

# **Controls of carbon dioxide concentrations and fluxes above central London: Supplementary Material**

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## **1 Introduction**

The data presented in this supplement provide additional details regarding some of the methods (including locations) and results of this study. Whilst not essential to the understanding of the main article, the following information aims at complementing it and is referred to in the main text.

## **2 Methods**

### **2.1 Site location**

The location of the BT tower is given in Fig. S1. This figure also shows the location of the Imperial College [CO<sub>2</sub>] measurement site (Rigby et al., 2008), and five traffic monitoring sites used in this study.

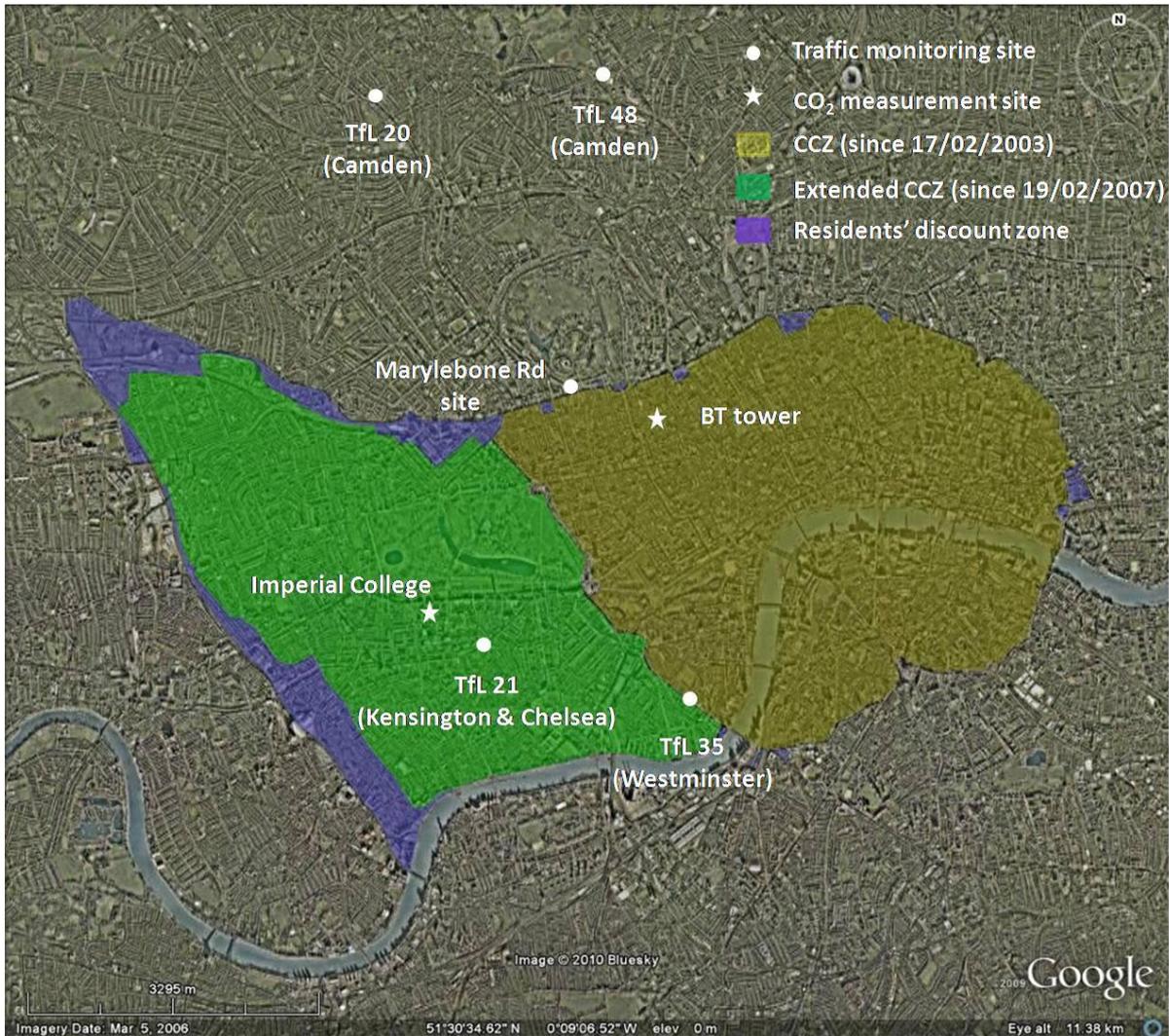


Figure S1: Location of the BT Tower and the Imperial College (Rigby et al., 2008) measurement sites (star-shaped symbols) and of some traffic monitoring stations (circles) on a satellite image of central London (Google<sup>TM</sup> Maps, 2006)<sup>1</sup>. The Marylebone Road site is funded by London local authorities (operated by the Environmental Research Group (ERG)<sup>2</sup>, King's College London) whilst the four remaining sites shown on this map are run by Transport for London (TfL)<sup>3</sup>. The original and extended congestion charging zones (CCZ) are also outlined.

## 2.2 Estimation of the CO<sub>2</sub> emissions from natural gas consumption

Natural gas provides 81% of the energy used for commercial and residential space and water heating as well as cooking for the whole of the UK, and a considerably larger fraction for UK

<sup>1</sup> <http://maps.google.co.uk/>

<sup>2</sup> <http://www.erg.kcl.ac.uk/>

<sup>3</sup> <http://www.tfl.gov.uk/>

