.01 (-8 %)	0.068 (67 %)	0.49
Harwell (GB)	0.80 (5177)	0.55	-0.26 (-32 %)	0.40 (50 %)	0.61	0.56	-0.25 (-31 %)	0.40 (49 %)	0.61
Hyytiälä (FI)	0.22 (25521)	0.21	-0.001 (-0.7 %)	0.11 (51 %)	0.63	0.26	0.05 (21 %)	0.13 (60 %)	0.61
Mace Head (IE)	0.23 (14141)	0.12	-0.11 (-47 %)	0.12 (54 %)	0.73	0.12	-0.10 (-46 %)	0.12 (53 %)	0.73
Melpitz (DE)	1.84 (14822)	0.57	-1.27 (-69 %)	1.28 (70 %)	0.65	0.62	-1.22 (-66 %)	1.24 (67 %)	0.65
Vavihill (SE)	0.24 (5647)	0.42	0.18 (74 %)	0.27 (110 %)	0.38	0.42	0.18 (76 %)	0.26 (107 %)	0.42
Aspvreten (SE)	0.22 (6509)	0.24	0.02 (11 %)	0.16 (73 %)	0.36	0.27	0.05 (22 %)	0.17 (79 %)	0.35
Birkenes (NO)	0.10 (6166)	0.13	0.03 (29 %)	0.09 (88 %)	0.42	0.09	-0.01 (-8 %)	0.068 (67 %)	0.49
Harwell (GB)	0.80 (5177)	0.55	-0.26 (-32 %)	0.40 (50 %)	0.61	0.56	-0.25 (-31 %)	0.40 (49 %)	0.61

The Figs. S1-S8 show time series plots for EC. Measured and modelled concentrations with two different ageing assumptions are included (FRESH=All EC emitted as hydrophobic and no atmospheric ageing of EC; STD=Standard model version, including atmospheric ageing of EC) [Note: model data only shown for periods with measurements.]

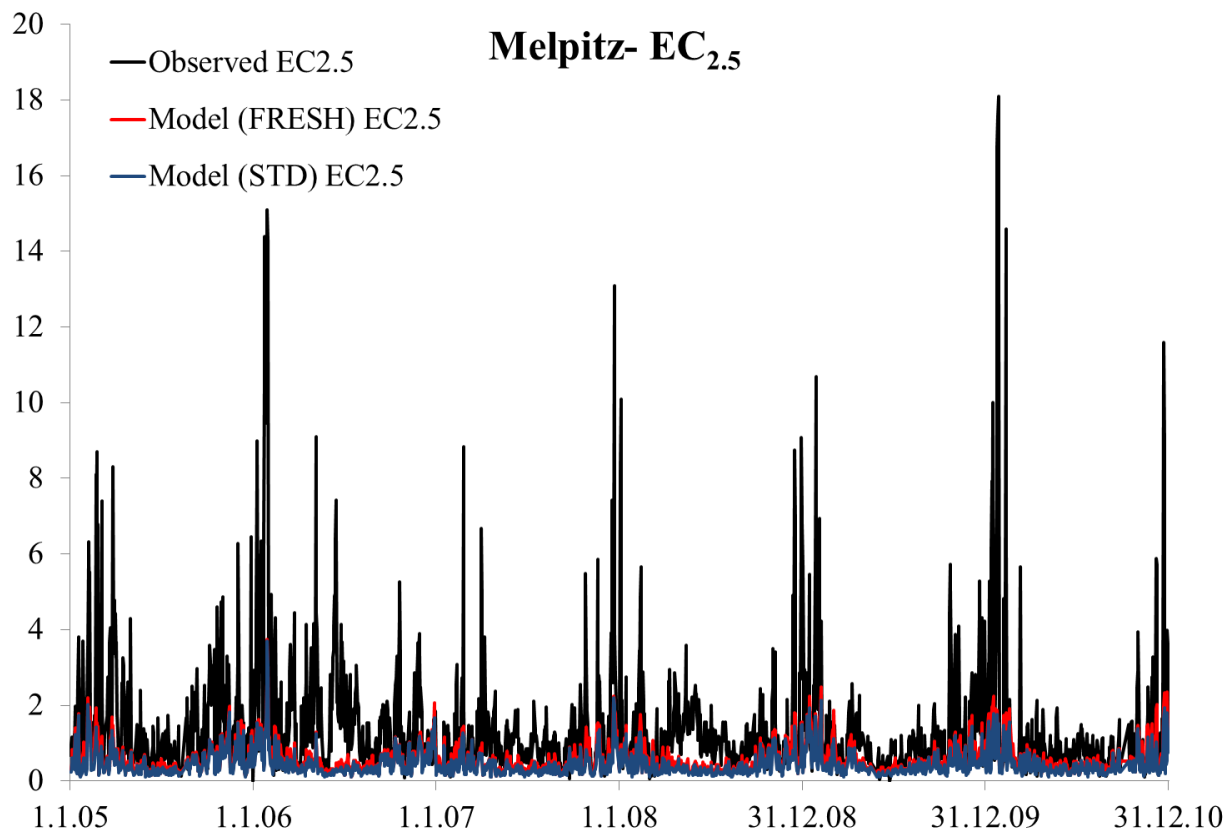


Figure S1. Measured and modelled EC_{2.5} concentrations at Melpitz. Unit: $\mu\text{g}/\text{m}^3$.

