

Supplement of: Simulating organic aerosol in Delhi with WRF-Chem using the VBS approach: Exploring model uncertainty with a Gaussian Process emulator

S1. WRF-Chem setup.

Table S1 shows an example of the parameters used to control the VBS scheme when building the model setup for anthropogenic and biomass burning sources. 111 namelist.input files were designed using the ranges of the 10 parameters in table 2.

Table S1: Example of a namelist.input file with parameters to control the VBS scheme.

Parameter	Value
Biomass Burning _VBS_ scaling	1
Anthropogenic VBS scaling	1
Biomass Burning VBS ageing rate	1.00E-13
Anthropogenic VBS ageing rate	1.00E-13
Biomass Burning VBS Oxidation rate	0.075
Anthropogenic _VBS_ Oxidation rate	0.075
Biomass Burning VBS FRAC_1	0
Biomass Burning VBS FRAC_2	0.12
Biomass Burning VBS FRAC_3	0.24
Biomass Burning VBS FRAC_4	0.24
Biomass Burning VBS FRAC_5	0.21
Biomass Burning VBS FRAC_6	0.13
Biomass Burning VBS FRAC_7	0.04
Biomass Burning VBS FRAC_8	0
Biomass Burning VBS FRAC_9	0
Anthropogenic VBS FRAC_1	0
Anthropogenic VBS FRAC_2	0.12
Anthropogenic VBS FRAC_3	0.24
Anthropogenic VBS FRAC_4	0.24
Anthropogenic VBS FRAC_5	0.21
Anthropogenic VBS FRAC_6	0.13
Anthropogenic VBS FRAC_7	0.04
Anthropogenic VBS FRAC_8	0
Anthropogenic VBS FRAC_9	0

Ageing rate is the reaction rate for all VBS reactions in that scheme

Oxidation rate is the fractional increase in oxidation of the VBS compounds per reaction step

FRAC[1-9] is the multiplier from the POA mass in the emission database, to give the emitted mass of VBS component in that volatility bin.

scaling is a scaling factor applied to all **FRAC[1-9]** values for that scheme, usually with the aim of ensuring that the condensed VBS mass at time of emission is roughly equivalent to the involatile POA mass in emission database used.

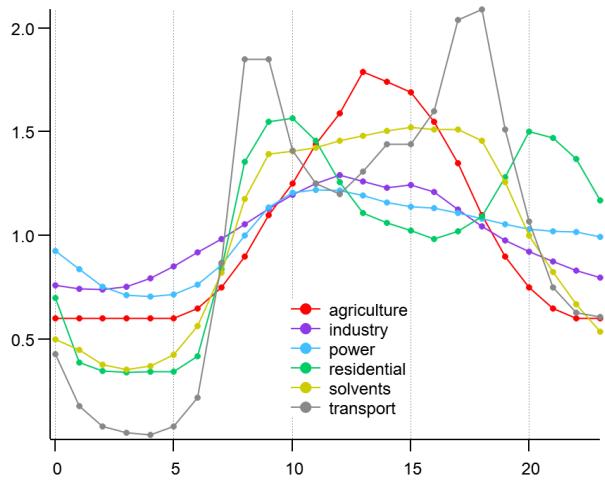


Figure S1 Diurnal fraction of seven activities used in WRF-Chem. Taken from (Olivier et al., 2003)

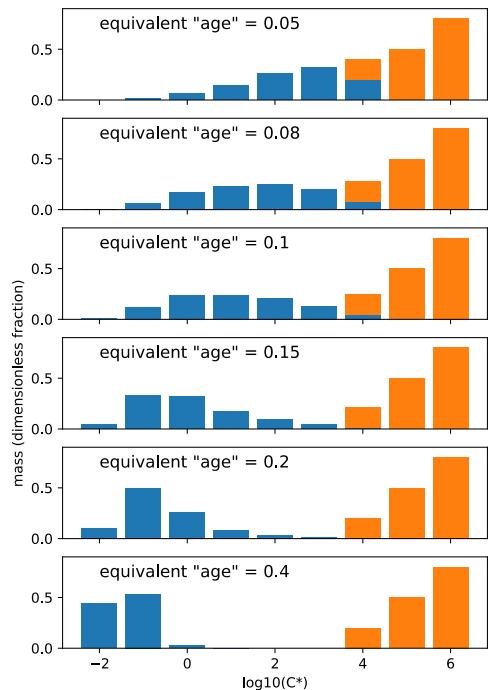


Figure S2 Example volatility distributions for emissions. Orange bars indicate IVOC emissions (with a fixed distribution). Blue bars indicate SVOC emissions, with volatility distributions calculated using the “equivalent age” calculation described in Section 2.3. For this plot both the IVOC and SVOC scaling factors are set to 1.

S2. Observations

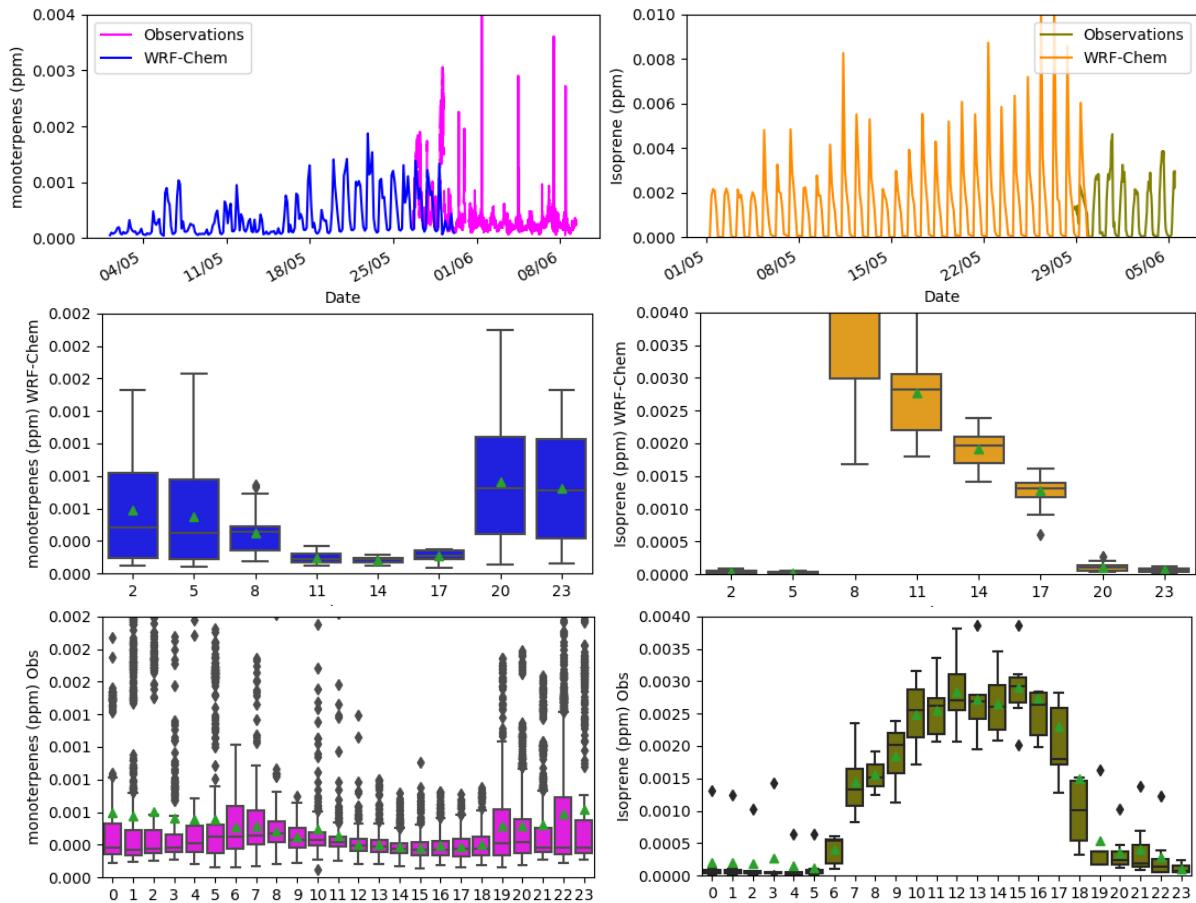


Figure S3. Monoterpenes (left) and isoprene (right) time series and diurnal cycles of WRF-Chem outputs and observations. Notice the different start-end times of model and observations. Triangles highlight the mean.

