We would like to thank reviewer 2 for their very helpful comments and suggestions. We will respond to each comment in turn below.

RC: Usually, ground flux measurements are conducted at the top of mast or scaffolding towers between two and four times higher than the average height of the roughness elements (see the Urban Flux Network, http://www.geog.ubc.ca/urbanflux/index.html) with the aim to be in the constant flux layer and observe fluxes from a spatial scale (10^2 - 10^4) of similar size to the cells in gridded emissions inventories. In this case the flux measurements were conducted at 200 m above ground, more than 13 times the average height of the surrounding buildings (page 17305, line 26). At this height many of the measured fluxes might not be representative of the ground fluxes, in particular during stable conditions at nighttime and cloudy days. The authors recognize that in some nights the measurements were de-coupled from the inertial sublayer (page17308, line 20) and were not representative of the ground fluxes. However, it is not clear how those periods were identified, neither if they were removed for further analysis.

AR: Without supporting measurements of boundary layer height from Lidar measurements it is impossible to unambiguously identify and remove these time periods. Therefore we chose to present all of our measured data, and discuss them in terms of local fluxes at the measurement height. This also makes sense in terms of chemical degradation mentioned by this referee below. We acknowledge in the text that the measured fluxes may not always be representative of the emissions occurring at the surface. However, we believe that the integration of our measurements across the day provides a reasonable daily flux value and we feel we are able to give a robust estimate of the fluxes occurring at this site.

RC: The estimation of the boundary layer height using a model, which cannot estimate heights below 250 m is useless to determine measurement periods de-coupled from the inertial sublayer. An alternative model needs to be used to determine those periods. Also the routine radio-sounding from the closer airport may be used.

AR: We contacted London City airport to enquire about LIDAR and radio sonde data. The airport could not supply us with LIDAR data and informed us that no airports in the SE of England use radio sondes due to the high density of air traffic. Therefore in the revised manuscript we will use an alternative boundary layer height model by Arya (1981). A comparison of this model against LIDAR measurements made during the subsequent REPARTEE II campaign showed reasonable agreement, and allowed for it's optimisation for for this site. This analysis gives us confidence in the model and a preliminary run for REPARTEE I using this model clearly indicates the boundary layer to be below 200 m on certain nights. The results from this model will be included in the revised manuscript and will replace the estimates currently shown in Fig 5.

Following the rule-of-thumb that says that typically the observed fluxes correspond to footprints between 100 and 300 times the measurement height (Grimmond and Oke, 1999), footprints between 18.5 and 55.5 km might be expected from the Telecom tower

(considering a measurement height of 185 m, zm = ztower – z0). Using a simple footprint model (Hsieh et al., 2000) and Zm / L = 1 for stable conditions, it was found that the fetch (80% of the flux) might extend up to 77.7 km. For unstable conditions (Zm / L = -1) it was found a similar fetch to that reported in the manuscript (page 17305, line 27). However, the extended footprints might jeopardize the identification of emission sources. During long periods of the day, the measured fluxes might include fluxes from the whole city and beyond its boundaries. The authors need to calculate the footprint for a wider range of atmospheric conditions, and explain how the periods with very long footprints were considered for further analysis, in particular for the comparison with the emissions inventory.

AR: The problem of (locally) stable conditions is not the extent of the footprint, but the decoupling. Due to the anthropogenic heat output, sensible heat fluxes from the urban matrix tend to be positive during night as confirmed with a sonic anemometer which was mounted at a lower height (Janet Barlow, pers. commun.; e.g. Curtis et al.,). Therefore footprints would always be expected to be well within the London conurbation. However, on the Tower negative heat fluxes were measured during some night-time periods, hinting at the presence of an inversion layer between the ground and the tower measurement height. Thus, negative heat fluxes bring us back to the question of the decoupling addressed in response to the previous comment. In addition, tracer release experiments (Petersson et al., 2009) confirmed that, during daytime, tracers emitted about 2 km upwind were picked up at the tower sampling height, supporting the view of efficient vertical mixing during the day.

RC: The disjunct eddy covariance technique does not allow an inspection of the spectra and cospectra of the measured variables. However, the parallel flux measurements of CO and CO2 by the traditional eddy covariance method might be used to investigate the influence of the measurement system in the attenuation of the turbulent signal. The massive Telecom tower might have a significant influence on the turbulence pattern.