urban centres (DTI, 2007)<sup>4</sup>. UK natural (commercial and residential) gas usage data as a function of average daily temperature were obtained from statistics of the UK Department of Energy and Climate Change (DECC, 2007)<sup>5</sup> and extrapolated to the London boroughs found in the flux footprint area of the BT tower. Daily demand for natural gas at the national scale ( $G_{nat}^{total}$  in GWh) was found to be a near linear function of temperature ( $R^2 = 0.78$ ). The daily gas usage at the borough scale ( $G_i^{daily}$ ) can be estimated from a linear function of temperature ( $\bar{T}_{daily}$ , °C) parameterised on daily and annual national demands ( $G_{nat}^{daily}$  and  $G_{nat}^{total}$  in GWh, respectively) and on annual demand at borough level ( $G_i^{total}$ , in GWh):

$$G_i^{daily} = \frac{G_{nat}^{daily}}{G_{nat}^{total}} \times G_i^{total} (-13.688 \times \bar{T}_{daily} + 397.54) \quad (S1)$$

Annual gas consumption for the borough of Westminster, calculated using Eq. (S1) parameterised on temperatures measured at the BT tower, was in good agreement with NAEI estimates (+ 12% with respect to NAEI data). Daily gas usage at borough-level was estimated using the parameterisation of Eq. (8) and converted into CO<sub>2</sub> emissions ( $F_{C-gas}$ ) using a conversion factor of 0.185 kg CO<sub>2</sub> (kWh)<sup>-1</sup> (Carbon Trust, 2009)<sup>6</sup>. Emissions were assumed to be uniformly-distributed at borough-level.

For the purpose of comparing eddy-covariance and NAEI data at sub 24-hour level, a diurnal (1 hour temporal resolution) gas consumption trend for Edinburgh (UK) city centre was applied to London figures (see Fig. S2), because for London gas supply statistics at hourly resolution were not available. This was especially useful when considering daytime (09:00 – 18:00) data only.

The methodology used to compare eddy-covariance estimates to bottom-up inventory data from NAEI is summarised as a flowchart in Fig. S3.