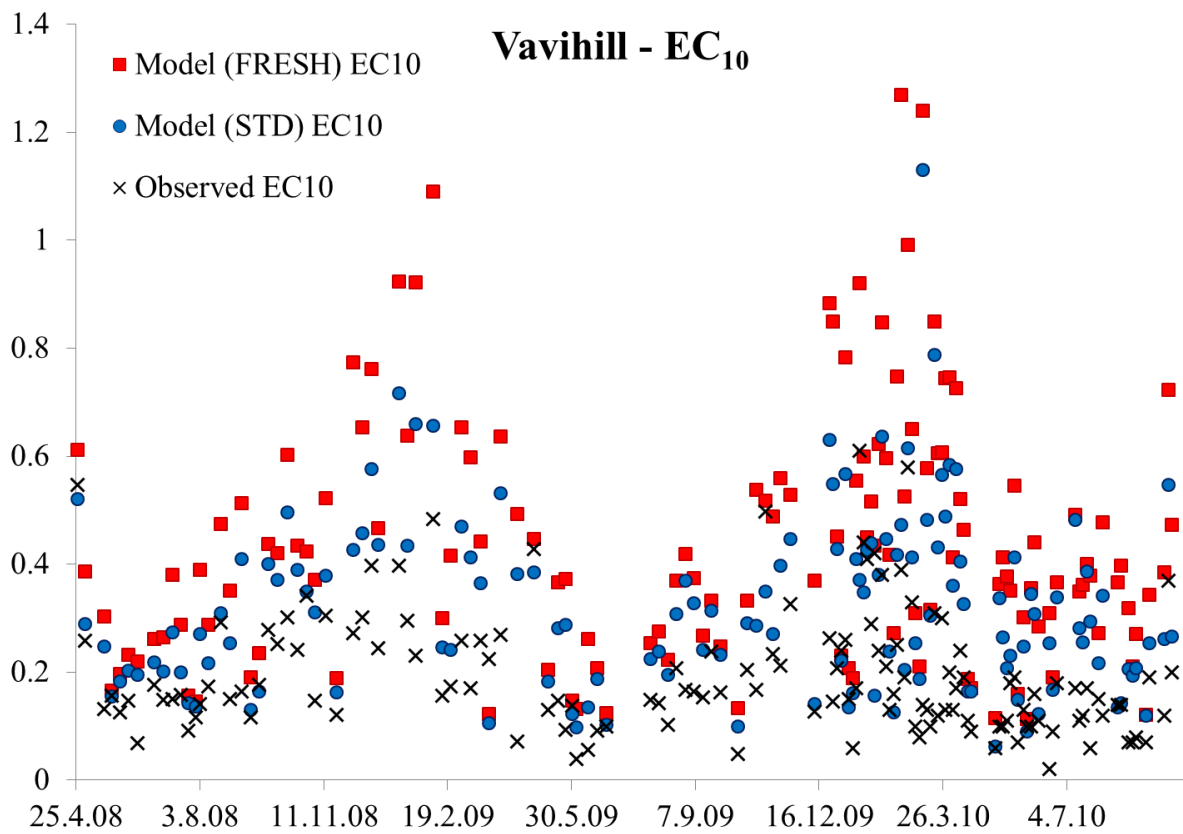


Figure S2. Measured and modelled EC₁₀ concentrations at Vavihill. Unit: $\mu\text{g}/\text{m}^3$.

