Supplement of Atmos. Chem. Phys., 16, 13773–13789, 2016
http://www.atmos-chem-phys.net/16/13773/2016/
doi:10.5194/acp-16-13773-2016-supplement

Atmospheric Chemistry and Physics Open Access EGU

Supplement of

Model simulations of cooking organic aerosol (COA) over the UK using estimates of emissions based on measurements at two sites in London

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1 Supplementary information for the Methods section

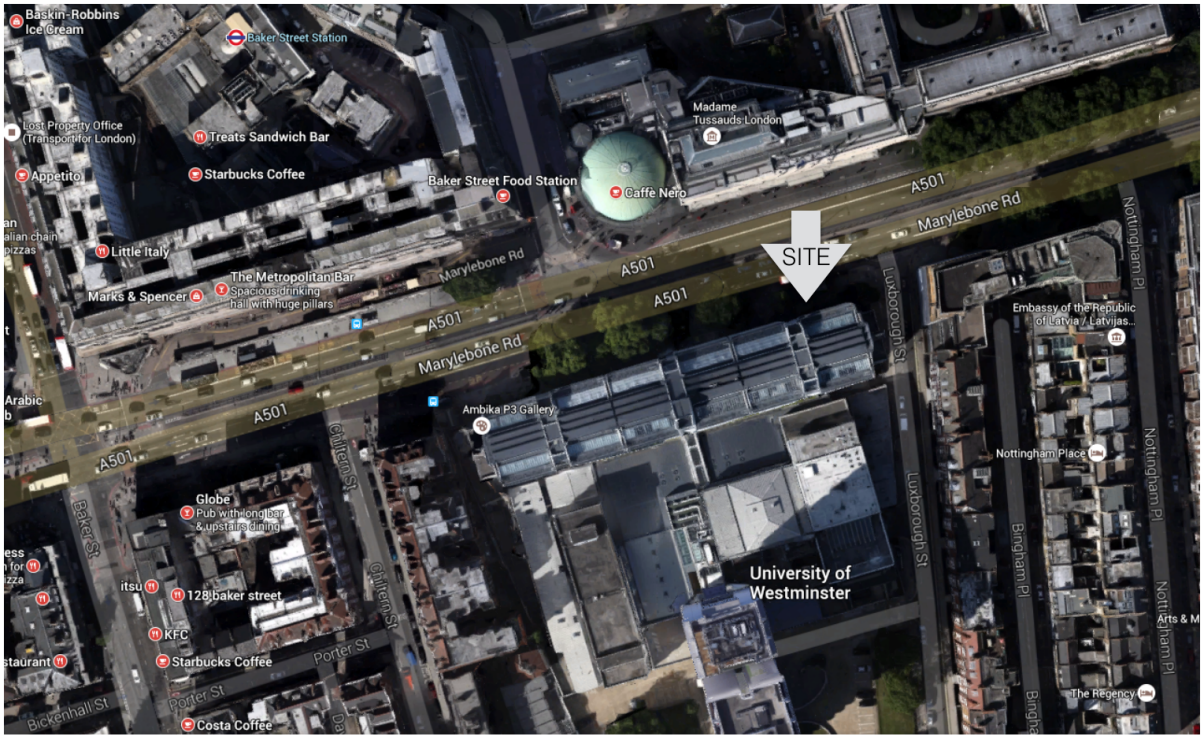

Figure S1: Location of the London Marylebone Road measurement site with some of the cafes and restaurants in the area marked. Imagery © 2015 Google Earth, map data © 2015 Google; last accessed on 10-Dec-2015.

1.1 Diurnal variation of COA emissions

The measured diurnal profile of COA at the Marylebone Road site was used as a basis for an emission profile for the atmospheric chemistry transport model (ACTM). The normalised emission diurnal profiles (for weekdays and weekends separately), denoted *Test1 (asMeasured)*, is given in Table S1, and is shown in Fig. S2. COA emissions were gridded to workday population density for an annual total of 7.4 Gg for the UK as this value minimised the biases between modelled concentrations and over the year of measurements at both the Marylebone Road and North Kensington sites. Average hourly diurnal profiles modelled for the two grid cells containing the measurements sites for the Test1 iteration are shown in Fig. S3. Using the measured profile yielded near perfect correlation coefficients between the measured and modelled diurnal cycles at the measurement stations (0.96 and 0.97 for Marylebone Road weekdays and weekends, respectively, and 0.98 for North Kensington for both weekdays and weekends), as well as good COE values (0.71 and 0.71 for Marylebone Road, and 0.33 and 0.67 for North Kensington). However, Fig. S3 also shows that at Marylebone Road, the modelled lunchtime peak is underestimated as the concentrations that are measured at that time would have been strongly diluted due to a deeper atmospheric boundary layer than in the evening. Therefore, sensitivity runs with altered emission profiles to give a relatively bigger emission during day time were conducted.

For the first sensitivity experiment the emissions around lunchtime were increased (particularly between 12.00 and 14.00) and night-time emissions were decreased. For the weekend profile, evening emissions (dinnertime) were also increased slightly. All emissions changes were normalised to ensure the total amount of COA emitted each day remained the same. The resulting emission profiles, denoted *Test2*, are given in Table S1, and are shown in Fig. S2. Average hourly profiles with the Test2 emission profiles are shown in Fig. S4. There is an improvement for the Marylebone Road site (r-values increase to 0.98 and 0.99 for the weekdays and weekend profiles, respectively; COE values are increased to 0.78 and 0.85, weekdays and weekends, respectively; Fig. S4a and b). For North Kensington, however, these metrics are slightly poorer in the Test2 run, but still remain high (r-values of 0.91 and 0.94, COE values of 0.30 and 0.58; Fig. S4cd).