<sup>4</sup> <http://www.berr.gov.uk/files/file43843.pdf>

<sup>5</sup> <http://www.decc.gov.uk>

<sup>6</sup> <http://www.carbontrust.co.uk>

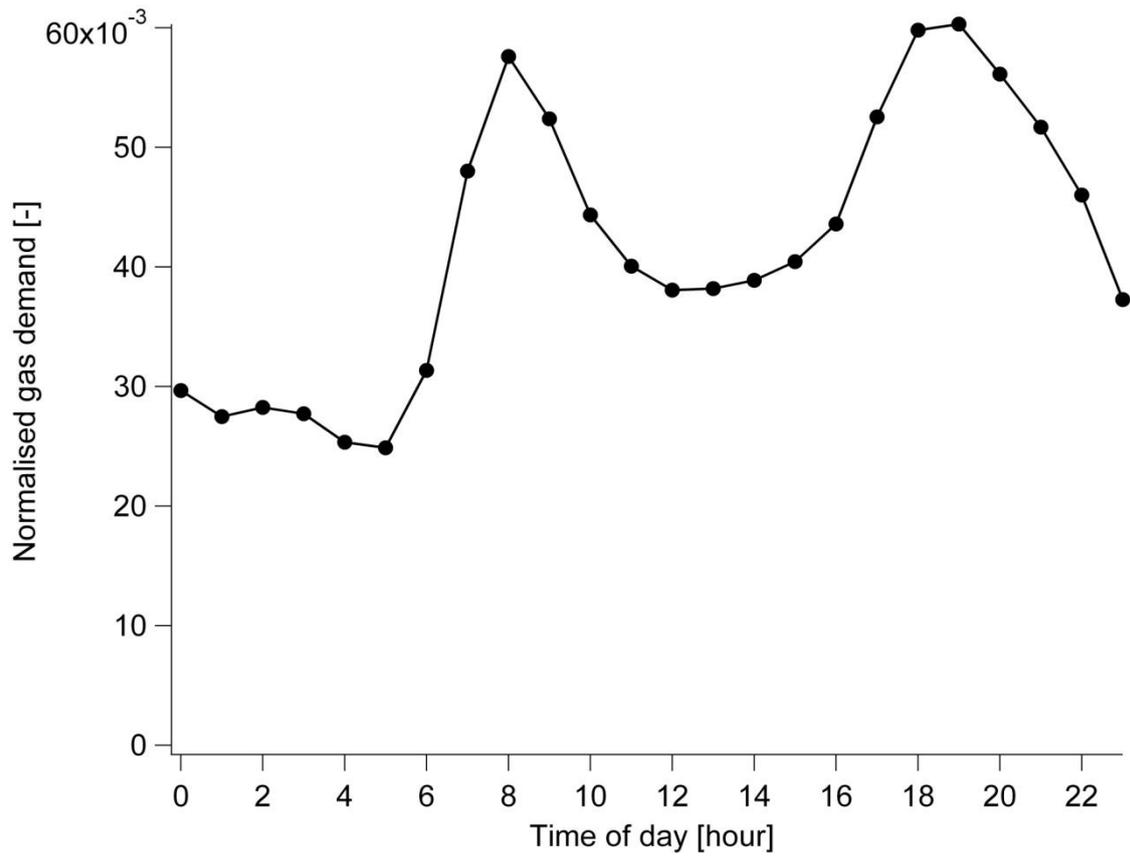


Figure S2: Diurnal trend in demand for natural gas in Edinburgh (UK) city centre (20/10/2000 – 30/11/2000) as provided by the operators of the gas supply network. A non-normalised version of the data appeared in Nemitz et al. (2002).