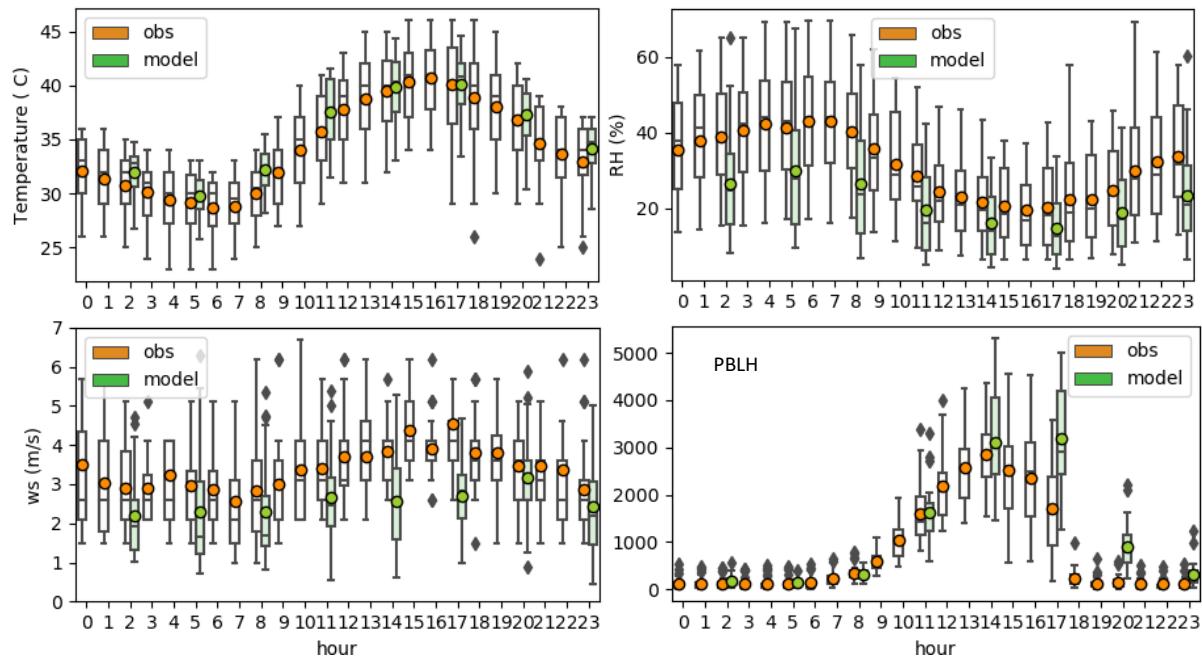


Figure S4. Diurnal cycles of temperature, RH, ws and PBLH. May – 2018. Circles highlight the mean.

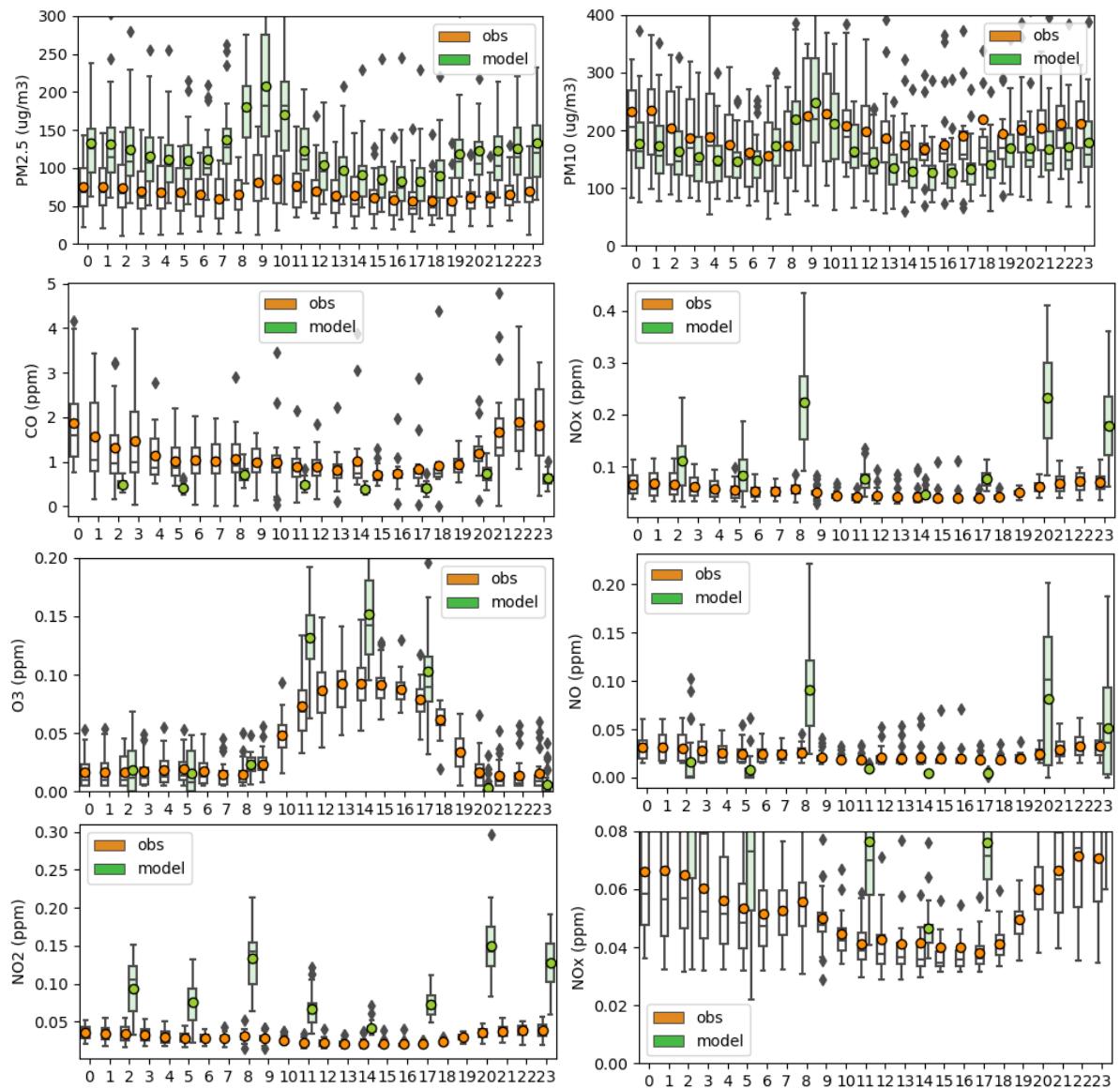


Figure S5. Diurnal cycles of particulate matter and gaseous species. Observations and model outputs. May 2018. The circles highlight the mean.

S3. WRF-Chem model evaluation

The parameters used for model evaluation were calculated with the OpenAir package (Carslaw and Ropkins, 2012). The following equations were extracted from the OpenAir manual, where O_i represents the i^{th} observed value and M_i represents the i^{th} modelled value for a total of n observations.

Fraction of predictions within a factor of two, FAC2

FAC2 is the fraction of modelled values within a factor of two of observations, which satisfy:

$$0.5 \leq \frac{M_i}{O_i} \leq 2.0$$

Mean bias (MB).

MB gives an indication of the mean over or underestimate of predictions; it has the same units as the quantities being considered.

$$MB = \frac{1}{n} \sum_{i=1}^n M_i - O_i$$

Index of agreement (IOA).

