

Response to comments from reviewer 2.

We thank the reviewers for their constructive comments which we will endeavour to address to their satisfaction. Below are our responses, in chronological order, to the comments (in bold italics) received from reviewer 2.

Response to general comments

- 1) Please provide more information on how traffic emissions were derived. As traffic counts were available, emissions per vehicle class should be distinguished (passenger cars, vans, etc.).**

This has been clarified in the new Section 3.4.2.3 (Seasonal controls of CO₂ emissions) as follows (new text in bold).

“Measured winter time emissions of CO₂ in central London were estimated to have been dominated by natural gas emissions (71.1%) whilst traffic and other fossil fuel burning contributed 22.9% (Table 5). **It is important to emphasize that traffic emissions from eddy-covariance measurements were not estimated from traffic counts but taken as the difference between the total eddy-covariance flux and the cumulative contributions from other sources (natural gas, biospheric and human). The NAEI takes fleet composition into account for the purpose of estimating traffic-based emissions of CO₂.**”

- 2) The authors find evidence for an inversion layer during which ‘the tower height is above the layer connected to the urban surface below’. Please state more clearly whether CO₂ measurements observed under such conditions were included in the analysis – clearly the footprint model is not able to properly account for these situations.**

For the purpose of comparing NAEI bottom-up and eddy-covariance top-down fluxes, the footprint model was applied to daytime periods dominated by near-neutral or unstable stratification for which it was implicitly assumed that the tower was connected to the surface. This has been clarified in Section 3.1 as follows (new text in bold).

“(…) **For the quantitative comparison between measured fluxes with NAEI predictions, the footprint model was applied to daytime periods (09:00 – 18:00) dominated by unstable stratification for which it was implicitly assumed that the tower was connected to the surface. This is consistent with the study of Wood et al. (2010a) which shows that the tower is indeed coupled to the surface under both neutral and unstable stratification.**”

- 3) Sections 3.3 and 3.4 contain a lot of important findings. However, these findings are somewhat difficult to distil from the bulky text. The manuscript could gain a lot from a better organisation of these sections. Maybe it would help to shorten the sections slightly.**

Sections 3.3 and 3.4.2 were given further sub-sections to improve readability. The text was however not shortened as the level of detail provided seemed useful for the understanding of the underlying mechanisms.

- 4) *The results from the bottom-up study, i.e. the comparison of EC data with emissions from NAEI should be illustrated and highlighted more clearly.*

Section 3.4.2.2 (Comparison of bottom-up inventory (NAEI) with eddy-covariance results) was created in order to make the results of the comparison of NAEI with eddy-covariance data more readily accessible.

- 5) *Table 2 shows almost constant traffic counts for Marylebone road, while at three other locations, traffic decreased very significantly from June to October 2007. It should be stated whether this observation was specific to the selected year (road closure nearby?) or whether it was a re-occurring pattern. In either case, it does not seem justified to select only one dataset (traffic counts of a single road) for the analysis. This may raise some concern on the analysis.*

This is addressed in the new Section 3.3.1 (Seasonal variability). The new text in the fragment below is given in bold.

“Road transport emissions, which amounted to 33 % of the annual emissions estimates for the borough of Westminster in 2006 and 42 % for all boroughs (NAEI, 2009), were fairly constant from February to July 2007 (Fig. 5). Traffic volume measured on Marylebone Road decreased by ca. 6 % in December 2006, August - September 2007 and December 2007, compared with all other months, but this was not unequivocally linked to changes in CO₂ concentrations measured at the BT tower. Marked drops in traffic loads (Table 2) were recorded between June and October at three out of four Transport for London (TfL) monitoring sites (Fig. S1, Supplementary Material). Whilst these sites are located in the boroughs contributing the most to the flux footprint measured at the tower (Westminster, Camden and Kensington & Chelsea), [CO₂] did not reflect these changes in traffic loads. **This could suggest that traffic was not the foremost mechanism affecting seasonal variations in concentrations. Rigby et al. (2008), who studied CO₂ mixing ratios atop a tall tower at the Imperial College in London (UK) between August 2006 and June 2007 (Fig. 5b), report low concentrations in summer - limited by traffic and biospheric uptake - whilst increased values in winter time are attributed to added emissions from natural gas burning. Furthermore, summer time concentrations are lower than winter time ones across the entire dominant stability range (Fig. 6a), which suggests that dilution by entrainment does not control seasonal trends to a great extent.**