Based on the results of Test2, for a further sensitivity experiment the lunchtime emissions were decreased relative to Test2 and breakfast emissions were elevated slightly. The weekend profile was left identical to Test2. The resulting emission profiles, denoted *Test3*, are given in Table S1, and are shown in Fig. S2. Average hourly profiles of COA concentrations modelled with these emission profiles are shown in Fig. S5. The r-values at the Marylebone Road site increase to 0.99 for both weekday and weekends and the weekday COE increases to 0.82. This was chosen as the final profile for COA emissions. Further evaluation of modelled COA concentrations at these sites is presented in the main paper.

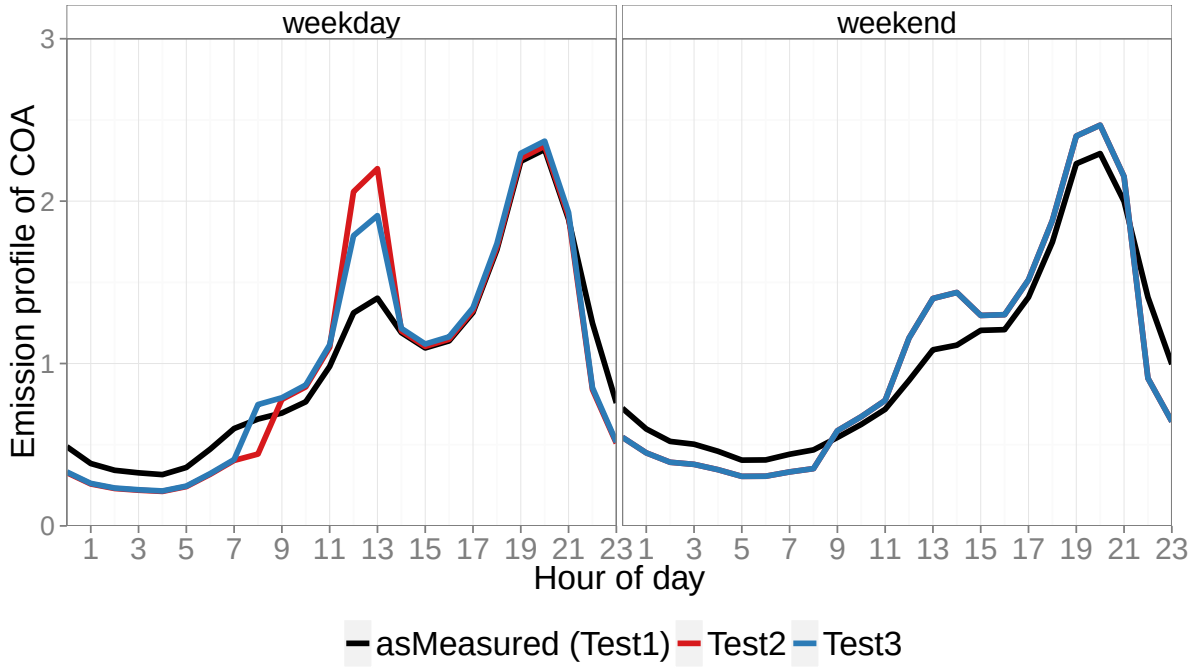

Figure S2: Diurnal normalised emission profiles for COA used in sensitivity runs. The timestamp is at the beginning of the hour. The specific hourly values are also given in Table S1.

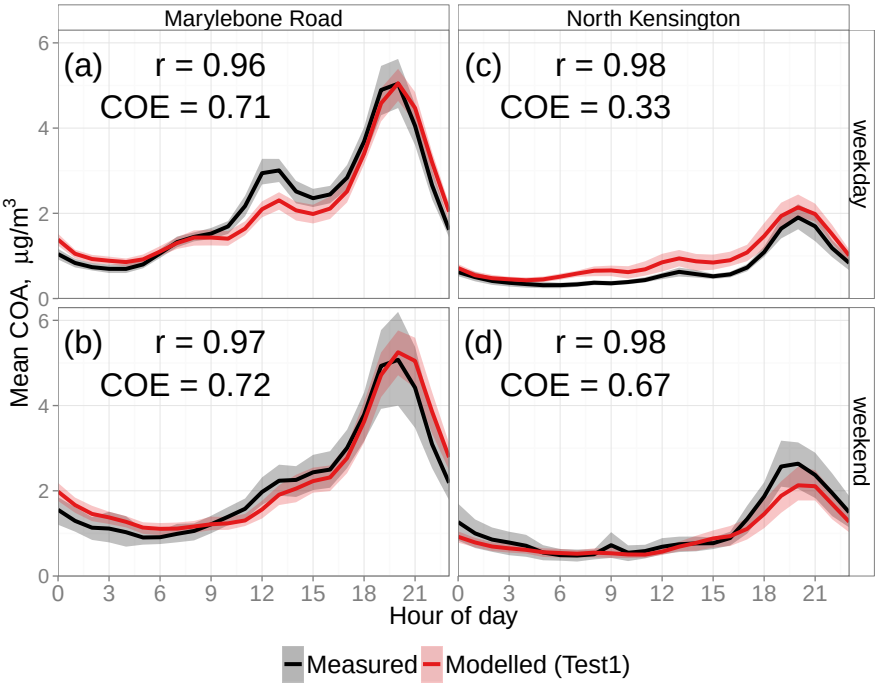

Figure S3: Average hourly profiles of measured and modelled COA using the first iteration diurnal emission profiles (Test1: asMeasured). The shading is the 95% confidence interval. Timestamp is at the beginning of the hour.