The IOA is commonly used in model evaluation (Willmott et al., 2012), ranging between -1 and +1, with values close to +1 representing a better model performance. An IOA of 0.5 indicates that the sum of the error magnitudes is one-half of the sum of the observed-deviation magnitudes. IOA, with $c = 2$, is defined as:

$$IOA = 1.0 - \frac{\sum_{i=1}^n |M_i - O_i|}{c \sum_{i=1}^n |O_i - \bar{o}|}, \text{ when}$$

$$\sum_{i=1}^n |M_i - O_i| \leq c \sum_{i=1}^n |O_i - \bar{o}|$$

$$IOA = \frac{c \sum_{i=1}^n |O_i - \bar{o}|}{\sum_{i=1}^n |M_i - O_i|} - 1.0, \text{ when}$$

$$\sum_{i=1}^n |M_i - O_i| > c \sum_{i=1}^n |O_i - \bar{o}|$$

S4. Comparison of the model runs with AMS observations

Table S2. List of 111 WRF-Chem model runs

*61 TRAIN runs				*20 VALID runs		*30 new TRAIN runs	
TRAIN000	TRAIN015	TRAIN030	TRAIN045	VALID000	VALID015	TRAIN100	TRAIN115
TRAIN001	TRAIN016	TRAIN031	TRAIN046	VALID001	VALID016	TRAIN101	TRAIN116
TRAIN002	TRAIN017	TRAIN032	TRAIN047	VALID002	VALID017	TRAIN102	TRAIN117
TRAIN003	TRAIN018	TRAIN033	TRAIN048	VALID003	VALID018	TRAIN103	TRAIN118
TRAIN004	TRAIN019	TRAIN034	TRAIN049	VALID004	VALID019	TRAIN104	TRAIN119
TRAIN005	TRAIN020	TRAIN035	TRAIN050	VALID005		TRAIN105	TRAIN120
TRAIN006	TRAIN021	TRAIN036	TRAIN051	VALID006		TRAIN106	TRAIN121
TRAIN007	TRAIN022	TRAIN037	TRAIN052	VALID007		TRAIN107	TRAIN122
TRAIN008	TRAIN023	TRAIN038	TRAIN053	VALID008		TRAIN108	TRAIN123
TRAIN009	TRAIN024	TRAIN039	TRAIN054	VALID009		TRAIN109	TRAIN124
TRAIN010	TRAIN025	TRAIN040	TRAIN055	VALID010		TRAIN110	TRAIN125
TRAIN011	TRAIN026	TRAIN041	TRAIN056	VALID011		TRAIN111	TRAIN126
TRAIN012	TRAIN027	TRAIN042	TRAIN057	VALID012		TRAIN112	TRAIN127
TRAIN013	TRAIN028	TRAIN043	TRAIN058	VALID013		TRAIN113	TRAIN128
TRAIN014	TRAIN029	TRAIN044	TRAIN059	VALID014		TRAIN114	TRAIN129
			TRAIN060				

*Anthropogenic SVOC scaling min = 0.5 and max = 4

+Anthropogenic SVOC scaling min = 0.1 and max = 0.5

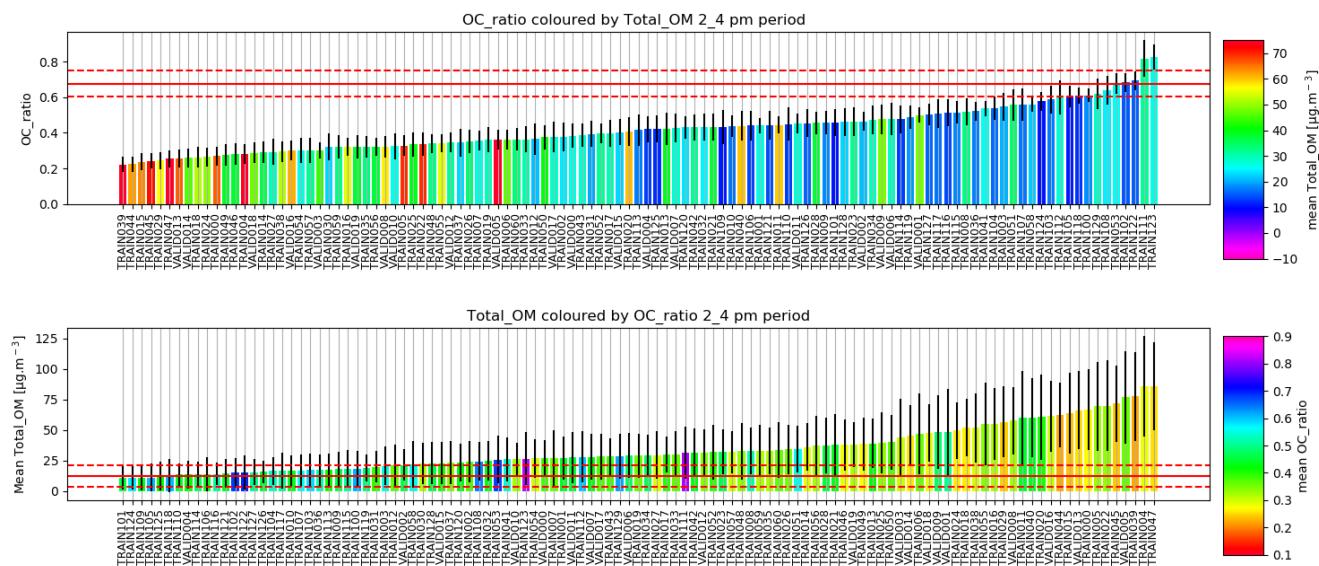


Figure S6. Analysis of the 111 model runs for the 2-4 pm period. Mean OC_ratio coloured by mean Total_OM (top) and mean Total_OM coloured by mean OC_ratio (bottom). The red line highlights the mean and SD of AMS observations (O:C top and OA bottom). The mean AMS values are O:C = 0.67 and OA = 12.20 $\mu\text{g.m}^{-3}$.

Table S3 Evaluation of the ensemble of 111 model runs ordered from high to low FAC2 values with O:C and OA for the 2-4 pm period.