The extent of the apparent spatial heterogeneity of traffic loads could not be probed any further due to lack of detailed spatial data. However, Marylebone road provides excellent temporal resolution (continuous hourly data) for 2007. The spatial data may be impacted by local activities (e.g. localised traffic disruptions to the traffic census sites TfL 20, 21 and 35 in October 2007), or the extension of the Congestion Charge Zone (CCZ), implemented in February 2007, but the magnitude of these impacts are unknown. However, diurnal trends at the four TfL sites used in this study were strongly correlated with traffic counts observed at Marylebone Road. ($R^2 \sim 0.8 - 0.9$; data not shown) which supports the use of the latter site as a proxy representative of central London.”

Response to specific comments

- 1) **Page 3, line 7: The authors state that CO₂ concentration and CO₂ fluxes were measured 'semi-continuously' from October 2006 to May 2008. Please state more clearly what data was available from when exactly. How many flux data points were used for the present study?**

Measurements were taken continuously but due to instrument downtime and offline data filtering 100% of data coverage could not be achieved. The number of half-hourly data points available for each season has been added to Table 1. "Semi-continuously" was changed to "continuously" in the text to avoid confusion.

- 2) **Page 3, line 30: Canary Wharf seems to have a substantially different roughness length than other sites surrounding the tower. How was this accounted for? Furthermore, the surface of Canary Wharf seems difficult to model – with a few buildings around 200 m tall. Again, how did the authors deal with possibly induced turbulence and eddies in this area?**

This is discussed in the revised manuscript under section 2.3 (Comparison of emissions estimates with NAEI) as follows (new text in bold).

"There are no operational footprint models for urban environments that fully account for topography and spatial variations in building height and surface heat flux. Here, as an approximation, the analytical footprint model proposed by Kormann and Meixner (2001) was applied, which accounts for non-neutral stratification but assumes homogeneous surfaces. The aerodynamic roughness length for momentum (z_0) was estimated to be 0.87 ± 0.48 m in central London (Padhra, 2009), with 1 m used in the present study.

Canary Wharf, which is home to some of the tallest buildings in London, has roughness length of the order of 2 – 2.5 m which differs from that in more (vertically) homogenous areas. No special treatment was however applied to Canary Wharf as the wind occurrence from that sector was low (ca. 7% in 2007) as is its relative surface area. Furthermore, Wood et al. (2010a) show that the Tower Hamlets borough, where Canary Wharf is located, does not contribute to flux measurements on the BT tower during unstable conditions. "

- 3) **Page 9, line 9: Only the extent of footprints for unstable stratification is listed. Please mention the according figures for neutral and stable stratification.**

The corresponding values for stable and near-neutral stratification were added to the text.

- 4) **Page 11, line 12: The authors suggest that traffic emissions contributed to 'background CO₂ concentration rather than dominating its seasonal fluctuations'. This is somewhat counterintuitive. If possible, refer to literature with similar findings.**

This discussion now appears in a new section (3.3.1 Seasonal variability) and has been revised for clarity (see below, new text in bold). Reference is also made to the new figure 6,

suggested by reviewer 1, which illustrates the relationships between concentrations, fluxes, traffic and atmospheric stability.