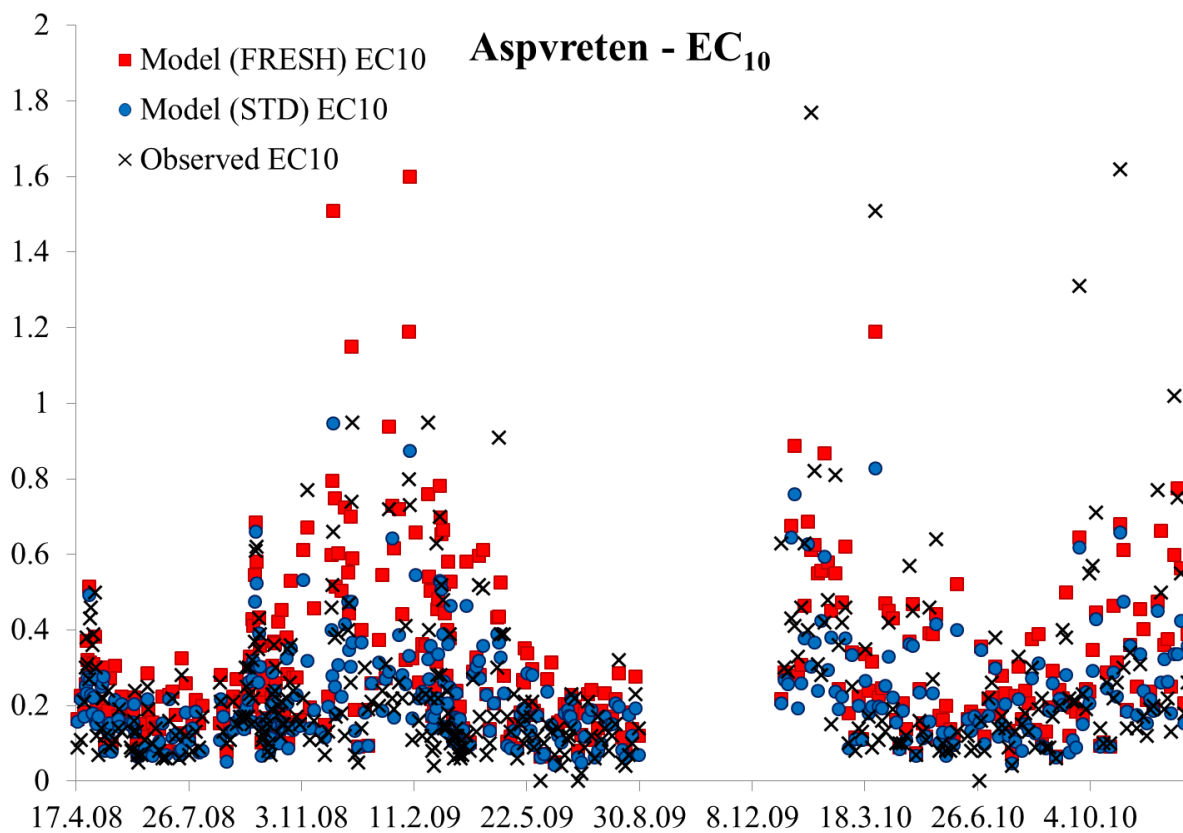


Figure S3. Measured and modelled EC₁₀ concentrations at Aspvreten. Unit: $\mu\text{g}/\text{m}^3$.