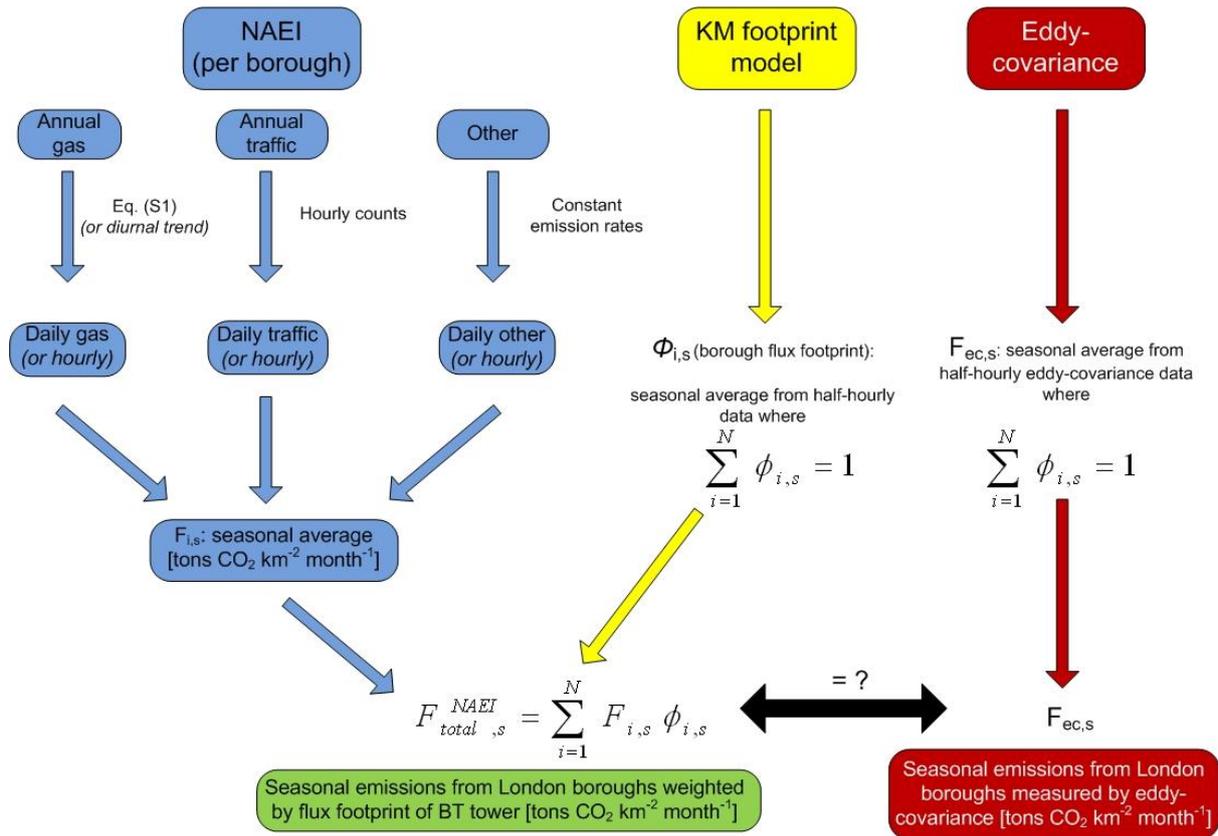


Figure S3: Summary of the procedure used to determine seasonal and – by simple up-scaling – annual bottom-up flux estimates of CO<sub>2</sub> from London boroughs from the NAEI, for comparison with the eddy-covariance measurements at the BT tower; annual emissions per borough obtained from NAEI were converted into seasonal estimates (diurnal trends were applied to natural gas usage and traffic data whilst other NAEI CO<sub>2</sub> sources were assumed to have constant emission rates throughout the year) and weighted by seasonal averages of flux footprint for the BT tower (obtained with the KM model). Finally, these bottom-up modelled emissions are compared with eddy-covariance estimates.

### 3 Results

#### 3.1 Flux losses due to high pass filtering

The size and frequency of the flux-carrying eddies scale with measurement height. Given the high measurement height, the largest potential flux losses would not be expected from the smearing of fast fluctuations by the limited frequency response of analyser or inlet system, but from low-frequency losses due to the limited flux averaging period of 30 minutes. In order to estimate this flux loss, half-hourly averages of sensible heat fluxes were compared

with 2-hour averages (Fig. S4) as described by Langford et al. (2010). This test provided an estimate of the proportion of low-frequency eddies not captured in 30 min averaging of high frequency data at this high measurement height.

Daytime sensible heat fluxes ( $H$ ) calculated for 2 h ( $H_{2h}$ ) periods were well correlated with fluxes obtained by averaging the four 30-min sub-periods ( $H_{4-30}$ ), in winter and summertime, with less than 1% discrepancy in either case. However, at night the  $H_{2h}$  values were 5% smaller than  $H_{4-30}$ , which could have been caused by non-stationarities resulting from changes in the nocturnal boundary layer. The comparison of  $H_{2h}$  and  $H_{4-30}$  during summer daytime conditions (Fig. S4d) shows more scatter, although no average bias. This is probably indicative of non-stationarities starting to affect the longer-term estimate.