“Road transport emissions, which amounted to 33 % of the annual emissions estimates for the borough of Westminster in 2006 and 42 % for all boroughs (NAEI, 2009), were fairly constant from February to July 2007 (Fig. 5). Traffic volume measured on Marylebone Road decreased by ca. 6 % in December 2006, August - September 2007 and December 2007, compared with all other months, but this was not unequivocally linked to changes in CO₂ concentrations measured at the BT tower. Marked drops in traffic loads (Table 2) were recorded between June and October at three out of four Transport for London (TfL) monitoring sites (Fig. S1, Supplementary Material). Whilst these sites are located in the boroughs contributing the most to the flux footprint measured at the tower (Westminster, Camden and Kensington & Chelsea), [CO₂] did not reflect these changes in traffic loads. **This could suggest that traffic was not the foremost mechanism affecting seasonal variations in concentrations. Rigby et al. (2008), who studied CO₂ mixing ratios atop a tall tower at the Imperial College in London (UK) between August 2006 and June 2007 (Fig. 5b), report low concentrations in summer - limited by traffic and biospheric uptake - whilst increased values in winter time are attributed to added emissions from natural gas burning. Furthermore, summer time concentrations are lower than winter time ones across the entire dominant stability range (Fig. 6a), which suggests that dilution by entrainment does not control seasonal trends to a great extent.**

The extent of the apparent spatial heterogeneity of traffic loads could not be probed any further due to lack of detailed spatial data. However, Marylebone road provides excellent temporal resolution (continuous hourly data) for 2007. The spatial data may be impacted by local activities (e.g. localised traffic disruptions to the traffic census sites Tfl 20, 21 and 35 in October 2007), or the extension of the Congestion Charge Zone (CCZ), implemented in February 2007, but the magnitude of these impacts are unknown. However, diurnal trends at the four TfL sites used in this study were strongly correlated with traffic counts observed at Marylebone Road. ($R^2 \sim 0.8 - 0.9$; data not shown) which supports the use of the latter site as a proxy representative of central London.

Based on traffic figures and the relationship between CO₂ concentrations and atmospheric stability it seems likely that temperature-dependent fluctuations in demand for natural gas combined with seasonal changes in biospheric uptake control the seasonal trends in atmospheric CO₂ concentrations. “

5) Page 12, line 6ff: Not clear, please re-formulate.

This paragraph has been rewritten and now reads “Analysis of mean concentrations as a bivariate function of wind speed and direction (Fig. 7c) shows that the largest concentrations were observed from the north and north-east of the tower especially at high wind speed ($\geq \sim 10 \text{ m s}^{-1}$). Rises in CO₂ concentrations in the north-east quadrant were not accompanied by surges in CO₂ fluxes, which were largest in the south-east quadrant where they ranged from $\sim 20 - 60 \mu\text{mol m}^{-2} \text{ s}^{-1}$, (Fig. 7d-f) whilst fluxes in the three other quadrants were in the $0 - 40 \mu\text{mol m}^{-2} \text{ s}^{-1}$ range. The level of CO₂ concentrations observed in the north-east quadrant could have been caused by horizontal transport (north-east winds in the UK are often associated with cyclonic transport of polluted European

continental air masses) and reduced vertical mixing. Averages for north and north-east conditions may further have been affected by the relative small number of observations under these conditions (Fig. 7g). This is also true for the south-east quadrant which had the lowest wind occurrence but the highest CO₂ fluxes.”

6) Page 12, line 17ff: Please explain – possible reason for findings?

Diurnal trends are now discussed in the new section 3.3.2 (Diurnal dynamics). The discussion has now been revised for clarity and now reads (new text in bold):

“...In winter and autumn a gradual decrease in concentrations occurs from ca. 10:00 until a minimum at mid-afternoon (ca. 14:00 - 15:00) (not shown). Concentrations increase from this point until ca. 17:00 - 18:00 after which they remain relatively constant throughout the night. These diurnal patterns are consistent with those observed in Edinburgh (Nemitz et al., 2002), Tokyo (Moriwaki and Kanda, 2004) and Basel (Vogt et al., 2006). In contrast, spring and summer concentrations have local minima at around 12 noon and 16:00, and local maxima around 14:00. Summer time concentrations were generally lower than any other season, whilst spring time values were the highest, especially at night time.