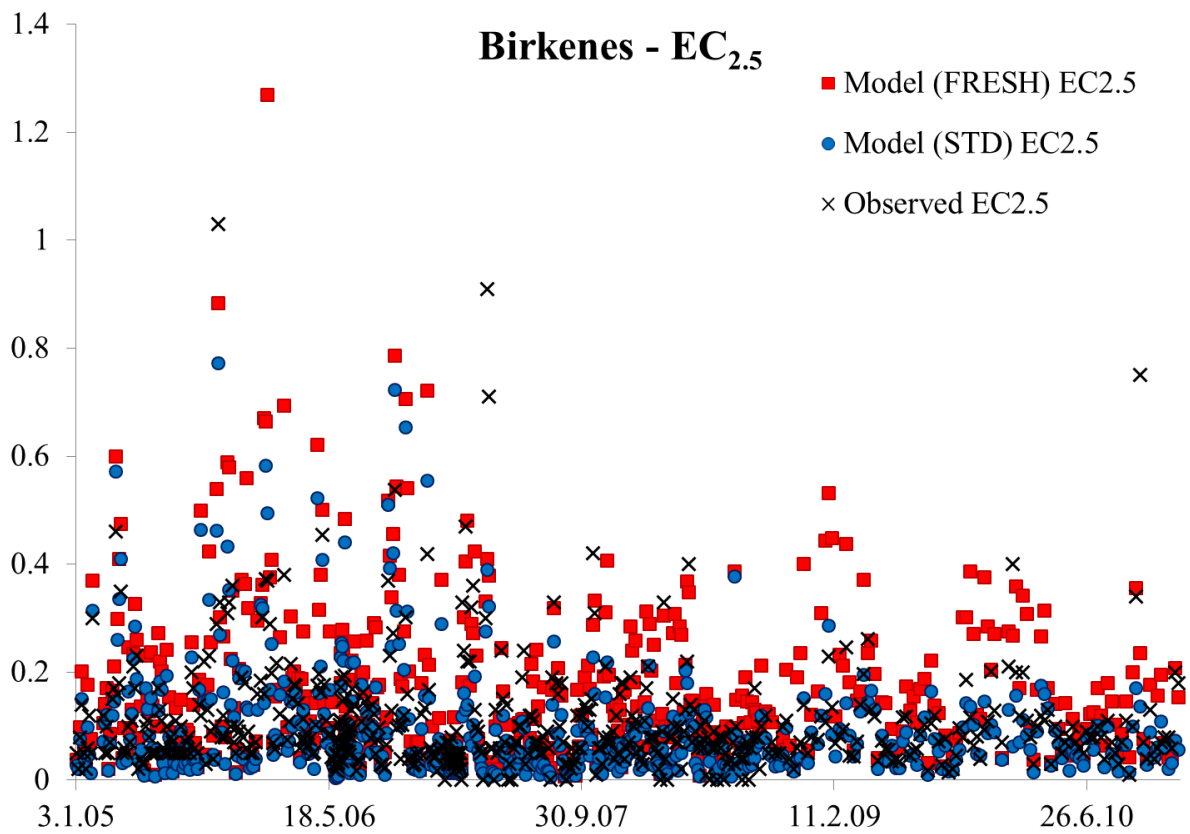


Figure S4. Measured and modelled EC_{2.5} concentrations at Birkenes. Unit: $\mu\text{g}/\text{m}^3$.

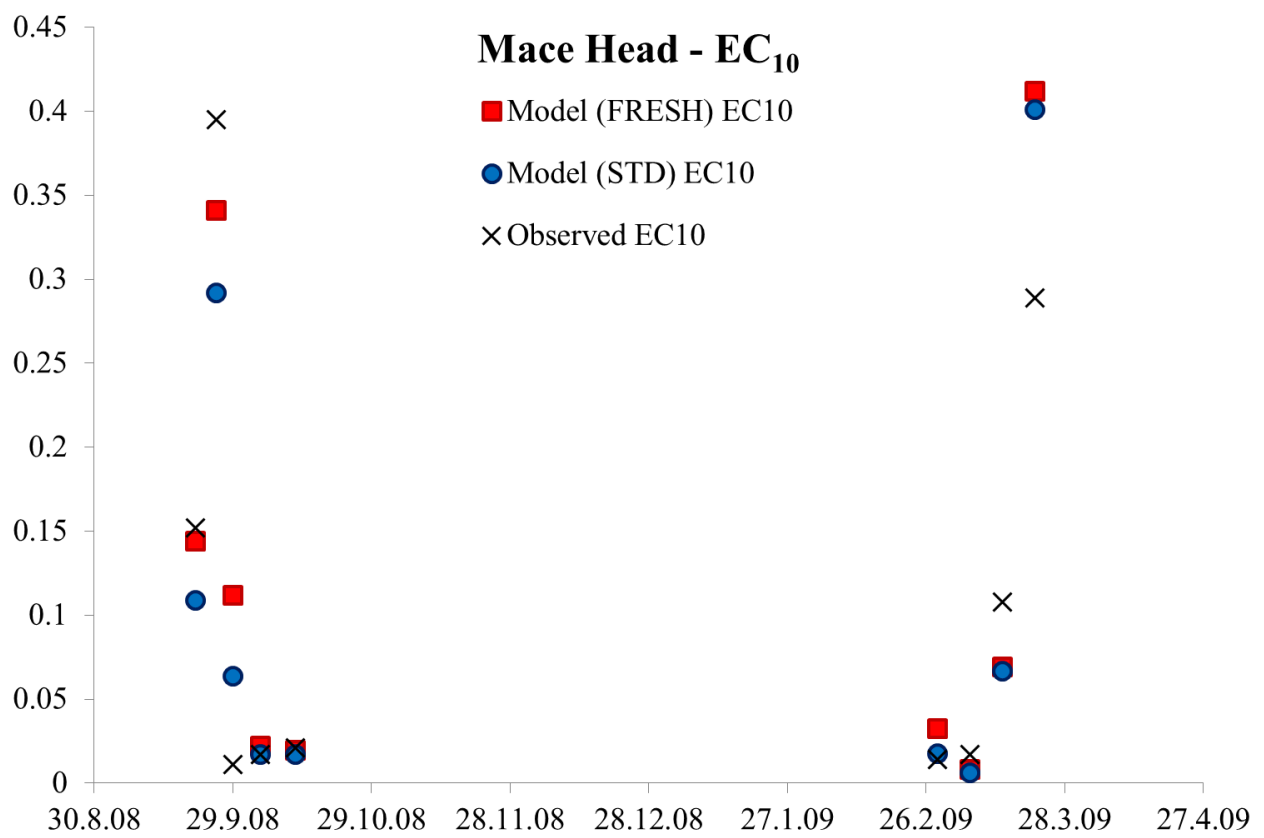


Figure S5. Measured and modelled EC₁₀ concentrations at Mace Head. Unit: $\mu\text{g}/\text{m}^3$.

