Response to comments from reviewer 1.

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 1.

Abstract: I'm not convinced that the abstract highlights the key findings of the text. You report a number of interesting results both relating to the seasonal and diurnal trends in CO2 concentration and flux. These could be more clearly stated in both the abstract and conclusion. For example you note that diurnal traffic counts and CO2 fluxes are well correlated but in the text you demonstrate that this is really only the case during the daytime period under well mixed conditions. This is an important and interesting qualification to your results.

The abstract has been modified slightly in order to address the reviewer's comment (see below; new text in bold).

"Eddy-covariance measurements of carbon dioxide fluxes were taken continuously between October 2006 and May 2008 at 190 m height in central London (UK) to quantify emissions and study their controls. Inner London, with a population of 8.2 million ( $\sim$ 5000 inhabitants per km<sup>2</sup>) is heavily built up with 8% vegetation cover within the central boroughs. CO<sub>2</sub> emissions were found to be mainly controlled by fossil fuel combustion (e.g. traffic, commercial and domestic heating). The measurement period allowed investigation of both diurnal patterns and seasonal trends. Diurnal averages of CO<sub>2</sub> fluxes were found to be correlated to traffic but also exhibited an inverse dependency on atmospheric stability in the near-neutral range, with higher fluxes coinciding with unstable stratification during most seasons and perhaps reflecting how changes in heating-related natural gas consumption and, to a lesser extent, photosynthetic activity controlled the seasonal variability. Despite measurements being taken at ca. 22 times the mean building height, coupling with street level was adequate, especially during daytime. Night-time saw a higher occurrence of stable or neutral stratification, especially in autumn and winter, which resulted in data loss in postprocessing and caused the tower to become decoupled from street level. CO<sub>2</sub> fluxes observed at night were not always correlated with traffic counts, probably reflecting this decoupling, but also the fact that at night heating was always a larger source than traffic. No significant difference was found between the annual estimate of net exchange of  $CO_2$  for the expected measurement footprint and the values derived from the National Atmospheric Emissions Inventory (NAEI), with daytime fluxes differing by only 3%. This agreement with NAEI data also supported the use of the simple flux footprint model which was applied to the London site; this also suggests that individual roughness elements did not significantly affect the measurements due to the large ratio of measurement height to mean building height."

Introduction: This is well written and well referenced. Given that later in the paper you refer to the results from a number of key previous studies (Edinburgh, Vancouver, Tokyo, Basel etc.) it would be nice to see a brief summary of the key seasonal and diurnal trends observed and brief discussion of any differences observed between the sites. You could make more use of your tables as well.

More information on trends observed at other urban sites has been added as suggested by the reviewer (see below; new text in bold).

"In recent years, monitoring of carbon dioxide  $(CO_2)$  exchange with natural and semi-natural environments has benefited from communication through international flux networks (FLUXNET and its regional components such as CarboEurope, AmeriFlux, AsiaFlux, etc.). Within some of these communities considerable effort has been made to standardise measurement techniques and data processing methods (Aubinet et al., 2000; Foken et al., 2004; Moncrieff et al., 2004). Databases with long-term micrometeorological data, heat and trace gas fluxes for sites on five continents have been set up. These span latitudes from 30 °S to 70 °N and encompass boreal, temperate and tropical forests, wetlands, crops and tundra vegetation types (Baldocchi et al., 2001). In contrast, relatively few measurements of CO<sub>2</sub> exchange have been performed in urban environments, where over 50% of the world's population are estimated to live (United Nations, 2007)<sup>1</sup>, and the majority of studies were conducted in temperate areas of the Northern hemisphere, e.g. Basel, Switzerland (Vogt et al., 2006); Chicago, USA (Grimmond et al., 2002); Edinburgh, UK (Nemitz et al., 2002); Tokyo, Japan (Moriwaki and Kanda, 2004), as compiled in the database of the International Association for Urban Climate<sup>2</sup>. Although these studies vary in length, all found that the urban environment was a net source of CO<sub>2</sub>. Summer fluxes in Tokyo were comparable to those measured in Chicago and approximately half of mean Basel values. Values observed in central Edinburgh were the largest of all (e.g. four times Chicago values and twice Basel values) reflecting the strength of  $CO_2$  sources found in the footprint of the very central measurement location. Seasonal variations are important as demonstrated for example in Tokyo, where wintertime fluxes were more than twice their summertime counterparts due to changes in the demand for fossil fuels for domestic and commercial heating as well as photosynthetic activity. Despite a recent surge in publications pertaining to emissions from urban environments, data remains sparse compared to the natural environment. (...)"