On a diurnal basis, CO₂ concentrations appear not to be correlated with traffic density with the minimum [CO₂] associated with afternoon peak traffic counts and whilst [CO₂] increased during the night when traffic counts were at their minimum. This is consistent with the observations made by Rigby et al. (2008) and can be explained by the overriding effect of the growth of the mixed layer and entrainment of air less concentrated in CO₂ from above during the day (Reid and Steyn, 1997). **As shown in Fig. 6a, there is a correlation between atmospheric stability (at the measurement height) and CO₂ concentrations in the near-neutral range, especially for values of the local stability parameter ($\zeta = z/L$) in the range -1.5 to +2, where over 90% of available data points were found. Strong positive linear dependencies were found in spring and autumn in the narrower stability range -0.5 to +1 (84% of the total annual data falls into this stability range); this is also true in summer for the stability range -0.5 to +0.5 (82% of the total annual data). In all three cases, [CO₂] exhibited an inverse correlation to traffic counts which supports the idea that entrainment / dilution is the dominant mechanism. No such trends were however observed during the winter period, perhaps due to increased emissions from natural gas burning, although this could not be verified.”**

7) Page 13, line 27: Please give examples for ‘other activities’.

This paragraph now reads “other activities such as demand for natural gas”.

8) Page 14, line 16ff: Is this possibly due to decoupling from the surface layer? Do typical wind directions differ for mornings and afternoons?

Morning and afternoon wind directions were not notably different; night time decoupling from the surface could have contributed to the observed trend although this could not be verified.

The paragraph now reads (new text in bold):

“Based on traffic data, the weekday / weekend comparison also suggests that the tower may have been decoupled from street level at night time when stable or neutral stratification were dominant throughout the year. Measured fluxes were very similar despite considerable difference in the traffic patterns: traffic was ~ 35% heavier between midnight and 04:00 at the weekends and reached a minimum at around 05:00 (possibly coinciding with night-club closing time and the resuming of public transport services). Traffic increased steadily - and almost linearly - thereafter before reaching an almost constant value from ~ 13:00 until 19:00. Weekday traffic volumes were on average 15% higher between 05:00 and 13:00 and grew more rapidly from 05:00 until 08:00. However, weekday and weekend CO₂ fluxes were almost identical in magnitude and trend between midnight and 08:00 which suggests that (a) the net CO₂ flux is dominated by other (non-traffic) sources, (b) that night time weekend Marylebone Road traffic loads are not representative of other areas within the flux footprint **or (c) that decoupling from the surface occurred at night (although it was not possible to determine how frequently this might have been the case).** Since there is no reason to assume that stability was (on average) different between weekdays and weekends, the lower flux / traffic ratio at weekends suggests that the night-time emission was dominated by non-traffic sources. While the nocturnal footprint was larger than during the day (and may therefore have been biased towards more residential areas) there was no significant difference in the wind frequency distribution during day and night-time.”

9) Page 23, Table 2: Please add figures for each month during the EC survey.

Only traffic counts for the months of March, June and October 2007 were available for the TfL sites, which was partly the reason for selecting Marylebone Road as a proxy for central London.

10) Page 26, Table 5: on page 16 it is stated that the emissions from natural gas usage etc. were estimated rather than observed. Please state this clearly in the Table captions.

The caption of Table 5 was amended as requested and now reads (new text in bold):

“Table 5: Comparison of observed fluxes with daytime (09:00 – 18:00) carbon dioxide fluxes (F_c), emissions from natural gas usage, biosphere exchange and human respiration (by season in 2007). The relative contribution from traffic emissions is the difference of F_c and ground sources. Natural gas usage was estimated from third-party data as described in Section 2.2 of the Supplementary Material.”

11) Page 31, Figure 5: Plot traffic counts of all sites.

All available traffic data obtained from Transport for London (TfL) was added to Figure 5 (see below).