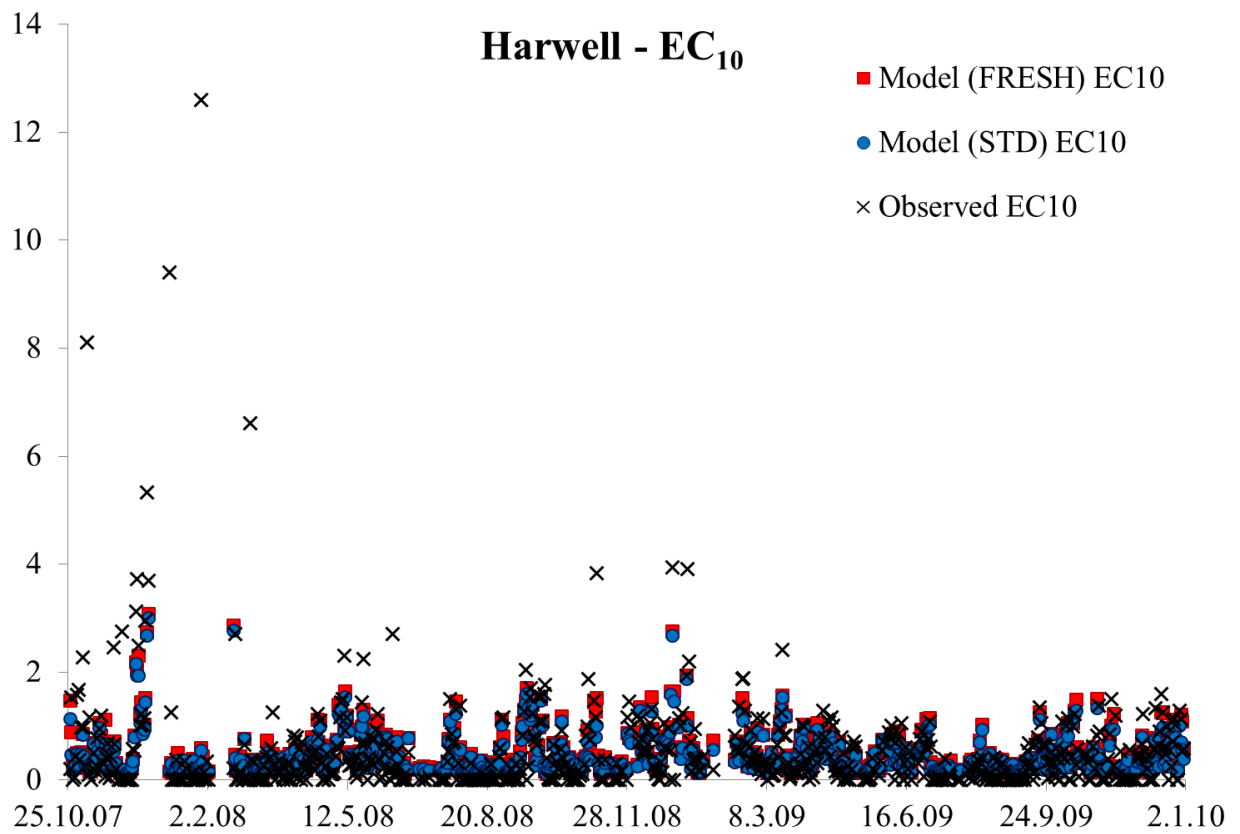


Figure S6. Measured and modelled EC₁₀ concentrations at Harwell. Unit: µg/m³.