2-4pm_O:C							2-4pm_OA								
model	FAC2	MB	IOA	model	FAC2	MB	IOA	model	FAC2	MB	IOA	model	FAC2	MB	IOA
TRAIN036	1.00	0.03	0.51	TRAIN050	0.86	-0.13	0.36	TRAIN127	0.73	4.37	0.44	TRAIN118	0.41	-2.39	0.36
TRAIN114	1.00	0.04	0.45	TRAIN028	0.85	-0.10	0.45	VALID004	0.72	3.30	0.51	TRAIN052	0.41	25.62	-0.12
TRAIN127	0.99	0.02	0.51	TRAIN019	0.85	-0.12	0.39	TRAIN121	0.72	1.02	0.48	TRAIN008	0.41	25.93	-0.13
TRAIN101	0.99	0.02	0.48	TRAIN011	0.85	-0.13	0.37	TRAIN126	0.72	4.35	0.43	TRAIN027	0.40	24.30	-0.08
TRAIN117	0.99	0.04	0.48	TRAIN105	0.85	0.15	0.25	TRAIN110	0.70	2.03	0.53	TRAIN105	0.40	-3.58	0.37
TRAIN115	0.99	0.05	0.46	TRAIN040	0.84	-0.08	0.46	TRAIN125	0.70	0.27	0.50	TRAIN124	0.38	-3.99	0.40
TRAIN121	0.98	0.00	0.52	VALID010	0.84	-0.14	0.34	TRAIN010	0.70	6.93	0.49	TRAIN051	0.38	27.77	-0.19
TRAIN106	0.98	0.00	0.52	TRAIN118	0.84	0.14	0.29	TRAIN009	0.70	8.94	0.43	TRAIN033	0.38	28.20	-0.19
TRAIN120	0.98	-0.04	0.52	TRAIN060	0.83	-0.14	0.33	TRAIN103	0.70	3.13	0.42	TRAIN035	0.38	28.62	-0.21
TRAIN119	0.98	0.01	0.51	TRAIN007	0.83	-0.16	0.25	TRAIN113	0.69	3.90	0.45	TRAIN057	0.37	27.82	-0.20
TRAIN109	0.98	0.02	0.50	TRAIN033	0.81	-0.13	0.37	TRAIN036	0.69	5.13	0.40	TRAIN007	0.37	27.76	-0.22
TRAIN126	0.98	0.01	0.50	TRAIN035	0.81	-0.16	0.27	TRAIN031	0.69	9.43	0.40	TRAIN026	0.37	30.73	-0.26
TRAIN058	0.98	0.05	0.48	TRAIN054	0.81	-0.16	0.27	TRAIN117	0.68	1.27	0.47	VALID011	0.35	29.12	-0.23
TRAIN104	0.98	0.03	0.47	TRAIN048	0.81	-0.17	0.24	TRAIN104	0.68	5.50	0.47	TRAIN059	0.35	30.79	-0.26
TRAIN003	0.98	0.04	0.46	TRAIN053	0.81	0.18	0.20	TRAIN116	0.68	2.09	0.43	TRAIN060	0.35	31.58	-0.28
TRAIN034	0.96	-0.02	0.54	VALID008	0.81	-0.18	0.19	TRAIN115	0.68	3.27	0.39	TRAIN021	0.33	28.24	-0.22
TRAIN041	0.96	-0.03	0.52	TRAIN108	0.81	0.16	0.17	TRAIN107	0.68	3.14	0.38	TRAIN048	0.32	31.49	-0.28
TRAIN128	0.96	-0.02	0.52	TRAIN027	0.81	-0.15	0.30	TRAIN012	0.67	10.89	0.39	VALID012	0.32	32.48	-0.30
VALID006	0.96	-0.01	0.51	TRAIN026	0.80	-0.15	0.31	TRAIN119	0.67	7.12	0.35	TRAIN014	0.30	33.28	-0.32
VALID002	0.96	-0.04	0.51	TRAIN030	0.80	-0.15	0.30	TRAIN058	0.67	8.52	0.33	TRAIN025	0.30	35.41	-0.36
TRAIN008	0.96	-0.01	0.49	VALID012	0.80	-0.16	0.27	VALID002	0.65	11.31	0.36	TRAIN049	0.28	37.63	-0.40
TRAIN125	0.96	-0.02	0.49	TRAIN025	0.80	-0.16	0.27	TRAIN106	0.64	0.92	0.44	TRAIN056	0.27	34.49	-0.34
TRAIN110	0.96	0.02	0.47	VALID019	0.80	-0.17	0.24	TRAIN120	0.63	11.83	0.33	TRAIN013	0.27	35.64	-0.36
TRAIN009	0.95	-0.04	0.52	TRAIN059	0.80	-0.17	0.22	TRAIN037	0.63	12.46	0.32	TRAIN046	0.26	38.32	-0.41
TRAIN031	0.95	-0.07	0.51	TRAIN006	0.80	-0.17	0.21	TRAIN003	0.63	9.95	0.31	VALID019	0.26	38.55	-0.41
TRAIN051	0.95	0.02	0.49	VALID005	0.80	-0.19	0.17	TRAIN053	0.63	10.41	0.28	TRAIN028	0.21	37.91	-0.40
TRAIN107	0.95	0.09	0.36	TRAIN014	0.80	-0.19	0.17	TRAIN129	0.63	14.04	0.25	VALID009	0.21	39.96	-0.43
TRAIN112	0.95	0.10	0.31	TRAIN055	0.80	-0.19	0.15	TRAIN108	0.62	11.43	0.28	TRAIN050	0.17	39.51	-0.43
TRAIN042	0.94	-0.06	0.53	TRAIN102	0.80	0.20	0.08	TRAIN123	0.62	10.77	0.26	TRAIN006	0.15	46.91	-0.52
TRAIN129	0.94	0.11	0.30	TRAIN057	0.79	-0.17	0.22	TRAIN128	0.59	11.86	0.26	TRAIN038	0.15	49.92	-0.54
TRAIN103	0.94	0.11	0.29	TRAIN022	0.79	-0.18	0.19	TRAIN002	0.58	13.48	0.31	VALID001	0.14	44.26	-0.49
VALID007	0.93	-0.06	0.53	TRAIN056	0.79	-0.19	0.18	TRAIN112	0.58	15.74	0.14	VALID003	0.12	54.70	-0.58
TRAIN113	0.93	-0.04	0.52	TRAIN122	0.79	0.21	0.08	TRAIN100	0.57	3.43	0.35	TRAIN040	0.10	52.93	-0.57
TRAIN010	0.93	-0.07	0.50	TRAIN005	0.78	-0.20	0.12	VALID015	0.57	17.83	0.18	TRAIN020	0.10	55.08	-0.59
VALID004	0.93	-0.04	0.49	TRAIN038	0.78	-0.21	0.09	TRAIN032	0.56	14.31	0.24	TRAIN011	0.09	58.80	-0.61
VALID015	0.93	-0.09	0.49	VALID016	0.74	-0.21	0.08	TRAIN109	0.54	-2.45	0.50	VALID018	0.07	50.69	-0.55
TRAIN021	0.93	-0.07	0.47	TRAIN016	0.74	-0.21	0.07	TRAIN101	0.54	-2.53	0.47	VALID014	0.07	51.50	-0.56
TRAIN116	0.93	0.07	0.40	TRAIN046	0.74	-0.21	0.07	TRAIN030	0.54	15.43	0.25	TRAIN055	0.07	56.82	-0.60
TRAIN023	0.91	-0.04	0.52	TRAIN049	0.74	-0.21	0.07	VALID000	0.54	17.86	0.16	TRAIN024	0.06	54.73	-0.58
VALID009	0.91	-0.05	0.49	TRAIN111	0.73	0.30	-0.24	TRAIN111	0.54	18.24	0.11	VALID008	0.06	58.47	-0.61
TRAIN013	0.91	-0.07	0.48	VALID018	0.70	-0.21	0.07	TRAIN041	0.54	21.17	0.03	TRAIN022	0.06	69.60	-0.67
TRAIN032	0.90	-0.06	0.50	TRAIN123	0.70	0.30	-0.25	TRAIN114	0.53	-2.14	0.42	TRAIN016	0.05	61.60	-0.63
VALID000	0.90	-0.09	0.46	TRAIN018	0.68	-0.23	0.00	TRAIN102	0.53	3.12	0.33	TRAIN018	0.04	57.44	-0.60
TRAIN017	0.89	-0.09	0.49	TRAIN004	0.65	-0.23	0.00	VALID007	0.53	18.29	0.16	TRAIN029	0.04	58.07	-0.61
TRAIN012	0.89	-0.08	0.48	VALID014	0.64	-0.23	-0.02	TRAIN122	0.51	1.03	0.33	VALID016	0.04	66.92	-0.66
TRAIN002	0.89	-0.09	0.46	VALID003	0.63	-0.23	-0.01	VALID006	0.51	19.37	0.11	TRAIN044	0.01	77.70	-0.71
VALID017	0.89	-0.10	0.45	TRAIN024	0.63	-0.23	-0.02	TRAIN043	0.51	20.22	0.09	VALID013	0.00	72.87	-0.69
VALID001	0.89	-0.09	0.44	VALID013	0.59	-0.25	-0.08	VALID017	0.49	19.28	0.13	TRAIN015	0.00	73.60	-0.69
TRAIN001	0.89	-0.10	0.44	TRAIN000	0.58	-0.24	-0.04	TRAIN034	0.49	19.41	0.11	TRAIN000	0.00	73.63	-0.69
TRAIN052	0.89	-0.11	0.42	TRAIN029	0.56	-0.24	-0.07	TRAIN042	0.49	21.14	0.03	TRAIN005	0.00	81.04	-0.72
TRAIN100	0.88	0.13	0.34	TRAIN047	0.48	-0.26	-0.11	TRAIN054	0.48	21.37	0.04	TRAIN045	0.00	82.62	-0.73
TRAIN124	0.88	0.13	0.31	TRAIN045	0.48	-0.26	-0.13	TRAIN023	0.46	21.18	0.03	VALID005	0.00	87.76	-0.74
TRAIN020	0.86	-0.11	0.42	TRAIN015	0.46	-0.27	-0.14	TRAIN017	0.46	22.30	0.00	TRAIN039	0.00	93.93	-0.76
TRAIN043	0.86	-0.11	0.42	TRAIN044	0.38	-0.27	-0.16	VALID010	0.43	22.14	0.02	TRAIN004	0.00	94.32	-0.76
VALID011	0.86	-0.11	0.42	TRAIN039	0.38	-0.28	-0.18	TRAIN001	0.43	22.86	-0.02	TRAIN047	0.00	101.81	-0.78
TRAIN037	0.86	-0.11	0.41					TRAIN019	0.42	23.43	-0.03				

Table S4 Evaluation of the ensemble of 111 model runs ordered from high to low FAC2 values with O:C and OA for the full period.