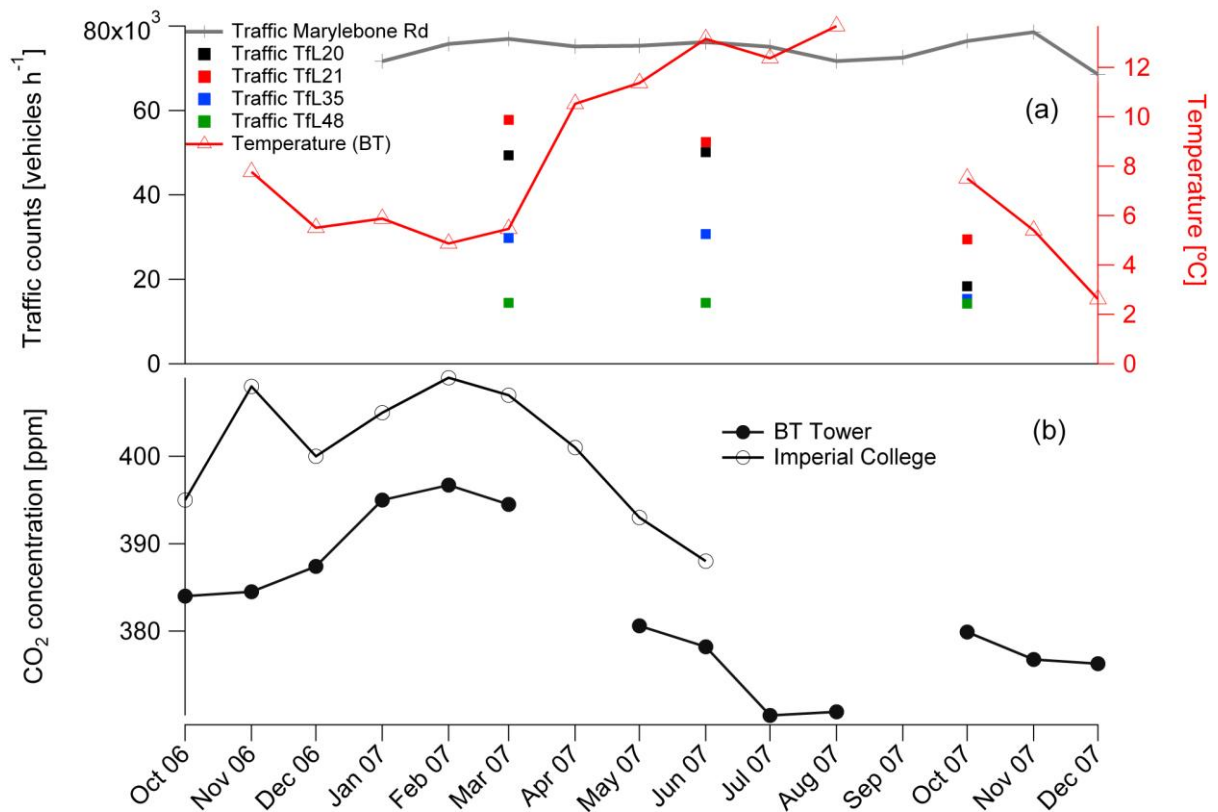