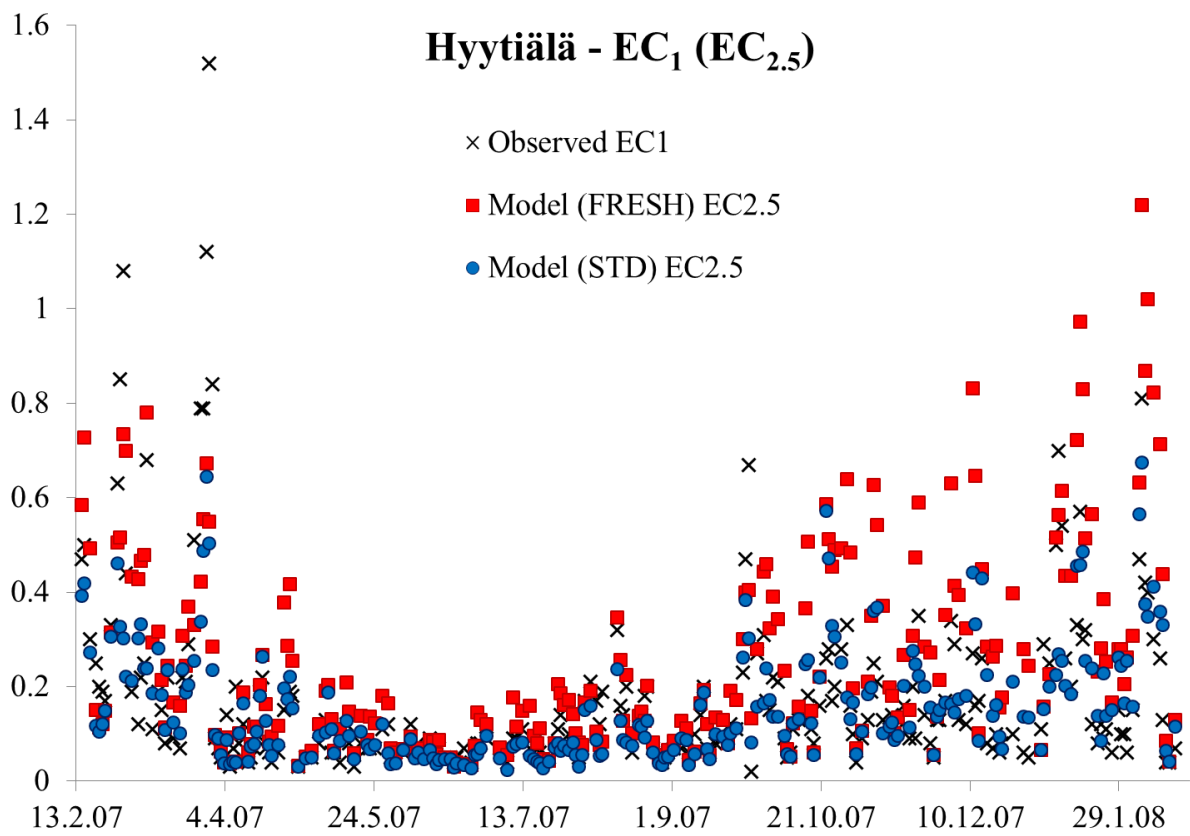


Figure S7. Measured EC₁ concentrations and modelled EC_{2.5} at Hyytiälä. Unit: µg/m³.