Full_O:C							Full_OA								
model	FAC2	MB	IOA	model	FAC2	MB	IOA	model	FAC2	MB	IOA	model	FAC2	MB	IOA
TRAIN041	0.98	-0.05	0.56	TRAIN108	0.87	0.11	0.40	TRAIN110	0.62	2.23	0.45	VALID010	0.39	24.06	-0.04
VALID002	0.98	-0.08	0.52	TRAIN002	0.87	-0.13	0.40	TRAIN126	0.61	5.13	0.38	TRAIN023	0.38	25.26	-0.08
TRAIN008	0.98	-0.05	0.54	TRAIN012	0.85	-0.13	0.39	TRAIN010	0.61	9.09	0.35	TRAIN019	0.37	26.28	-0.11
TRAIN036	0.97	-0.01	0.56	VALID017	0.85	-0.14	0.38	TRAIN113	0.60	6.79	0.36	VALID011	0.37	31.43	-0.24
TRAIN127	0.97	-0.02	0.55	TRAIN053	0.85	0.14	0.36	TRAIN119	0.60	9.54	0.31	TRAIN007	0.37	31.16	-0.24
TRAIN003	0.97	0.00	0.55	TRAIN102	0.85	0.15	0.33	TRAIN117	0.59	3.18	0.41	TRAIN052	0.37	29.10	-0.18
TRAIN119	0.97	-0.04	0.54	TRAIN020	0.84	-0.15	0.37	TRAIN031	0.59	11.94	0.28	TRAIN008	0.36	28.92	-0.18
TRAIN104	0.97	-0.01	0.56	TRAIN037	0.83	-0.15	0.35	TRAIN009	0.59	10.54	0.30	TRAIN051	0.33	30.19	-0.20
TRAIN051	0.97	-0.02	0.55	TRAIN019	0.83	-0.16	0.35	TRAIN121	0.59	2.87	0.41	TRAIN027	0.33	29.90	-0.19
TRAIN117	0.97	-0.01	0.53	TRAIN122	0.83	0.16	0.31	VALID004	0.59	4.01	0.41	TRAIN033	0.33	31.49	-0.23
TRAIN128	0.97	-0.06	0.51	TRAIN050	0.81	-0.17	0.33	TRAIN104	0.58	5.77	0.39	VALID012	0.31	35.63	-0.31
VALID006	0.96	-0.06	0.51	TRAIN060	0.81	-0.18	0.31	VALID002	0.58	13.27	0.24	TRAIN057	0.31	31.72	-0.24
TRAIN034	0.96	-0.06	0.53	TRAIN033	0.81	-0.17	0.32	TRAIN003	0.57	12.65	0.24	TRAIN048	0.31	35.26	-0.30
TRAIN120	0.96	-0.08	0.49	VALID012	0.78	-0.19	0.26	TRAIN116	0.57	1.97	0.39	TRAIN035	0.30	33.34	-0.27
TRAIN009	0.96	-0.08	0.51	TRAIN007	0.78	-0.19	0.26	TRAIN103	0.56	4.28	0.37	TRAIN060	0.30	36.02	-0.31
TRAIN023	0.96	-0.08	0.50	VALID010	0.78	-0.18	0.28	TRAIN127	0.56	4.78	0.37	TRAIN059	0.29	36.02	-0.32
TRAIN115	0.96	0.01	0.53	TRAIN026	0.77	-0.18	0.28	TRAIN106	0.56	1.97	0.40	TRAIN026	0.29	36.05	-0.31
VALID009	0.96	-0.09	0.49	TRAIN048	0.76	-0.19	0.25	TRAIN125	0.56	1.55	0.42	TRAIN021	0.28	35.19	-0.30
TRAIN101	0.95	-0.03	0.51	TRAIN111	0.76	0.25	0.01	TRAIN012	0.56	13.07	0.27	TRAIN025	0.25	41.70	-0.40
TRAIN126	0.95	-0.04	0.51	TRAIN025	0.75	-0.19	0.26	TRAIN107	0.55	5.05	0.34	TRAIN014	0.25	39.43	-0.37
TRAIN121	0.95	-0.05	0.50	TRAIN030	0.75	-0.19	0.27	TRAIN120	0.55	13.61	0.23	TRAIN013	0.24	40.12	-0.38
TRAIN114	0.95	-0.01	0.50	TRAIN123	0.75	0.26	0.00	TRAIN128	0.55	14.80	0.18	TRAIN056	0.24	41.11	-0.40
TRAIN058	0.95	0.02	0.56	TRAIN006	0.74	-0.20	0.24	TRAIN037	0.54	16.17	0.18	TRAIN028	0.23	43.64	-0.43
TRAIN042	0.95	-0.10	0.48	TRAIN035	0.73	-0.20	0.24	TRAIN115	0.53	4.74	0.34	VALID019	0.22	43.21	-0.42
TRAIN106	0.94	-0.05	0.50	TRAIN027	0.72	-0.20	0.23	TRAIN036	0.53	6.90	0.31	TRAIN049	0.22	42.84	-0.42
TRAIN010	0.94	-0.10	0.48	TRAIN054	0.72	-0.20	0.23	TRAIN108	0.53	13.86	0.18	TRAIN046	0.22	43.93	-0.43
TRAIN110	0.94	-0.04	0.48	TRAIN056	0.72	-0.21	0.20	TRAIN109	0.53	-2.10	0.46	TRAIN050	0.21	43.76	-0.43
TRAIN001	0.94	-0.12	0.44	VALID019	0.71	-0.21	0.22	TRAIN114	0.53	-0.18	0.40	VALID009	0.19	47.60	-0.47
TRAIN112	0.94	0.05	0.49	TRAIN059	0.71	-0.21	0.21	TRAIN101	0.52	-2.10	0.44	VALID001	0.18	56.05	-0.55
TRAIN107	0.94	0.04	0.50	VALID008	0.69	-0.21	0.20	TRAIN002	0.51	16.48	0.19	TRAIN006	0.14	57.68	-0.56
TRAIN125	0.94	-0.07	0.47	TRAIN055	0.69	-0.22	0.18	TRAIN058	0.51	11.30	0.19	TRAIN038	0.14	59.48	-0.58
TRAIN021	0.93	-0.12	0.44	VALID005	0.69	-0.21	0.20	TRAIN129	0.50	17.26	0.12	VALID003	0.13	62.45	-0.60
TRAIN109	0.93	-0.04	0.48	TRAIN022	0.68	-0.21	0.19	TRAIN030	0.50	18.22	0.12	VALID014	0.13	57.86	-0.57
VALID001	0.93	-0.12	0.48	TRAIN057	0.68	-0.21	0.20	VALID015	0.50	19.22	0.09	VALID018	0.12	57.88	-0.57
VALID007	0.93	-0.10	0.47	TRAIN005	0.64	-0.23	0.14	TRAIN112	0.50	18.32	0.09	TRAIN024	0.12	59.97	-0.58
TRAIN116	0.93	0.02	0.50	TRAIN014	0.64	-0.22	0.15	TRAIN053	0.49	12.64	0.17	TRAIN055	0.11	70.18	-0.64
TRAIN103	0.92	0.06	0.48	TRAIN016	0.63	-0.23	0.12	TRAIN123	0.49	12.79	0.18	TRAIN040	0.10	66.26	-0.62
VALID004	0.92	-0.09	0.47	TRAIN038	0.62	-0.23	0.12	TRAIN032	0.48	18.26	0.11	TRAIN018	0.10	65.93	-0.62
TRAIN129	0.92	0.07	0.47	TRAIN049	0.58	-0.24	0.10	TRAIN100	0.48	4.66	0.31	TRAIN020	0.10	68.88	-0.63
VALID011	0.92	-0.13	0.43	VALID016	0.58	-0.24	0.10	VALID000	0.47	20.06	0.08	VALID008	0.10	69.06	-0.64
TRAIN032	0.92	-0.11	0.46	TRAIN046	0.58	-0.24	0.10	TRAIN111	0.47	20.42	0.01	TRAIN029	0.09	70.25	-0.64
TRAIN113	0.92	-0.08	0.48	VALID018	0.58	-0.24	0.10	TRAIN041	0.46	22.32	-0.01	TRAIN016	0.09	70.82	-0.65
TRAIN031	0.91	-0.11	0.44	VALID003	0.54	-0.25	0.06	VALID007	0.46	21.42	0.04	TRAIN011	0.09	73.91	-0.66
VALID015	0.91	-0.13	0.42	TRAIN018	0.50	-0.26	0.03	TRAIN102	0.46	2.82	0.33	VALID016	0.08	78.82	-0.68
TRAIN028	0.91	-0.13	0.44	TRAIN024	0.50	-0.26	0.03	TRAIN122	0.45	1.56	0.33	TRAIN022	0.06	86.57	-0.71
TRAIN013	0.90	-0.12	0.43	TRAIN004	0.49	-0.26	0.03	TRAIN034	0.45	22.03	0.02	TRAIN015	0.05	86.22	-0.71
TRAIN052	0.90	-0.14	0.40	VALID014	0.49	-0.26	0.03	VALID006	0.44	22.47	0.00	TRAIN044	0.05	88.31	-0.71
TRAIN040	0.90	-0.12	0.43	TRAIN000	0.45	-0.26	0.01	TRAIN124	0.43	-3.36	0.39	VALID013	0.05	86.38	-0.71
VALID000	0.90	-0.13	0.42	VALID013	0.42	-0.27	-0.02	TRAIN118	0.43	-1.25	0.37	TRAIN000	0.04	86.77	-0.71
TRAIN017	0.89	-0.13	0.42	TRAIN029	0.41	-0.27	-0.02	VALID017	0.43	23.14	-0.01	TRAIN005	0.03	96.26	-0.74
TRAIN118	0.89	0.10	0.46	TRAIN047	0.36	-0.28	-0.05	TRAIN105	0.42	-2.74	0.38	TRAIN045	0.03	97.09	-0.74
TRAIN100	0.89	0.10	0.46	TRAIN045	0.33	-0.29	-0.07	TRAIN042	0.42	25.05	-0.07	VALID005	0.02	105.05	-0.76
TRAIN124	0.89	0.09	0.44	TRAIN015	0.32	-0.29	-0.08	TRAIN043	0.41	24.75	-0.07	TRAIN039	0.01	109.17	-0.77
TRAIN105	0.89	0.10	0.43	TRAIN044	0.28	-0.30	-0.10	TRAIN001	0.41	26.56	-0.13	TRAIN004	0.01	114.79	-0.78
TRAIN011	0.88	-0.15	0.39	TRAIN039	0.27	-0.30	-0.12	TRAIN054	0.40	24.17	-0.05	TRAIN047	0.01	121.60	-0.79
TRAIN043	0.88	-0.14	0.40					TRAIN017	0.39	25.67	-0.08				