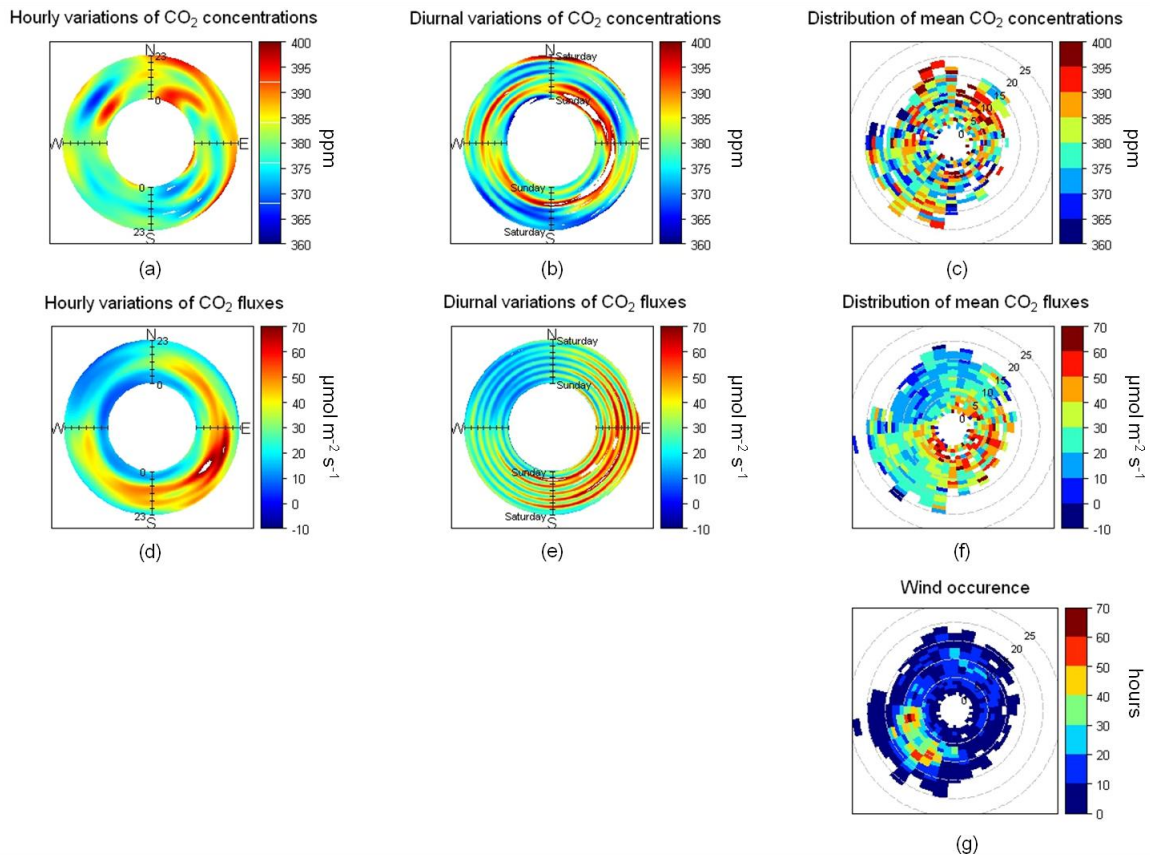