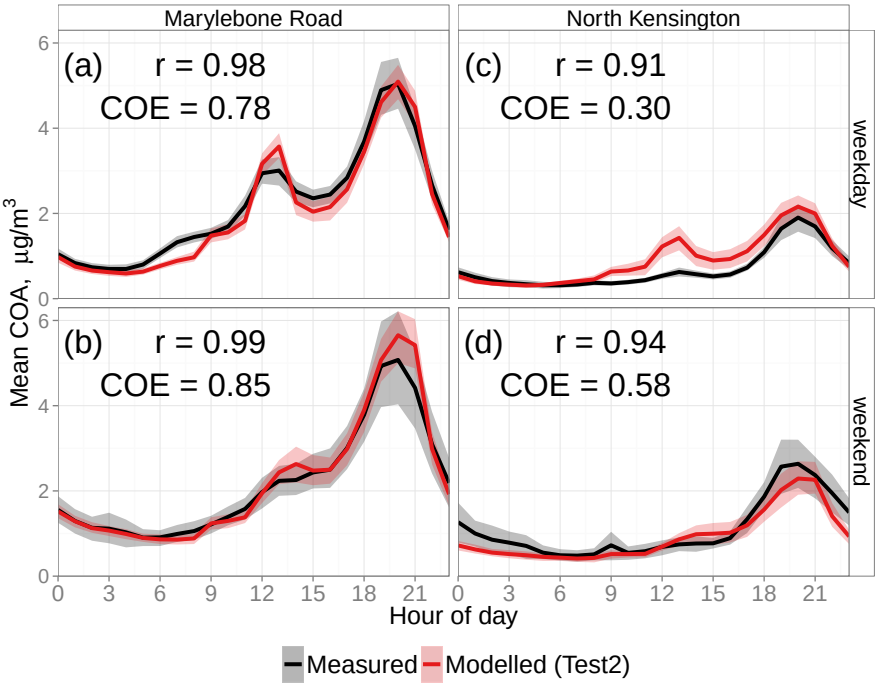

Figure S4: Similar to Fig.S3, but with modified diurnal emission profiles (Test2).

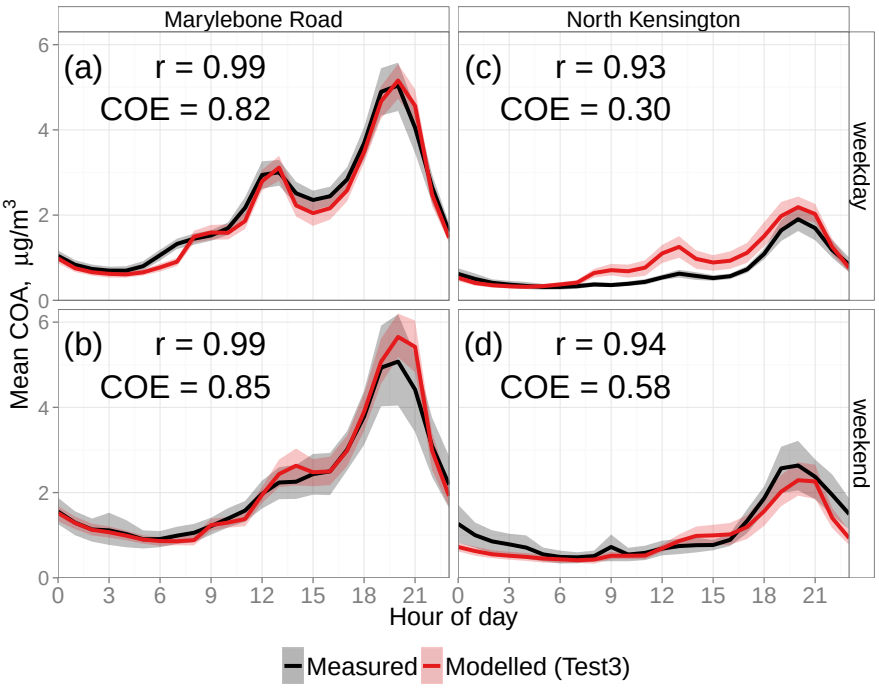

Figure S5: Similar to Fig.S3, but with modified diurnal emission profiles (Test3).

Table S1: Different normalised diurnal emission profiles for COA emissions used in sensitivity runs. The timestamp is at the beginning of the hour. The weekend values for Test2 and Test3 are identical.

	Test1 (as measured)		Test2		Test3 (final)	
	weekdays	weekend	weekdays	weekend	weekdays	weekend
0:00	0.49	0.72	0.32	0.55	0.33	0.55
1:00	0.38	0.60	0.26	0.45	0.26	0.45
2:00	0.34	0.52	0.23	0.39	0.23	0.39
3:00	0.33	0.50	0.22	0.38	0.22	0.38
4:00	0.32	0.46	0.21	0.35	0.21	0.35
5:00	0.36	0.40	0.24	0.30	0.25	0.30
6:00	0.47	0.41	0.32	0.30	0.32	0.30
7:00	0.60	0.44	0.40	0.33	0.41	0.33
8:00	0.66	0.47	0.44	0.35	0.75	0.35
9:00	0.69	0.54	0.78	0.59	0.79	0.59
10:00	0.76	0.63	0.86	0.67	0.87	0.67
11:00	0.98	0.72	1.10	0.77	1.12	0.77
12:00	1.31	0.90	2.06	1.16	1.79	1.16
13:00	1.40	1.08	2.20	1.40	1.91	1.40
14:00	1.19	1.11	1.20	1.44	1.22	1.44
15:00	1.10	1.20	1.11	1.30	1.12	1.30
16:00	1.14	1.21	1.15	1.30	1.16	1.30
17:00	1.31	1.41	1.33	1.52	1.34	1.52
18:00	1.70	1.75	1.72	1.88	1.74	1.88
19:00	2.25	2.23	2.26	2.40	2.29	2.40
20:00	2.32	2.29	2.34	2.47	2.37	2.47
21:00	1.89	2.00	1.90	2.15	1.93	2.15
22:00	1.25	1.41	0.84	0.91	0.85	0.91
23:00	0.76	1.00	0.51	0.64	0.52	0.64
SUM	24.00	24.00	24.00	24.00	24.00	24.00