Figure S7. Relative variation (%) of the 5 anthropogenic PPE (1 – 5) for the full period. Each pentagon represents the 5-D parameter space and the positions of the dots connected with lines show the position of each parameter within its range for that specific ensemble member. The filled area within the dots represents the explored parameter space in each ensemble member. Anticlockwise from top there are the five anthropogenic parameters: VBS_AGERATE (1), SVOC_VOLDIST (2), SVOC_OXRATE (3), IVOC_SC (4) and SVOC_SC (5). The values of the five parameters have been normalised dividing by their respective maximum values, hence their values in this plot range from 0 – 1. Example of interpretation in bottom right: the five parameters are towards their high values. The colour in the lines and dots represents the FAC2 values from the O:C analysis and the fill colour represents the FAC2 values from the OA analysis. Red = 0 – 0.2, orange = 0.2 – 0.4, yellow = 0.4 – 0.6, green = 0.6 – 0.8, light blue = 0.8 -0.9 and blue = 0.9 -1.0

S5. Emulator

S5.1 Selection of periods for building and testing the emulator.

S5.1.1 Emulators for the four periods (high and low OA concentrations)

Four period time-slots were considered to build and test the emulator based on OA-AMS concentrations; one period with high OA concentrations (P1) and one period with low OA concentrations (P2). and two time-slots; 13:00 – 16:00 (A) and 20:00 – 23:00 hrs (B).

- 13:00-16:00 hrs (P1A) and 20:00-23:00 hrs (P1B), on 10 – 12 May, with high org mass loadings
- 13:00-16:00 hrs (P2A) and 20:00-23:00 hrs (P2B), on 20 – 23 May, with low org mass loadings

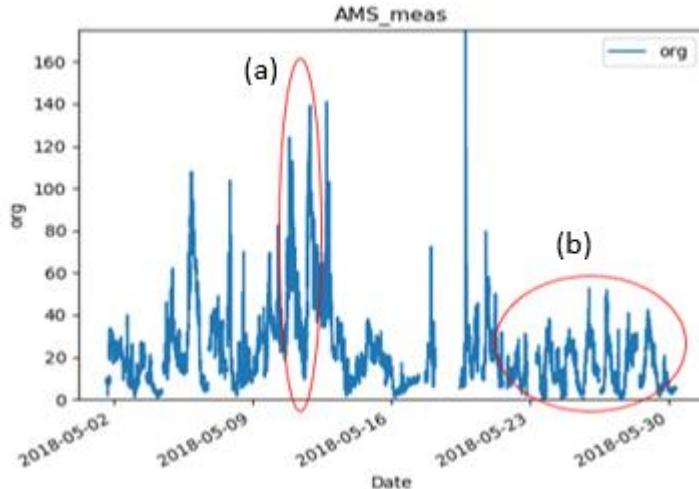


Figure S8. Time series of OA concentrations measured with the AMS highlighting the period with high (a) and low (b) Org concentrations.