Methodology: This was very clear. It might have been helpful to discuss the issues surrounding data coverage upfront however. For example you state that only 54% of the data was used- is this spread equally throughout the day and year? Are there any notable consistent patterns in data loss (i.e. night time during the winter?) What impact does this have on your conclusions? (You do comment on this sporadically through the paper e.g. section 3.4.1 - but it is hard to assess the overall impact.)

The following paragraph was added at the beginning of the Results section: "Data coverage was high in winter and summer (76% and 84% respectively) whilst less than 50% of seasonal data was available in spring and autumn (45% and 44% of data available, respectively). Instrument

<sup>&</sup>lt;sup>1</sup> http://unstats.un.org/unsd/demographic/products/dyb/dyb2.htm

<sup>&</sup>lt;sup>2</sup> http://www.urban-climate.org

downtime was the main cause of missing data throughout the 2007 study and since it occurred randomly, there is no reason to believe that diurnal trends discussed in this paper were biased towards daytime or night time regimes. The impact on seasonal trends is however more difficult to quantify, especially since it was the transitional seasons (here we mean transitions between regimes of vegetative dormancy and high natural gas demand towards heightened vegetative activity and reduced needs for domestic and commercial heating) that were the most affected. It was therefore assumed that the available data was representative of the whole season in each case."

Results:

## 1) In general these are well written. However, the figures could be more carefully woven into the discussion. For example I found Figure 6 and the related discussion quite confusing. The discussion of this figure could be clarified. Perhaps it would be helpful to plot a diurnal graph of two of the key wind directions to illustrate your points.

We appreciate that the readability of the discussion of figure 6 can be improved; we will however try to achieve this by text only as the manuscript contains a rather large number of figures.

## 2) "A diagram to illustrate the seasonal variability in diurnal fluxes would also be helpful as discussed on p23753 second paragraph".

This comment was difficult to interpret as the second paragraph on page 23753 discusses concentrations rather than fluxes. In addition, the diagram suggested by the reviewer already exists (Fig. 8b). Consequently, no further action was taken for this comment.

## 3) "Can you plot Figure 7 by stability as well?"

This is a very interesting point and we are grateful to the reviewer for raising it.

This point is discussed in the new sub-section 3.4.2.1 (traffic) which now reads (new text in bold):

"Possible reasons for the non-linearity in the traffic dependence of the flux include: (a) other activities, such as demand for natural gas, scale non-linearly with traffic, (b) fuel consumption increases at high traffic volumes, (c) the Marylebone Road traffic counts underestimate the full dynamic range of the average traffic activity in the flux footprint due to saturation at high traffic volumes, and (d) changing meteorology (e.g. boundary layer height) with season at rush hours.

The latter is illustrated in Fig. 6c which shows that high traffic counts were generally recorded at times dominated by unstable stratification.  $CO_2$  fluxes ( $F_c$ ) are inversely correlated to stability (i.e. unstable stratification is often accompanied by higher flux values), as shown in Fig. 6b and positively correlated to traffic counts (6d). The relationship between traffic counts and stability

(this is obviously not causal) is of a sigmoidal nature over the stability range -1.5 to +2, where over 90% of available data points were found. In light of this, it is difficult to quantify unequivocally how much of the variability in the  $CO_2$  flux measured well above the city is associated with the variability in traffic volume and how much reflects changes in atmospheric factors."