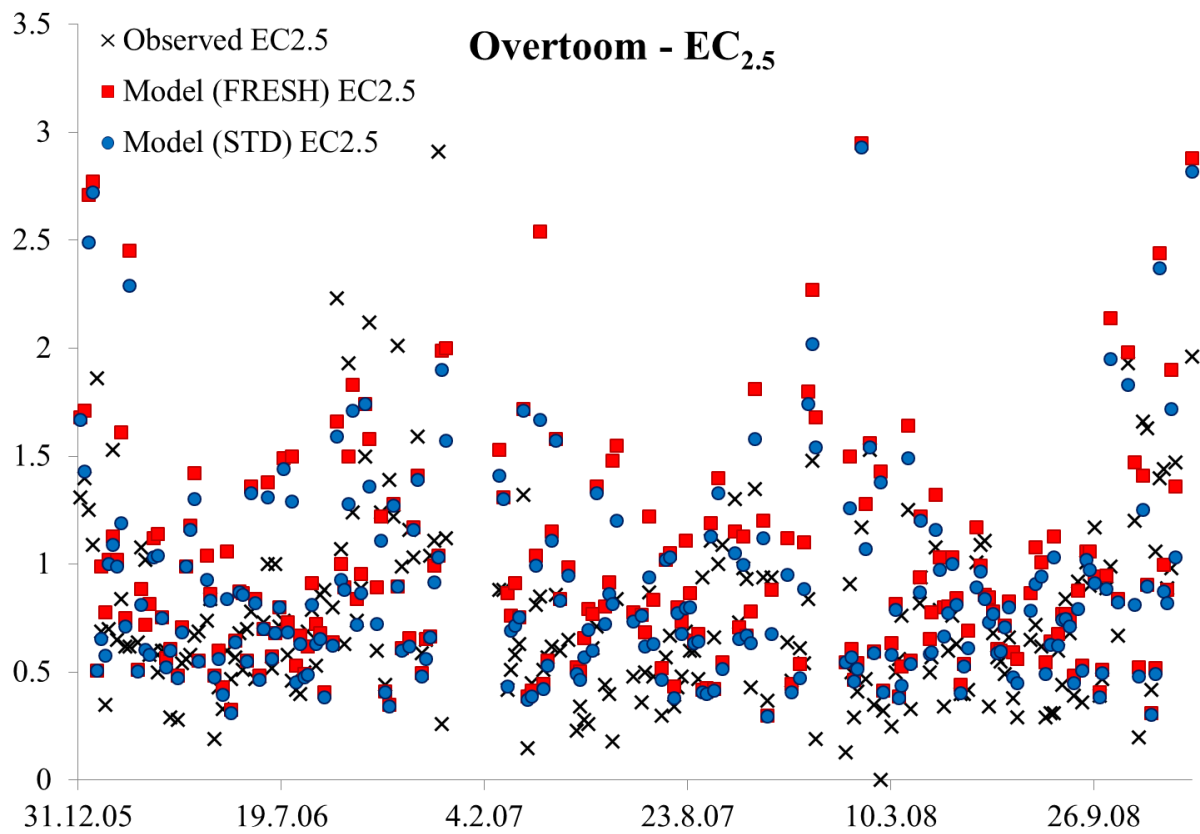


Figure S8. Measured and modelled EC_{2.5} concentrations at Overtoom. Unit: $\mu\text{g}/\text{m}^3$.

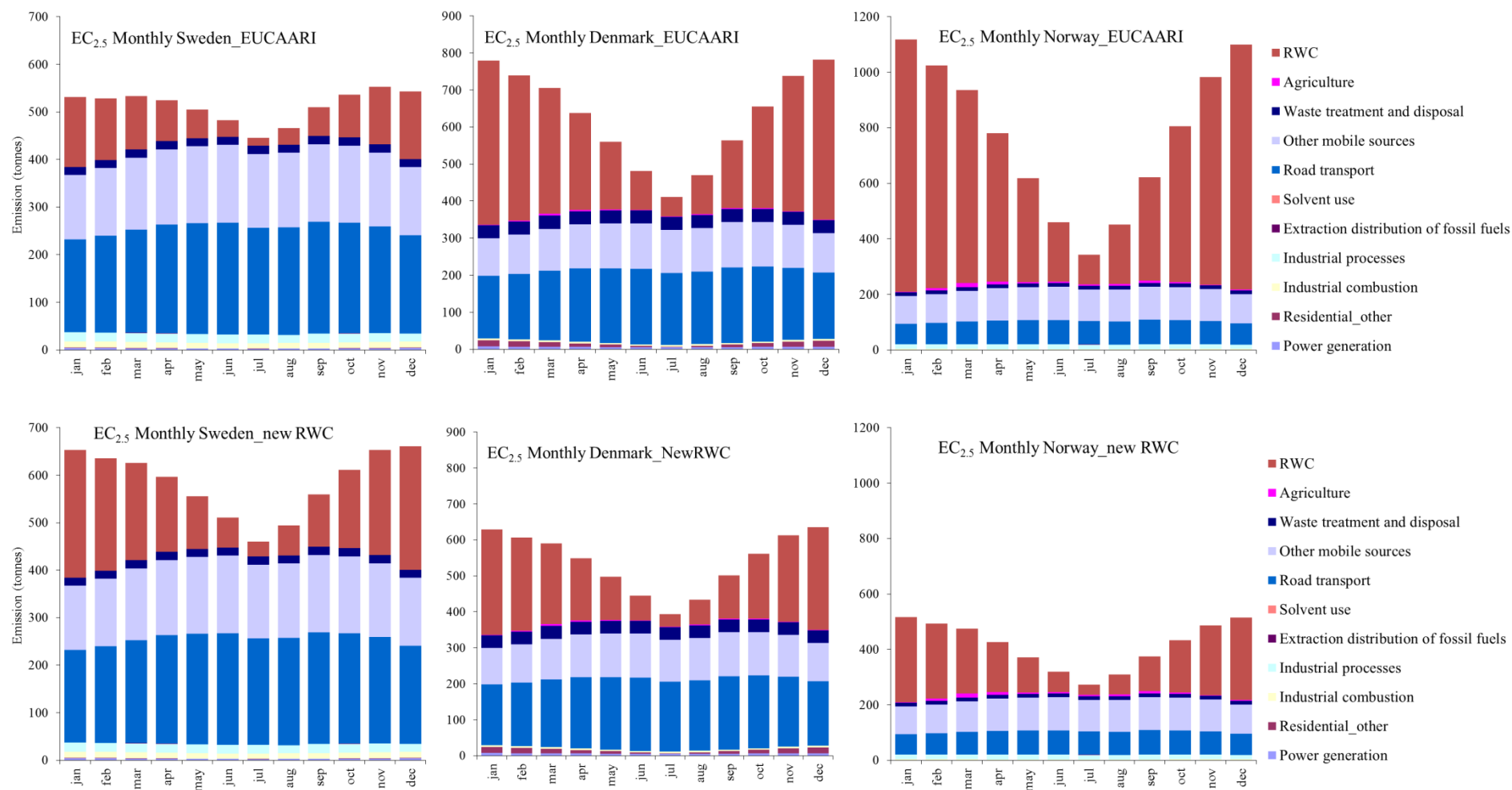


Figure S9. Monthly variation of EC emissions from different sources in the Scandinavian countries (Sweden, Denmark and Norway) with two different emission inventories EUCAARI (top) and the new TNO inventory for Residential Wood Combustion (bottom). Note that only the RWC emissions (the red top part of each monthly bar) are different in the two inventories and that the y-axis levels have been fixed by country to illustrate absolute emission changes. Unit: tonnes.