AR: We agree with the reviewer that the Telecom tower could influence our turbulence measurements. However, the 15m lattice structure on top of the tower makes this a relatively ideal urban measurement location. Indeed, using the data available to us, our analysis of the integral turbulence statistics shown in the discussion paper suggests that wake turbulence from the tower structure was within an acceptable range, and so was the vertical angle of the anemometer rotation. Within our manuscript we also direct readers to papers by Wood et al. (2009, submitted to BLM) and Helfter et al. (2009, in preparation for ACPD) who provide a more detailed analysis of the turbulence observed at this site.

RC: As suggested in the section describing the ratios between VOC and CO (page 17311, line 15), the chemical degradation might have an important impact on the measured fluxes because of the height at which the measurements were conducted. It is necessary

to asses the sensitivity of the VOC fluxes to chemistry, in particular the isoprene fluxes, since isoprene is a very reactive VOC. This can be done applying a transport model coupled with a simple chemical mechanism, such as the model proposed by Rinne et al. (2007). The life time of the measured species under the chemical conditions of the atmosphere of London needs to be clearly longer than the time consumed by an air parcel to reach the top of the flux tower.

AR: We did discuss the effect of chemistry on the measured local fluxes in the manuscript, but did not conduct an analysis of the time-scales involved.
The reviewer makes a good point here and we agree it is important to show the sensitivity of the measured VOC to chemistry given our very high measurement location. With this in mind we calculated the daytime convective velocity scale:

$$t_* = \frac{z_m}{w_*}$$

Where z_m is the measurement height (200 m) and w_* is equal to:

$$w_* = \left(\frac{g \times zi}{T_v} F_H \right)^{\frac{1}{3}}$$

***zi* is the measurement height (800 m), *g* is the acceleration due to gravity, T_v (285 K) is the virtual temperature and F_H (1.1143 m s^{-1}) is the kinematic heat flux.**
This gives a transport time of approximately 1 minute, which is considerably shorter than the typical atmospheric lifetime of isoprene (the most reactive compound measured during our study) which is approximately 1.4 hours (Atkinson, 2005). From this we conclude that our flux measurements are not sensitive to chemistry.

RC: Both, ambient concentrations and fluxes need to be analyzed in terms of wind direction. Currently the manuscript does not consider the wind direction in the results discussion. For example, winds blowing from the north might be related to higher VOC fluxes and concentrations due to the presence of a large number of important roadways in that direction. In the same context, the Regents Park located 1 km from the Telecom tower might produce larger fluxes of isoprene due the biogenic emissions when the wind blows from the northwest direction. For the ambient concentration discussion is necessary to keep in mind that concentrations depends on emission patterns, meteorology and chemical processes, while the fluxes depend mainly on the emissions and a few meteorological variables.

AR: In our revised manuscript we will analyse our data with respect to wind direction and include a new figure showing the normalized concentration and flux

wind sector dependence plots for each of the measured compounds as well as a wind frequency chart.

RC: It is not completely clear how the comparison between the measured fluxes and estimated emissions in the official emissions inventory was conducted. The only information provided of the emissions inventory is the grid scale; nothing is said about the temporal and seasonal distributions. Does the emissions inventory provide hourly, daily or annually emissions of anthropogenic and natural sources? How many species include? Certainly, more information regarding to the emissions inventory is required, as well as an explanation of the method used to extract the VOC emissions from the grid cells. Finally, if the parameterization used to evaluate the emissions inventory represents only a "snap-shot" of the annual emissions and considering that the seasonal changes in the VOC emissions are significant, a direct comparison of the measured fluxes with the emissions estimated for a day of October in the emissions inventory might be more appropriate.

AR: In response to a similar comment made by reviewer 1 we have added a more detailed description of the comparison as well as the methodology used by the NAEI to generate its emission estimates.
Briefly, the NAEI routinely provide detailed annual emission maps for ~ 25 important atmospheric pollutants. Of these, only two VOCs, benzene and 1-3 butadiene are explicitly included as well as total VOC emissions. The total VOC emission map contains emissions from >540 compounds which are listed along with their source apportionment in the NAEI VOC speciation database (available on request from www.**naei**.org.uk). **Emission maps for any of the 545 compounds can then be manually separated out from the total VOC emission map using the VOC speciation.**
As we will make clear in our revised manuscript, the NAEI emission maps only provide annual data. Unfortunately this severely limits the comparisons that can be made between our measured fluxes and the inventory. We agree with the reviewer that a comparison between fluxes on a shorter time scale (for example Velasco et al. (2009)) would have been far more informative, but in this instance this was not possible.