2 Supplementary information for the Results section

2.1 Hourly timeplots

Hourly averaged time-plots of modelled and measured COA concentrations for the whole year are shown in Figs. S6 and S7 (Marylebone Road) and Figs. S8 and S9 (North Kensington).

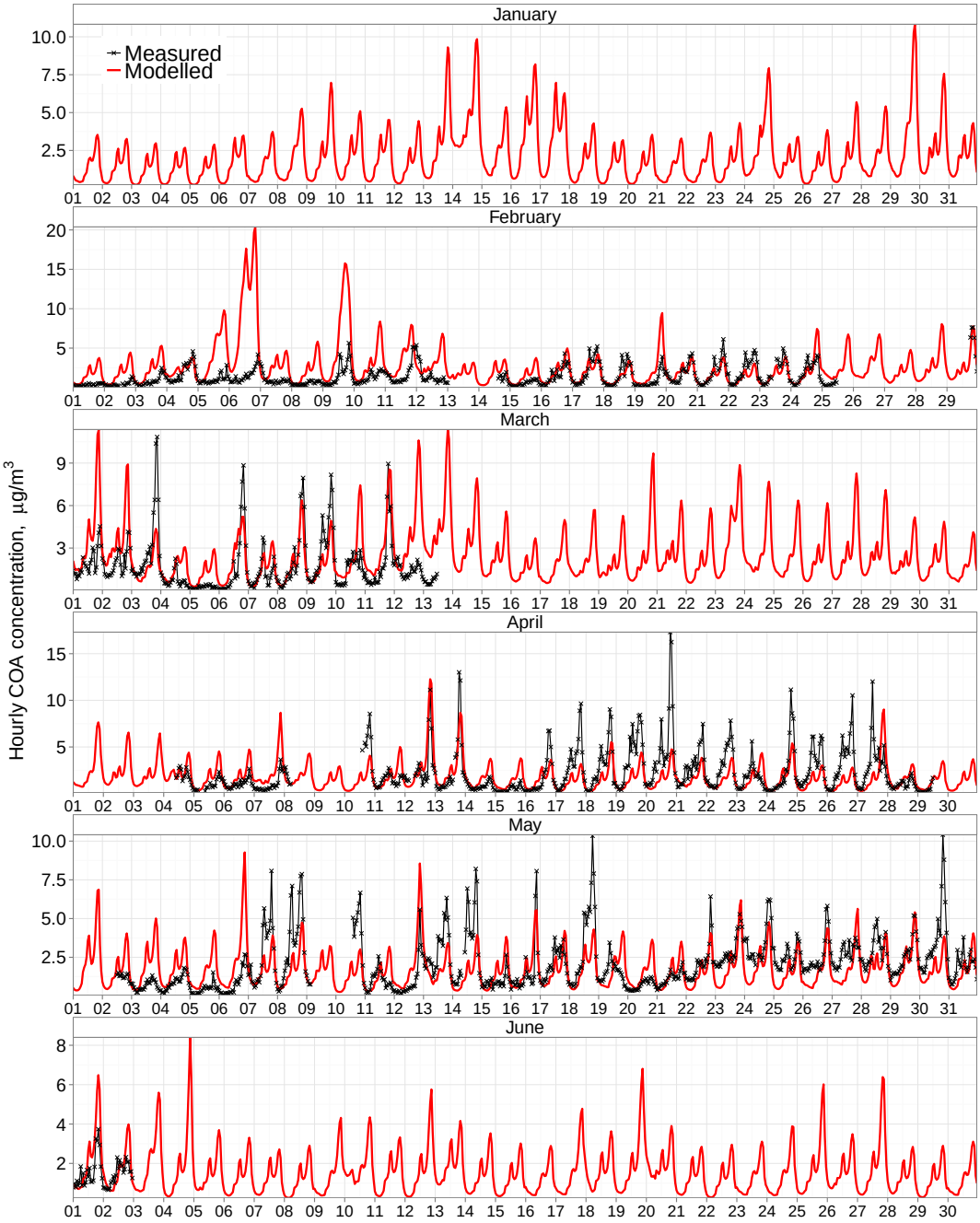

Figure S6: Hourly time-plots of measured and modelled COA concentrations at the Marylebone Road measurement site for January–June 2012.