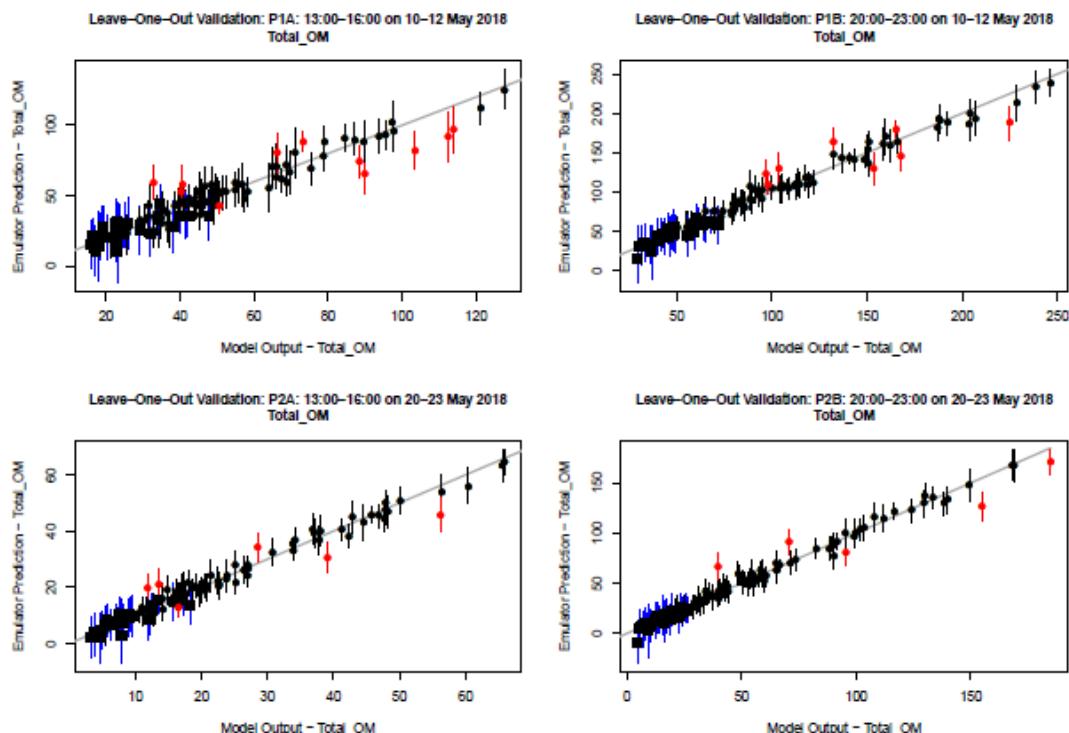


Figure S9. Validation of the four tested emulators for Org concentrations. Circles are the original 81 runs. Squares with error bars in blue are the new 30 runs with low settings of the anthropogenic SVOC scaling parameter (which has led to low aerosol mass). Runs where the actual model output lies outside the 95% prediction interval of the emulator are shown in red.

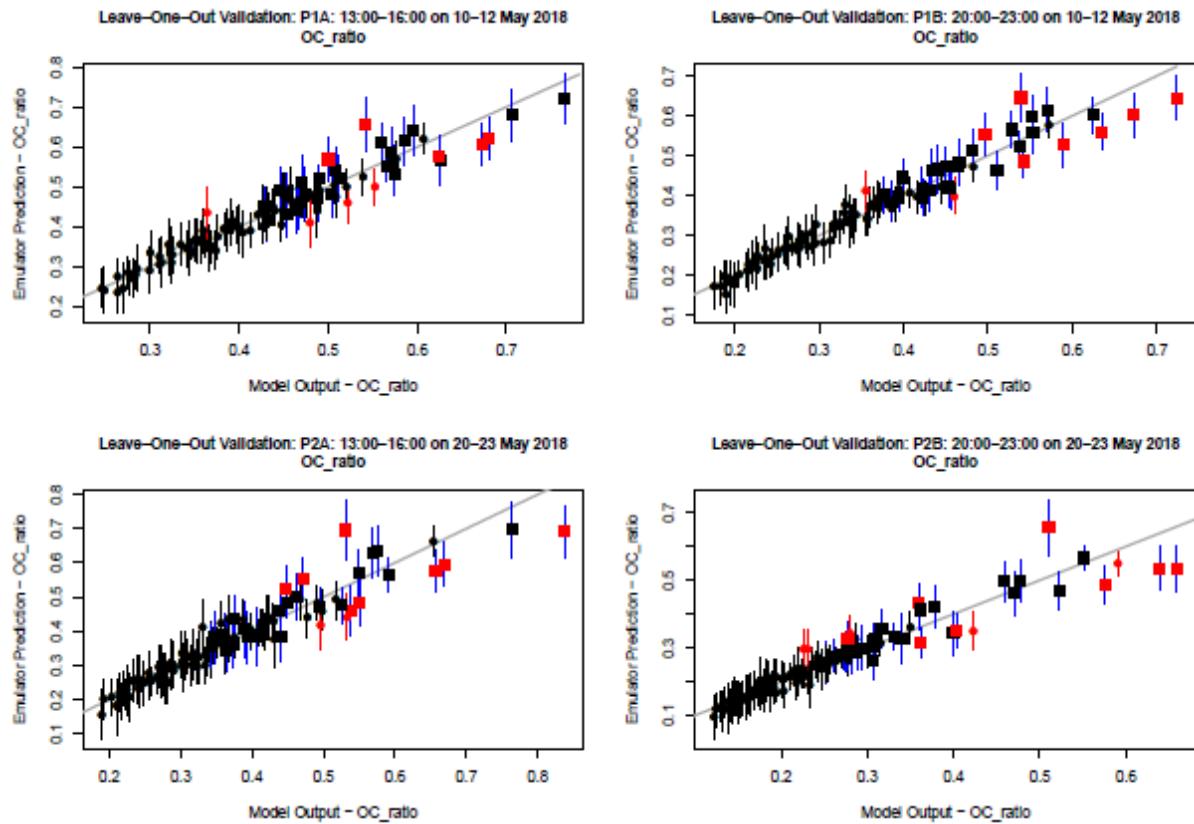


Figure S10 Validation of the four tested emulators for O:C ratios. Circles are the old 81 runs. Squares with error bars in blue are the new 30 runs with low aerosol mass. Red are the runs that are not within the 95% CI from prediction

S5.1.2 Emulator for the 2-4 pm period

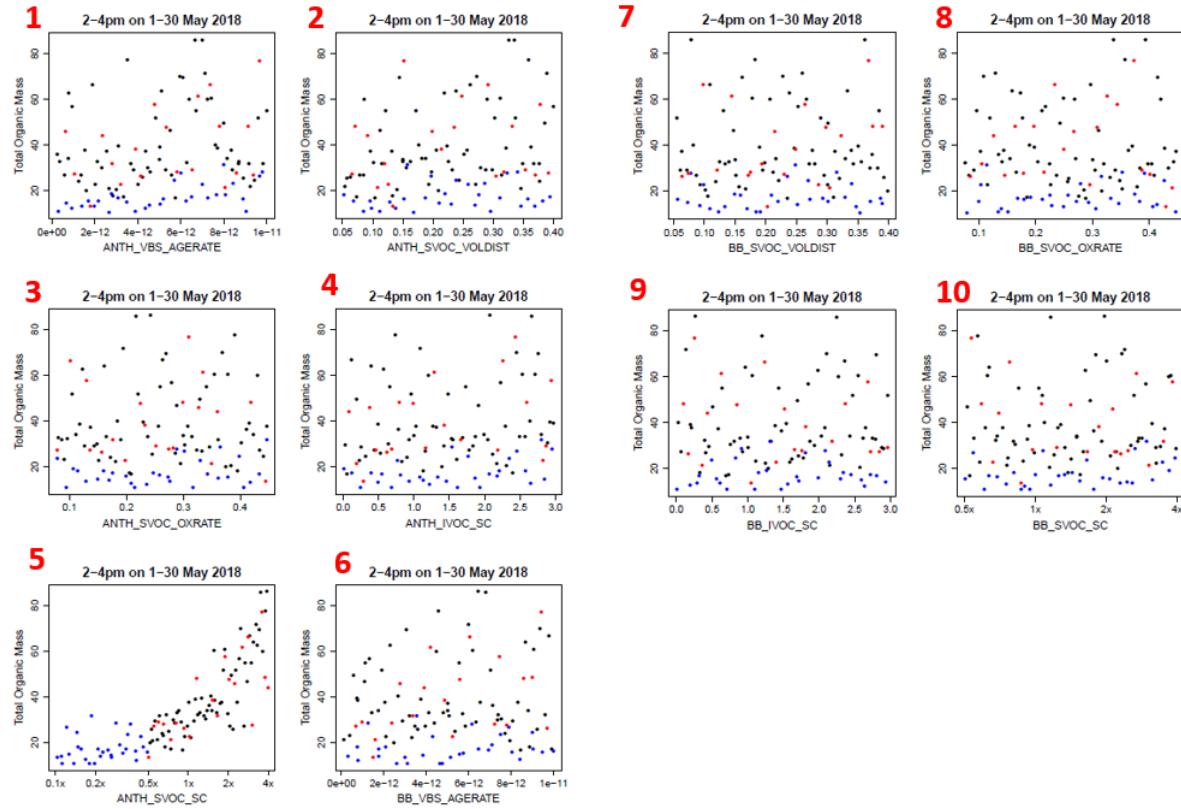


Figure S11 Spread of the total Organic mass for the 111 model runs vs the 10 parameters for the period 2-4 pm period. Red = 20 VALIDATE runs. Black = 61 TRAIN runs. Blue = 30 new TRAIN runs.