Specific and technical comments
1) Page 17299, line 13: Amman et al. (2006) reports water vapor fluxes and not fluxes of biogenic VOC as indicated in the text.

AR: We thank the reviewer for pointing out our mistake. We will remove this reference in the revised manuscript.

2) Page 17299, line 17: Please define a suitable elevation above street level to perform eddy covariance measurements.

AR: Measuring fluxes above urban areas is complicated as the surface is highly irregular and often roads, buildings, urban parkland and construction sites are

found in close proximity. Thus, flux measurements need to be made above the blending height, at which the small scale fluxes merge into a net flux and the conditions for eddy-covariance (homogeneity, stationarity) are fulfilled. For example, Nemitz et al. (2002) and Dorsey et al. (2002) concluded that fluxes could be reliably measured over an urban environment provided the measurement height exceeded at least 3 times the average building height. Similarly, in this study, the fluxes fulfilled these criteria. We will make a statement in the revised manuscript to this effect.

3) Page 17299, line 17: To reduce the impact of the surface heterogeneity, variability on the emissions distribution and effects of advection, the eddy covariance measurements are limited to fairly uniform urban areas in terms of topography and patterns of buildings, roads, emission sources and vegetation.

AR: We thank the reviewer for this suggestion and will include these comments within our revised manuscript.

4) Page 17299, line 3: Instead of "emission estimates" better use "fluxes". Emissions refer only to upward fluxes. A downward flux is called deposition.

AR: "Emission estimates" will be changed to "fluxes"

5) Page 17300, line 3: Which were the prevalent meteorological conditions during the field study?

AR: The campaign meteorology has been discussed in detail by Dall'Osto et al. (2009) and Nemitz et al. (2009). In the revised manuscript we will direct readers to these descriptions and we will include a brief description of the prevalent meteorological conditions.

6) Page 17300, line 5: In two lines indicate the main objectives of the REPARTEE project. Not all readers are familiar with it, in addition that (Dall'Oslo et al., 2009) is still under preparation.

AR: A short summary of the main objectives of the CityFlux project and the REPARTEE campaign will be added to the revised manuscript.

7) Page 17300, line 12: Please indicate the average height of the surrounding buildings.

AR: The average building height (15 m) will be included in the revised manuscript.

8) Page 17300, line 13: Please indicate the land cover fraction of buildings, vegetation and impervious ground of the monitored district, at least during daytime under convective conditions (_4.7 km).

AR: The fraction of land cover within the daytime flux footprint (4.7 km) is as follows:

Buildings/roads = 82%
Urban parkland = 13%
Impervious ground = 5%

This information will be incorporated into the revised manuscript.

9) Page 17301, line 7: Which was the eleventh monitored mass? Only 10 masses are indicated in the text.

AR: The eleventh mass monitored was m/z 43. This unidentified mass was only measured between the 19th – 30th October and therefore was not included within the final analysis.

10) Page 17301, line 12: Indicate that m/z 107 (C8-aromatics) includes the three xylone isomers and benzaldehyde, in addition to ethylbenzene.

AR: We will make the suggested change in the revised manuscript.

11) Page 17302, line 5: Please provide more details of the flux post-processing. Was the Web correction applied? What about the presence of possible trends in the 30-min series, were they eliminated using a low pass filter?

AR: The WPL correction was not applied to our flux data. A coordinate rotation was applied to turbulence data which was subsequently block averaged. This additional post-processing information will be added to the manuscript.

12) Page 17305, line 1: Was the same fraction of rejected periods due to stationarity and limit of detection for all the monitored species?

AR: No, the rejected fractions differed slightly (but not significantly) between each compound. The numbers presented in the manuscript represent the average values.

13) Page 17305, line 5: Which were the criteria to consider a data period of high or low quality?