We propose to add a new figure (Figure 6, below) in which traffic counts,  $CO_2$  fluxes and concentrations are plotted as a function of the atmospheric stability parameter. The revised manuscript will discuss the points raised above.



Figure 6 (new figure; the old Fig. 7 is now Fig. 6d): (a)  $CO_2$  concentrations, (b)  $CO_2$  fluxes, (c) traffic counts as a function of atmospheric stability and (d)  $CO_2$  fluxes as a function of traffic counts.

## Conclusion: In your conclusions you recommend that concentration gradient measurements be made to reducing remaining uncertainties about storage. The limitations of not having this information and their implications for the study are not adequately discussed in this paper.

Discussion of this comment has been added to the revised manuscript which now reads (new text in bold):

"The close agreement between measured fluxes and bottom-up emission inventory estimates (within 3% during daytime) suggests that the 190 m tall BT Tower in central London was a suitable measurement site for characterising average emissions during daytime, while some care needs to be taken to interpret night-time fluxes. This would imply that long-term flux measurements can be used to track changes in emissions, and to quantify emissions of pollutants for which urban area sources are more poorly understood and more variable than those of CO<sub>2</sub>. Measurements for a year reveal that the seasonal dynamics of atmospheric carbon dioxide concentrations are strongly linked to the natural background vegetation cycle. Seasonal variations in natural gas consumption (e.g. commercial and domestic heating, referred to as background emissions) and their diurnal trends were regulated by dilution, consistent with cycles of growth and shrinkage of the boundary layer. Diurnal fluxes of CO<sub>2</sub> were strongly correlated with traffic counts and, to a lesser extent, with atmospheric stratification in the dominant stability range (-1.5 to + 2). Because traffic correlates with atmospheric stability it is difficult to untangle the two effects. Night-time fluxes were not unequivocally correlated to traffic counts which could be explained by decoupling of the tower from street level and / or dominance by non-traffic sources. Gradient measurements would have provided useful information on the storage term and periods of decoupling from the surface. It is recommended that future flux measurements at such high measurement heights be accompanied with concentration gradient measurements to reduce remaining uncertainties due to storage effects. However, we were able to demonstrate good agreement between the eddy-covariance results and a bottom-up inventory which can be considered as a validation of both approaches as far as daytime averages are concerned. Without gradient data we can only speculate as to the causes of the discrepancies (although not statistically significant) between inventory and eddycovariance data but there is no indication that the absence of this information had a critical impact on the analysis carried out in this study.

Seasonal variations in heating-related emissions and vegetative assimilation through photosynthesis during the growing season had an impact on CO<sub>2</sub> fluxes measured at the tower. Whilst the urban environment of central London was a net source of CO<sub>2</sub> in 2007, summer net emissions were 20% lower than their wintertime counterparts due to the combined effects of a reduction in heating emissions and uptake by photosynthesising plants. Natural gas demand was found to dominate emissions (ca. 71%) in winter, whilst a more balanced partitioning, comparable to values reported for Edinburgh by Nemitz et al. (2002), was observed in autumn: 47.8% of autumn CO<sub>2</sub> emissions were attributed to natural gas burning, compared to 48.9% for traffic. Gross plant assimilation in the central London borough of Westminster - where green spaces account for ca. 15% of borough surface area compared to ca. 8% elsewhere - was estimated at 4330 t CO<sub>2</sub> y<sup>-1</sup>, which represents only 0.4% of the total annual emissions. Grass was estimated to make the largest contribution to net assimilation, with trees being responsible for a mere 5%. These results suggest that the total surface area allocated to green spaces would have to be increased by a factor of 250 in order to neutralise anthropogenic emissions of carbon dioxide in central London. Considering how heavily-urbanised the studied area was in 2007, it seems likely that central London will remain a strong source of  $CO_2$ in the foreseeable future. It is recommended that future flux measurements at such high measurement heights should be accompanied with concentration gradient measurements to reduce remaining uncertainties due to storage effects."