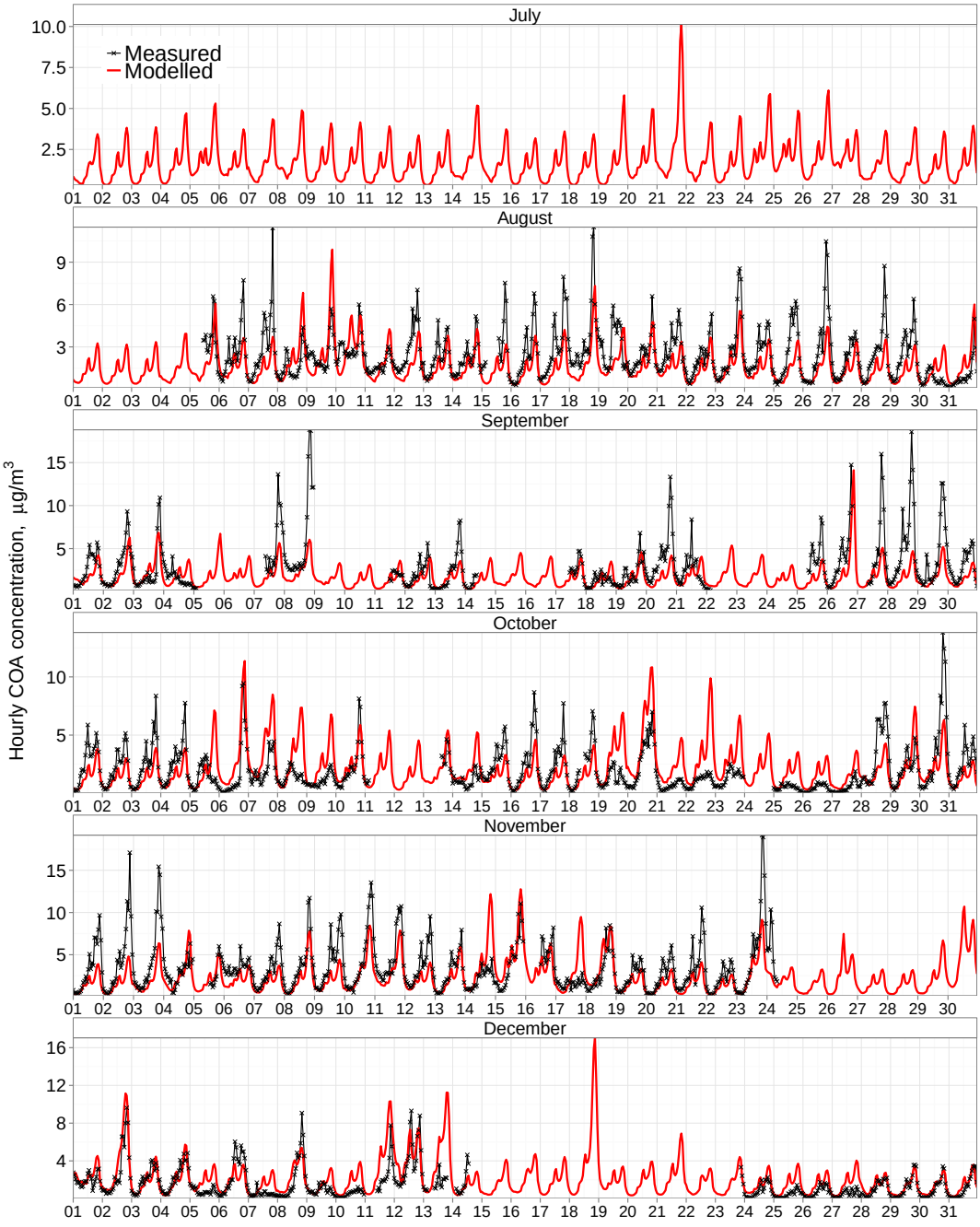

Figure S7: Hourly time-plots of measured and modelled COA concentrations at the Marylebone Road measurement site for July–December 2012.