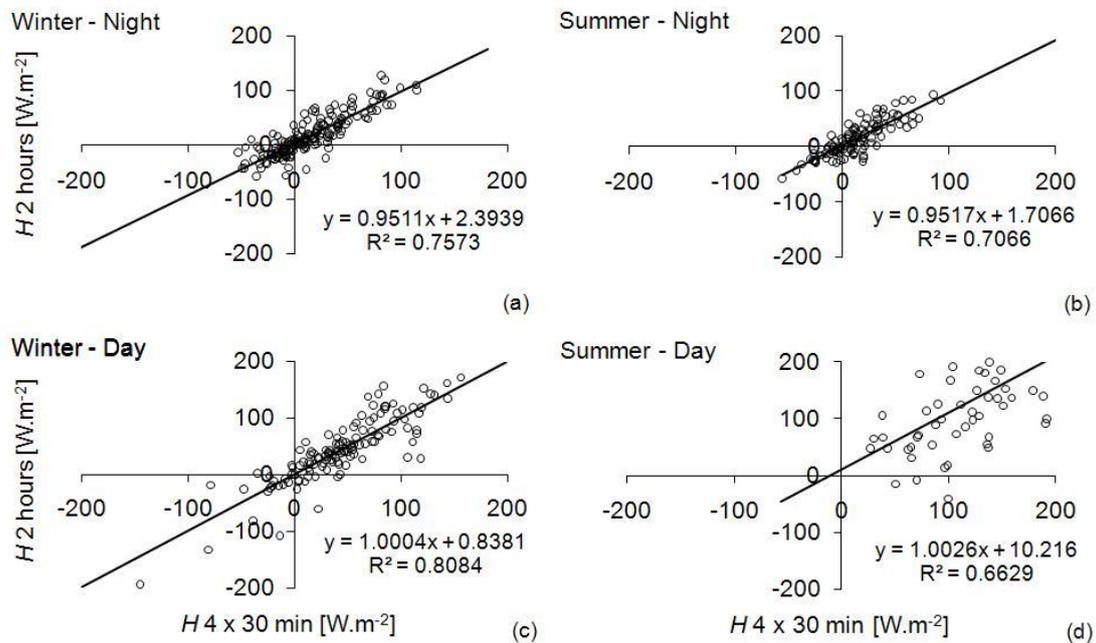


Figure S4: Sensible heat fluxes calculated using block-averaging of 2-h and 30 min (mean of the four periods). (a) Winter night, (b) summer (night), (c) winter day and (d) summer day.

### 3.2 Ecosystem exchange

Exchange rates of  $CO_2$  by vegetation in London were estimated using the methodology presented in Section 2.4 of the main article and summarised in Table S1 below.

Table S1: Estimated exchange of CO<sub>2</sub> by vegetation in the London borough of Westminster during the summer of 2007. Net exchange by trees is applicable to all London boroughs within the tower flux footprint.

Biogenic source / sink	Flux [t CO <sub>2</sub> km <sup>-2</sup> .month <sup>-1</sup> ]
Assimilation grass	- 305
Respiration grass	+ 150
Net exchange trees	- 19

#### 4 References

Langford, B., Nemitz, E., House, E., Phillips, G. J., Famulari, D., Davison, B., Hopkins, J. R., Lewis, A. C., and Hewitt, C. N.: Fluxes and concentrations of volatile organic compounds above central London, UK, *Atmos. Chem. Phys.*, 10, 627-645, 2010.

Nemitz, E., Hargreaves, K. J., McDonald, A. G., Dorsey, J. R., and Fowler, D.: Meteorological measurements of the urban heat budget and CO<sub>2</sub> emissions on a city scale, *Environ. Sci. Technol.*, 36, 3139-3146, 2002.

Rigby, M., Toumi, R., Fisher, R., Lowry, D., and Nisbet, E. G.: First continuous measurements of CO<sub>2</sub> mixing ratio in central London using a compact diffusion probe, *Atmos. Environ.*, 42, 8943-8953, 2008.