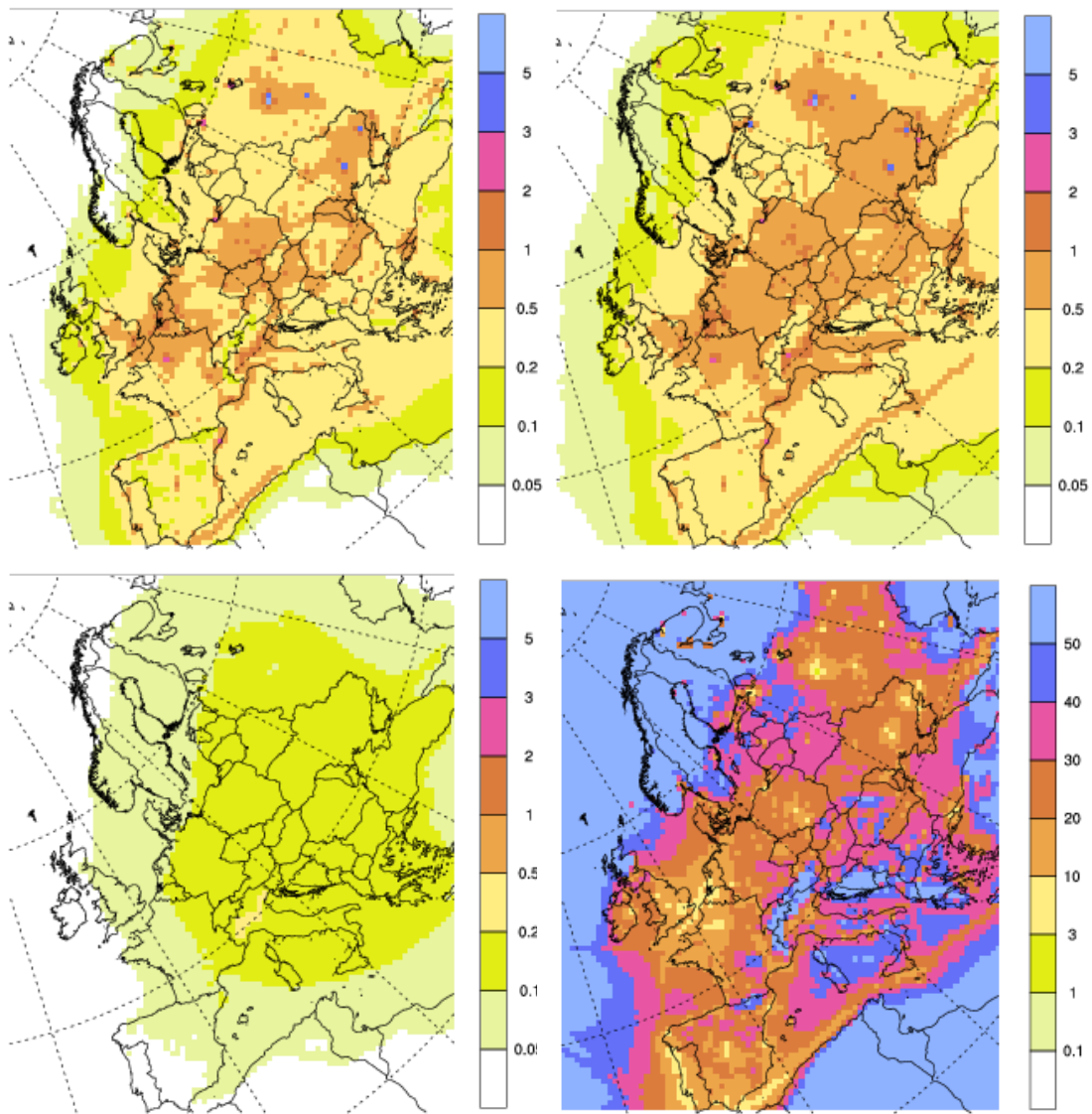


Fig. S10: Calculated EC₁₀-concentration (six-year average, 2005-2010) with the Standard model assumptions regarding ageing (STD, upper left), assuming no ageing of EC after emission (FRESH, upper right) and the difference, No ageing - Standard ageing, (FRESH - STD, lower left) and relative difference in EC₁₀ between FRESH and STD runs. Unit: $\mu\text{g}/\text{m}^3$, except for the relative difference, which is in %.