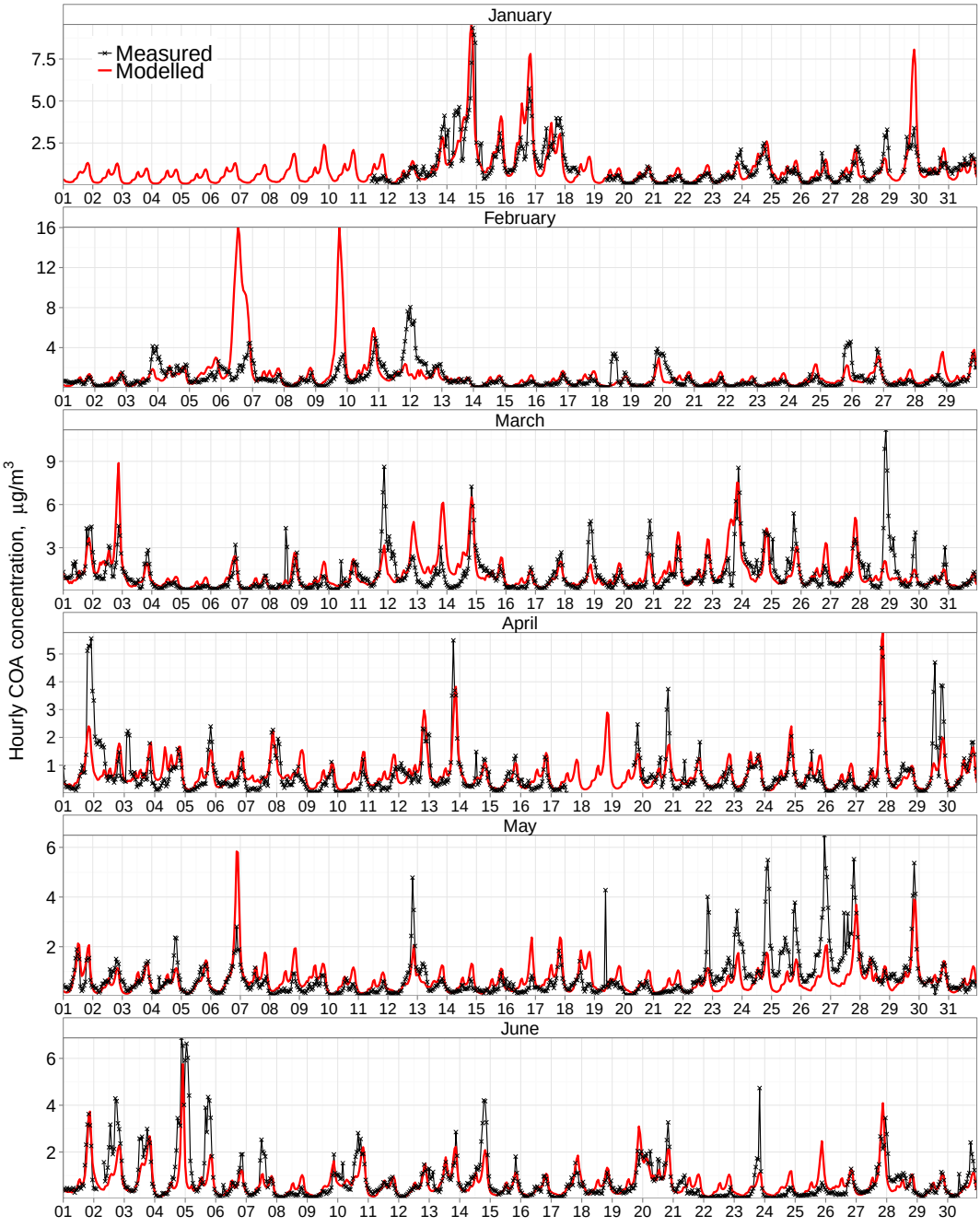

Figure S8: Hourly time-plots of measured and modelled COA concentrations at the North Kensington measurement site for January–June 2012.

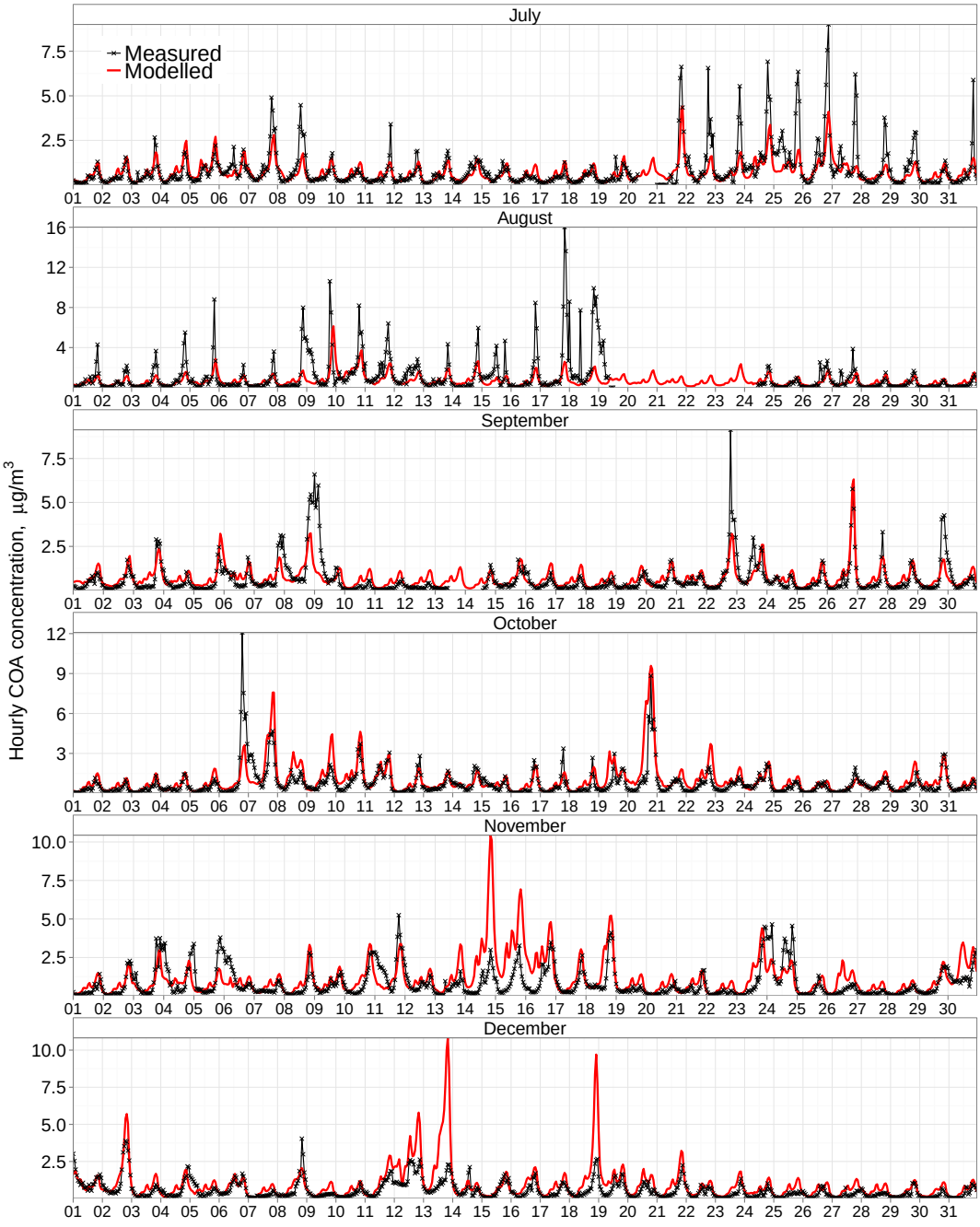

Figure S9: Hourly time-plots of measured and modelled COA concentrations at the North Kensington measurement site for July–December 2012.