AR: The stationarity test followed the theory outlined by Foken & Whichura (1996), which states that a time series χ is stationary, when the flux ($F\chi$) is equal to the mean average flux of its components ($F\chi1$, $F\chi2$, $F\chi3$…). Here we took $F\chi$ to be the flux over the averaging periods, and the components $F\chi1$ to $F\chi6$ to be the flux calculated from individual 5 minute blocks of the original time series. Following criteria specified by Velasco et al. (2005; 2009), if the mean of $F\chi1$ – $F\chi6$ differed by more

than 60% of the value of $F\chi$ the time series was considered non stationary and the data were discarded. Time series where the fluxes differed between 30% and 60% were considered stationary, but to be of a lower quality. High quality stationary data was taken to be any time series where the fluxes differed by less than 30%.

14) Page 17306, line 8: In addition to the evolution of the boundary layer and emission patterns, the ambient concentration of any pollutant depends also on wind transport and chemical processes. The finding that the highest VOC concentrations were observed for oxygenated species shows the importance of the photochemistry in the VOC levels in the atmosphere of London.

AR: We thank the reviewer for this observation, which we will incorporate into the revised manuscript.

15) Page 17306, line 13: How does the diurnal profile of CO compare to the diurnal profiles of VOCs?

AR: As you would expect, the diurnal CO profile is very similar to that of the aromatic VOCs measured, but there are slight differences hence our analysis of the VOC/CO ratio and its evolution over the course of the day. As we mention in our response to reviewer 1, the CO and CO_2 flux data measured during the campaign is being written up in separate papers by Helfter et al. (2009) and Phillips et al. (2009) and will be submitted to the special issue very soon. These papers will include the diurnal profiles of CO and CO_2 and therefore we do not wish to present the same data again here.

16) Page 17307, line 8: Is the benzene to toluene ratio in units of ppbv/ppbv or ppbC/ppbC?

AR: The units are ppbv/ppbv.

17) Page 17307, line 9: Is there any reason for this significant increment in the benzene to toluene ratio?

AR: The ratio increases from the 19-30 October as toluene concentrations show a marked decrease from the period of 01-13 October. When the ratio was high, benzene and toluene concentrations and fluxes showed very similar trends suggesting a similar source (traffic). When the ratio was low, toluene concentrations were larger and show a very different trend to benzene, whereas toluene and benzene fluxes still show a similar trend. As we state in the manuscript, this is strong evidence for a source of toluene outside of our flux footprint between 01-13 October which are contributing to elevated concentrations, but not fluxes.

18) Page 17307, line 16: Were the canister samples collected also at 200 m of height?

AR: The canisters did not sample from directly below the sonic anemometer. Instead they sampled air from a balcony approximately 30 m below. We will include this information in our revised manuscript.

19) Page 17308, line 9: If this statement is true, one of the basic assumptions of the eddy covariance technique is not fulfilled. The monitored footprint must be homogeneous in terms of topography and emission sources to avoid interferences by advection.

AR: All urban flux measurements will, to some extent, be affected by advection errors, which cannot be quantified by single point measurements. Because London is a large conurbation (megacity), we believe advection effects to be relatively small, but we do not have the data to confirm this.

20) Page 17308, line 19: Please indicate for all the times within the text if they are referred to UTC or local time? Because of the strong relationship between VOC fluxes and anthropogenic activities, it might be better to use local time.

AR: The reviewer makes a good point. As the emissions are closely linked to anthropogenic activities we will change the times from UTC to Local time.

21) Page 17308, line 23: According to Fig. 5, the boundary layer height passed from 600 m at midnight m _400 m at noon, being always above the measurement height.

AR: As we point out within the text, the de-coupling from street level emissions occurred occasionally and was not observed every day. In figure 5, this is much more apparent in Panel A then B. Therefore after consideration we have decided to remove panel B from this figure.

22) Page 17309, line 18: Please define “cold starts”.

AR: “Cold starts” refers to the starting of an automobile when the engine is cold. A definition will be inserted into the revised manuscript.

23) Page 17310, line 16: Define the acronym HGV.

AR: We will define Heavy Goods Vehicles (HGV) in the revised manuscript.

24) Page 17310, line 27: Provide examples of other major toluene sources.