Figure 5: (a) Traffic counts at Marylebone Road and four Transport for London (TfL) census sites, and temperature measured at the BT Tower in 2007; (b) comparison of mean monthly CO₂ concentrations measured at the BT Tower and a tower site at Imperial College London (at 87 m) (Fig. S1, Supplementary Material) (Rigby et al., 2008). BT Tower data for April and September 2007 were excluded since the temporal data coverage was less than 10%.

12) Page 32, Figure 6: Please annotate colour bars with measures/units.

Units were added to the colour keys in Figure 6, which has become Figure 7 due to the insertion of additional material as requested by reviewer 1.



New Figure 7 (formerly Figure 6).

13) Page 32, Figure 6: Is there possibly a shadowing effect of the tower?

Work by Barlow et al. (2009)¹ and Wood et al. (2009)² does not find any evidence of such a shadowing effect. No further action was consequently taken on that point.

14) Page 33, Figure 7: Would results look similar for mean traffic emissions rather than traffic counts?

This would have been an interesting comparison indeed; however, mean traffic emissions are not available at the time resolution used for CO₂ fluxes. No further action was taken.

15) Page 34, Figure 34: Please state the period selected for winter, spring, summer and autumn.

¹ Barlow JF, Dobre A, Smalley RJ, Arnold SJ, Tomlin AS, Belcher SE (2009) Referencing of street-level flows measured during the DAPPLE 2004 campaign. *Atmos Environ* 43:5536–5544

² Wood CR, Arnold SJ, Balogun AA, Barlow JF, Belcher SE, Britter RE, Cheng H, Dobre A, Lingard JJN, Martin D, Neophytou M, Peterson FK, Robins AG, Shallcross DE, Smalley RJ, Tate JE, Tomlin AS, White IR (2009) Dispersion experiments in central London: the 2007 DAPPLE project. *Bull Am Meteorol Soc* 90:955–969

Seasons were defined in the caption of Figure 8 as requested. The caption now reads (new text in bold):

“Figure 9: (a) Monthly breakdown of CO₂ flux statistics for 2007 (bottom and top of boxes correspond to 25th and 75th percentiles, respectively; bottom and top whiskers correspond to 10th and 90th percentiles, respectively; monthly means and medians are indicated by solid markers and horizontal lines, respectively), and seasonal trends in (b) CO₂ and (c) latent heat fluxes. **Winter is defined as December 2006 – February 2007, spring is March – May 2007, summer is June – August 2007 and autumn is September – November 2007.**”

16) Page 35, Figure 35: Data should be plotted separately for winter, spring, summer and autumn.

Figure 9 has been revised and data is now presented per season. The caption was amended accordingly (new text in bold; see below).

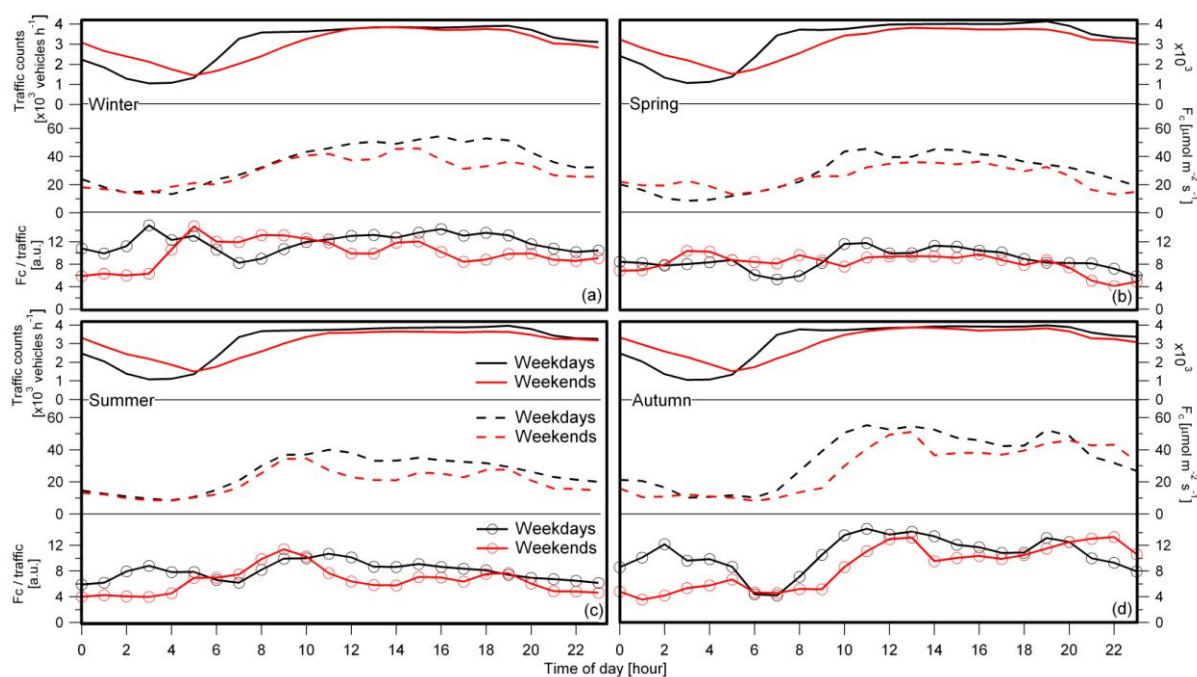


Figure 9: Weekday-weekend segregation of traffic counts (figures for Marylebone Road used as a proxy for central London), carbon dioxide fluxes measured at the BT Tower and ratio of fluxes to traffic in (a) winter 2007, (b) spring 2007, (c) summer 2007 and (d) autumn 2007.