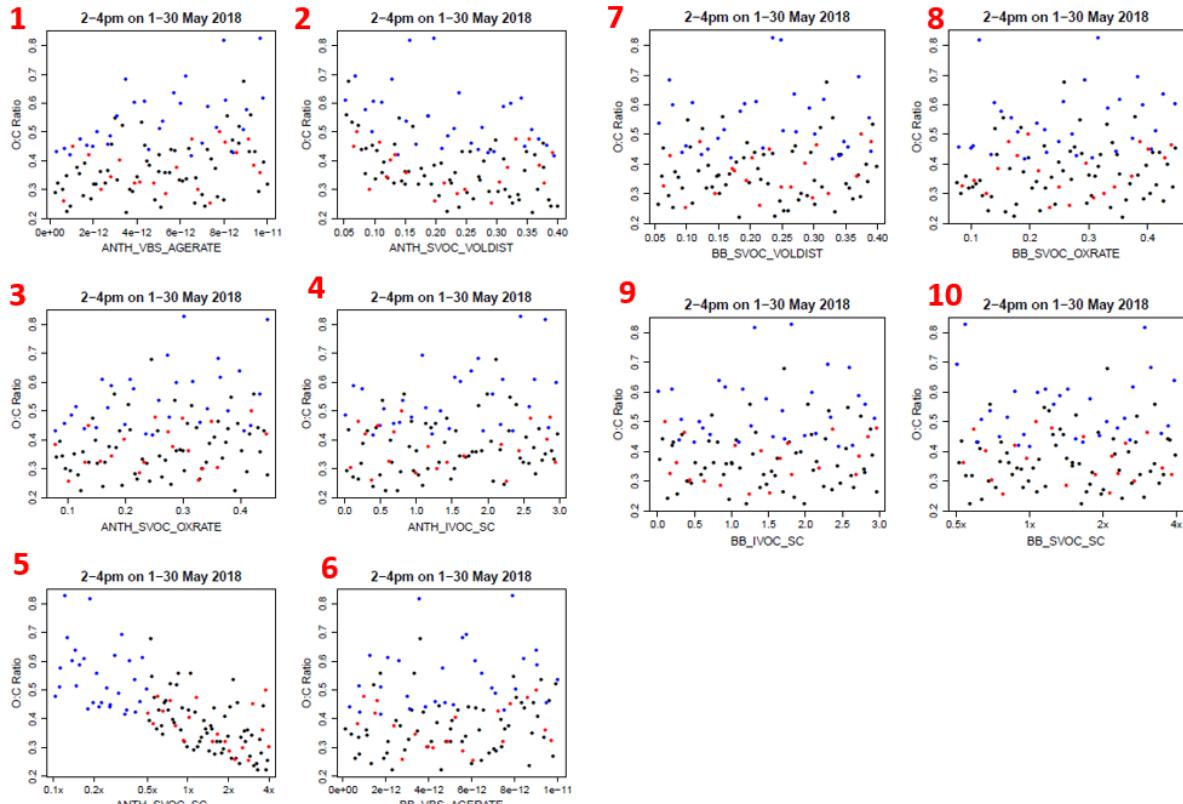


Figure S12. Spread of the total O:C ratio for the 111 model runs vs the 10 parameters for the 2-4 pm period. Red = 20 VALIDATE runs. Black = 61 TRAIN runs. Blue = 30 new TRAIN runs.

S5.2 Total_OM and O:C 2-d histogram analysis with and without constraint

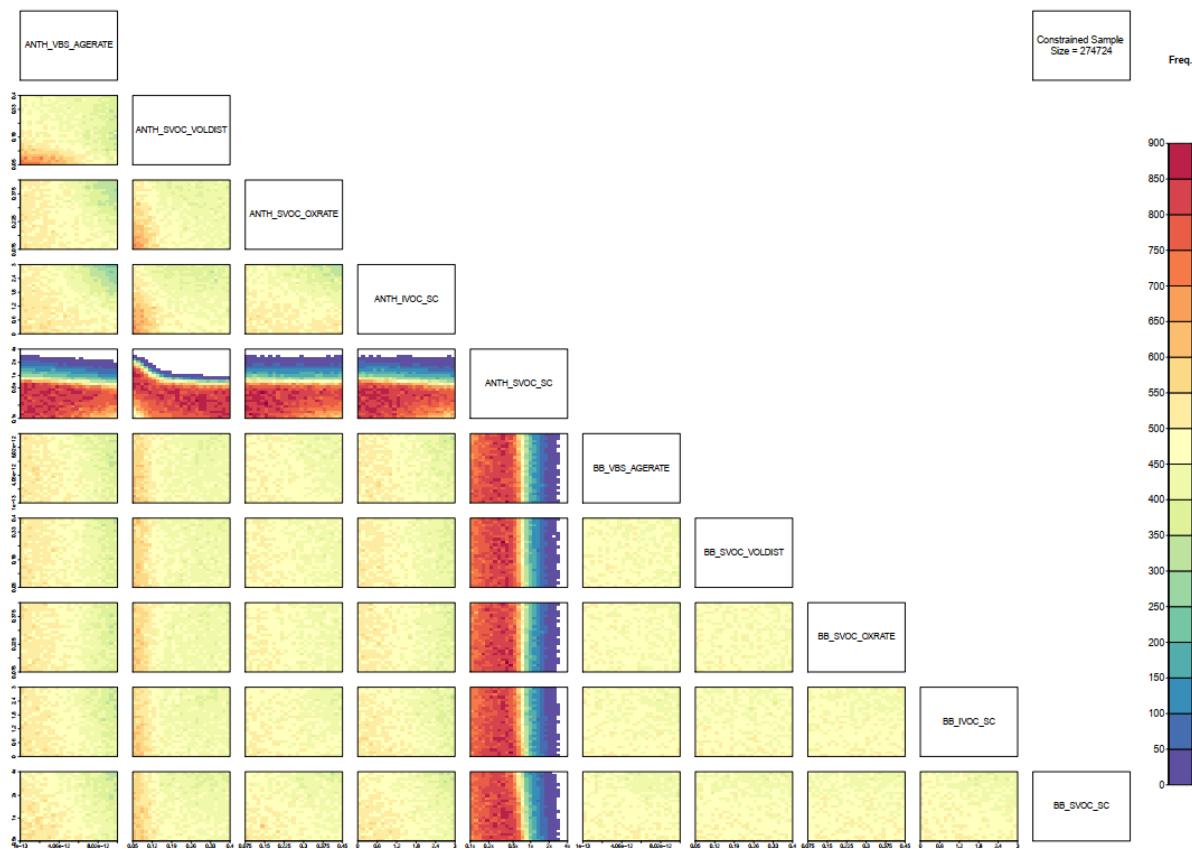


Figure S13. 2-d histogram for joint constraint effect (Total_OM and OC_ratio) for the full period accounting for emulator uncertainty. Retain 274724 variants from 0.5 million (~54.94%).

Carslaw, D. C., and Ropkins, K.: openair - An R package for air quality data analysis, Environ Modell Softw, 27-28, 52-61, DOI 10.1016/j.envsoft.2011.09.008, 2012.

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Willmott, C. J., Robeson, S. M., and Matsuura, K.: A refined index of model performance, International Journal of Climatology, 32, 2088-2094, <https://doi.org/10.1002/joc.2419>, 2012.