2.2 Daily evaluation with wind vectors

Scatterplots of daily-averaged modelled and measured concentrations at the Marylebone Road site conditioned by four divisions of wind directions are shown in Fig. S10. The FAC2, *r*-value, and the number of data points are also presented in each panel. During the period of measurements at Marylebone Road in 2012 (total number of days with measurements was 191) the prevalent daily-averaged wind direction was SW (91 days). The model generally underestimates daily COA concentrations when wind was from this direction (Fig. S10c: most points below the 1:1 line), but most points are still within a factor of two of the measurements (Fig. S10c: FAC2 = 80%). FAC2 values are noticeably lower for the northerly directions (Fig. S10a (NW): FAC2 = 67%, Fig. S10b (NE): FAC2 = 42%), with the model generally overestimating COA concentrations on days when the prevalent wind direction was northerly. As explained in the main paper (Sect. 3.2), this is probably a sub-model-grid effect caused by Regent's Park to the north of the measurement site (Fig. 7).

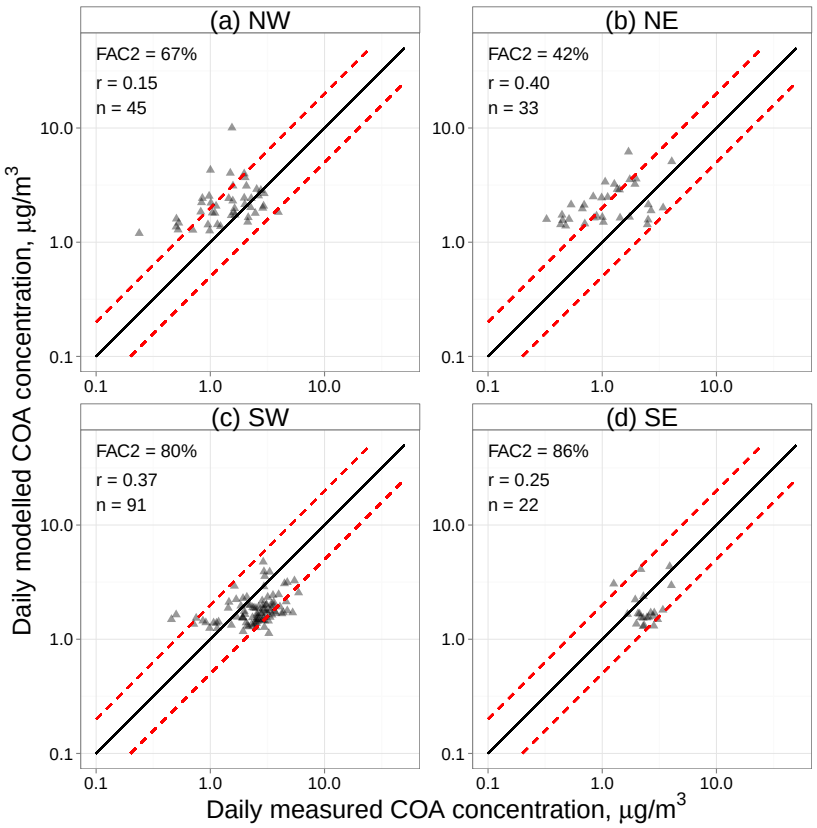

Figure S10: Scatterplots of daily-averaged modelled versus measured concentrations at the Marylebone Road site panelled by four divisions of wind directions measured at the Heathrow Airport meteorological station (Met Office, 2012): (a) from 270°–360° (denoted NW: north west), (b) from 0°–90° (denoted NE: north east), (c) from 180°–270° (denoted SW: south west), (d) from 90°–180° (denoted SE: south east).

Scatterplots of the daily-averaged modelled and measured concentrations at the Marylebone Road site panelled instead by wind speed quantiles but coloured by wind directions are shown in Fig. S11. The number of data points on each quantile panel is equal, but the number of points with the same colouring is largest for the SW direction

(180°–270° from north), as was shown in Fig. S10. During low wind speeds (Fig. S11a), COA concentrations for winds from the NE and NW directions are overestimated, but concentrations for winds from the SW and SE directions are well reproduced by the model. For all other wind speeds (Fig. S11b, c and d) COA concentrations for winds from the northerly directions are overestimated, but COA concentrations for winds from the southerly directions are underestimated. It is interesting, but currently unexplained observation, that COA concentrations are not underestimated when winds from the southerly directions are of low wind speed. Measurements at different locations and more modelling studies (including different models, for example an urban dispersion model) of COA concentrations in London, as well as in other cities would be necessary to draw further conclusions from these findings.