AR: Anthropogenic sources of toluene include industrial solvents, paint thinners, ink and paint manufacturing and printing. These examples will be added to the text.

25) Page 17311, line 5: The units of ppvb-1/ppbv-1 are not clear. They must be ppbv of VOC /ppbv of CO

AR: We will make the reviewers suggested change in the revised manuscript.

26) Page 17311, line 8: Change etheylbenzene by C2-benzenes. See comment 10.

AR: Ethylbenzene will be changed to C_2-benzenes throughout the text.

27) Page 17312, line 9: Was the flux analysis in terms of traffic activity restricted to periods with north winds?

AR: No, all available data was used in the comparison with traffic counts.

28) Page 17313, line 14: Which are those potential sources of methanol?

AR: Methanol is emitted from numerous biogenic sources, including direct emissions by plants (Huve et al., 2007), leaf wounding (Warneke et al., 2002) and also from soils (Schade and Custer, 2004) during the degradation of plant material (Warneke et al., 1999). Anthropogenic sources include industrial solvents, adhesives, dyes, paints, varnishes and antifreeze. We will mention these sources in the revised paper.

29) Page 17313, line 17: It is true that the biogenic emissions of isoprene depend on ambient temperature and solar radiation, but there are other parameters which might be also important under certain conditions, such as the plant phonology, relative humidity, pollution and water stress, etc. (see Guenther et al., 2006).

AR: We thank the reviewer for pointing out these additional parameters which we will include within our revised manuscript.

30) Page 17315, line 7: Which footprint area was used to extract the emissions from the emissions inventory?

AR: As our night time flux measurements are not always representative of the emissions occurring at the surface, we use the typical daytime flux foot print (4.7 km).

31) Page 17315, line 8: Which of the measured VOCs are not reported in the emissions inventory?

AR: All the measured VOC are included within the emission inventory. However, the NAEI only generates emission maps specifically for Benzene. Emission maps for the remaining compounds have to be calculated manually using the Total VOC emission map and the VOC speciation data base which are both provided by the NAEI (see our response to an earlier comment). We will make this clear in the revised manuscript.

32) Page 17315, line 11: The statement that the flux estimates are larger than NAEI estimates in all cases is not true. According to Fig. 9 the benzene emissions in the emissions inventory are larger than the emissions estimated in this work.

AR: We thank the reviewer for pointing out our mistake. We will make the necessary changes in the revised manuscript.

33) Page 17315, line 19: (Polson et al., 2009) is missing in the reference section.

AR: We thank the reviewer for pointing out our oversight. We will insert the reference in the revised manuscript.

34) Page 17313, line 20: According to the references Famulari et al. (2009) report NOx fluxes.

AR: The paper of Famulari et al. (2009) reports nitrous oxide (N_2O) fluxes above Edinburgh in relation to fluxes of CO_2 and CO. It describes in more detail the CO flux system also used in this study and compares fluxes of N_2O, CO_2 and CO with the NAEI values for Edinburgh. The paper is now available online.

35) Page 17316, line 7: The statement that the traffic contribution to the VOC fluxes changes from season to season was not demonstrated in this study.

AR: Our analysis shows that for isoprene the relative contribution from traffic is strongly dependent on temperature (season). We will rephrase this sentence to read "Traffic within the city has been shown to be the primary source of VOC fluxes to the atmosphere within central London, but its relative contribution varies from compound to compound and also temporally, with changes occurring from hour to hour and in the case of isoprene from season to season."

36) Page 17323, Table 2: It will be good to include the ±1 standard deviation of the 24-h flux averages listed in this table to have an idea of the day to day variability. The average toluene flux reported by Karl et al. (2009) is 14.1±4.0 mg m-2 h-1.

AR: We agree with the reviewer and will insert the ± 1 standard deviation to the fluxes shown in Table 2.

37) Page 17323, Table 2: The VOC fluxes inside of a city vary according to the emissions sources in each neighborhood. For example the fluxes reported by Karl et al. (2009) correspond to the industrial sector of Mexico City, while the fluxes reported by Velasco et al. (2009) correspond to a busy residential neighborhood. To give a better idea of the average fluxes measured in previous studies it will be good to add a column to indicate the type of monitored district in each study.

AR: This is a good suggestion and we will include an additional column detailing the monitored district of each study.

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