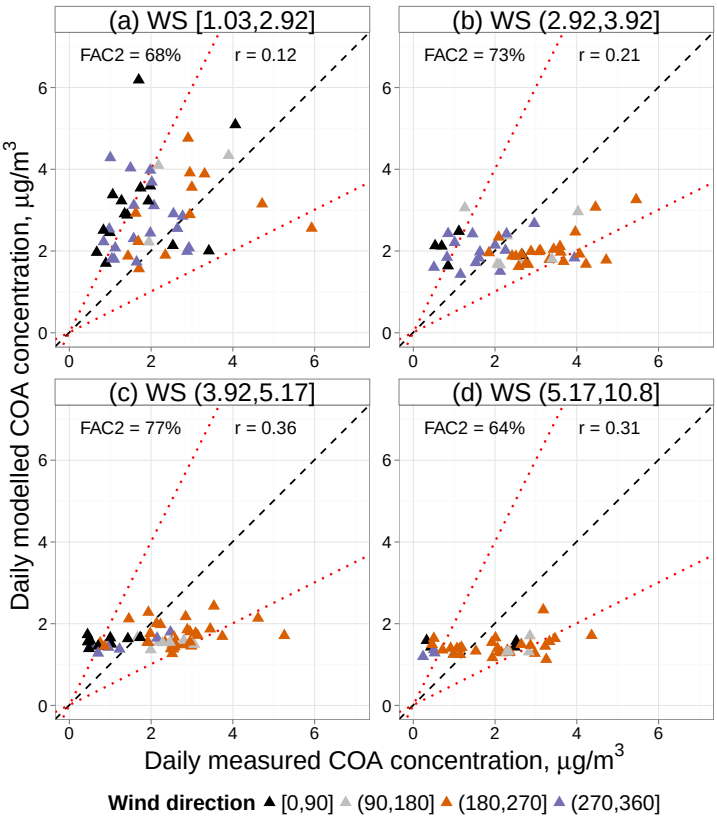

Figure S11: Scatterplots of daily-averaged modelled versus measured concentrations at the Marylebone Road site panelled by wind speed (WS) quartiles measured at the Heathrow Airport meteorological station (Met Office, 2012): (a) first quartile, (b) second quartile, (c) third quartile, (d) fourth quartile; units of wind speeds displayed on panel labels $\mathrm{m\,s^{-1}}$. Points are coloured by wind directions (see Fig. S10 for number of points from each direction), units: degrees from north.

2.3 COA concentrations in the vicinity of London

Modelled surface concentrations at the Harwell supersite near London for the 18 highest days (95th percentile: 0.43 $\mu\mathrm{g\,m^{-3}}$) are shown in Fig. S12. See Sect. 3.5 in the main paper for more informations and time plots of COA

concentrations at this site.

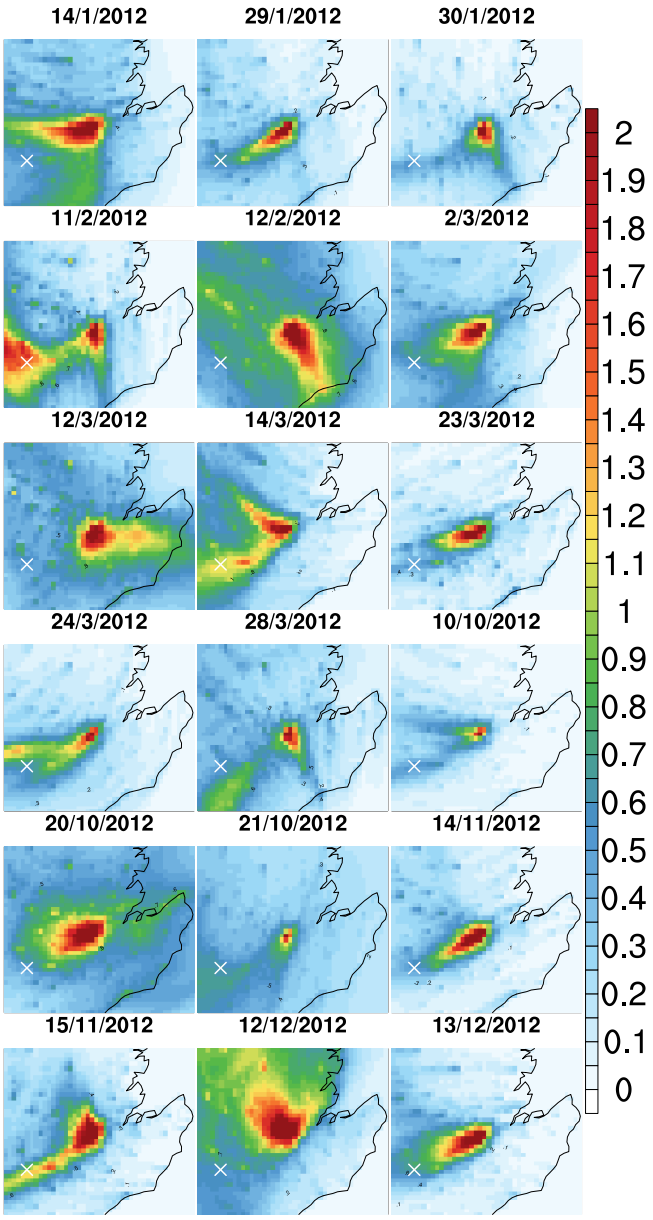

Figure S12: Daily-average modelled COA surface concentration maps for the 18 days when modelled COA concentrations for the Harwell location (marked with a white cross) were highest (95th percentile of days in the year: daily-average $COA \geq 0.43\ \mu g\ m^{-3}$). Units: $\mu g\ m^{-3}$.

References

Met Office: Met Office Integrated Data Archive System (MIDAS) Land and Marine Surface Stations Data (1853-current), Tech. rep., NCAS British Atmospheric Data Centre, URL http://catalogue.ceda.ac.uk/uuid/220a65615218d5c9cc9e4785a3234bd0, 2012.