

***Interactive comment on “Atmospheric chemistry and physics in the atmosphere of a developed megacity (London): an overview of the REPARTEE experiment and its conclusions” by R. M. Harrison et al.***

**R. M. Harrison et al.**

r.m.harrison@bham.ac.uk

Received and published: 16 February 2012

Atmospheric Chemistry and Physics Discussions

Interactive comment on “Atmospheric chemistry and physics in the atmosphere of a developed megacity (London): an overview of the REPARTEE experiment and its conclusions” by R. M. Harrison et al.

FINAL AUTHOR COMMENTS

The authors are most grateful to the reviewers for their thorough and thoughtful com-  
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mentaries on this paper. We are pleased to respond as follows:

#### Anonymous Referee #1

My main concern is the representability of the sites, and of the measurement period. For a reader not so directly connected with urban measurements, it would be very useful to have a concise section to explain how representable are the London measurements to other European metropolies. On some of measurements, comparative values from other cities and earlier studies are mentioned, but for some, no mention is made. It would be very useful to also mention, if this is the first time such and such measurement is done (at least to author's knowledge). Particular factors only observed in London should be mentioned, e.g. does the congestion charge affect the traffic patterns and thus the emission factors? Secondly, the periods of measurement are rather short, for understandable reasons. There should, however, be discussion on how representative the periods are of the overall pollutant levels on the other parts of the year. RESPONSE: The reviewer raises very valid points which we have done our best to address through additional text added to the synthesis and conclusions section. This is reproduced below.

“A key question is how representative the sampling sites are of Europe in general, and whether the two one-month snapshots well represent the longer-term position. Comparative data for particulate matter are available from Putaud et al. (2010). Concentrations of PM<sub>10</sub> and PM<sub>2.5</sub> at urban background sites in London (including North Kensington, as used in REPARTEE) are fairly typical of north-western Europe (excluding Scandinavia), similar to central Europe and generally lower than southern European sites. It is less easy to compare chemical composition, as analytical protocols were not identical across Europe, but the composition of PM<sub>10</sub> at London sites is fairly typical, as is the number concentration of sub- $\mu\text{m}$  particles (Putaud et al., 2010). Data for roadside sites is less abundant, but Marylebone Road lies within the range of other European sites (Putaud et al., 2010).

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In REPARTEE I, the pollution levels were fairly typical of the annual mean. PM<sub>10</sub> at NK averaged 26  $\mu\text{g m}^{-3}$  during the month of REPARTEE, while the annual mean was also 26  $\mu\text{g m}^{-3}$ . At M. Road, the monthly mean was 56  $\mu\text{g m}^{-3}$ , compared to an annual mean of 47  $\mu\text{g m}^{-3}$ . The month of REPARTEE II was also fairly representative with the monthly PM<sub>10</sub> averages at NK and M. Road respectively 29  $\mu\text{g m}^{-3}$  and 48  $\mu\text{g m}^{-3}$  compared to annual means of 25  $\mu\text{g m}^{-3}$  and 45  $\mu\text{g m}^{-3}$ .

The site at M. Road is on the northern boundary of the London congestion zone. Introduction of the zone had a relatively minor impact upon the emissions of traffic-generated pollutants (Beevers and Carslaw, 2005) and consequently only a small effect upon pollutant concentrations (Atkinson et al., 2009). The weather during the two campaigns was fairly typical of the autumn in London. Weather during the REPARTEE I campaign was markedly warmer than in REPARTEE II (mean 15.5°C and 10.4°C respectively). This compares with long-term averages for October of 12.05°C (corresponding to REPARTEE I) and 9.9°C for the mean of October and November (corresponding to REPARTEE II).”

I recommend the authors to either consistently use shortened versions of the sites, or full names. Now there are three different ways to reference e.g. Marylebone Road (M. Road and MR). RESPONSE: We attempted to do this, but in such a long paper it is inevitable that some mistakes would be made. We have corrected as many errors in site-naming as we could find.

Some of the figures are already published in earlier studies, some even in the same special issue. Please indicate clearly earlier publication or if the figures are directly based on earlier publications, in the figure captions. RESPONSE: There are rather few figures taken from earlier papers, but those which are have been identified as such.

Many figures are almost impossible to read in this resolution. I will mention some later on, but please review the figures to make sure they are in readable resolution. RESPONSE: Many of the figures have been redrawn to improve readability.

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30156:10 Did the change in RP measurement site affect the measurements? RESPONSE: The same measurement site was used in REPARTEE I and REPARTEE II, the difference being in relation to the different platforms and sampling height for the particulate matter intake. The site is within a large park without nearby pollutant sources, and although small gradients will inevitably exist between 10 metres and 2 metres altitude, they are unlikely to create any major differences.

30159:4 How did the REPARTEE II arrangement changed from REPARTEE I? Does it matter? RESPONSE: As indicated above, some parameters were sampled at the same height in both campaigns but the 10 metre sample pipe used for particulate matter in REPARTEE I at Regents Park was not available in REPARTEE II. We believe that any consequent differences will be small, and since the vast majority of data analyses were within a campaign and did not combine data from the two campaigns, any consequences should be negligible.

30162:2 "REPARTEE special issue (link)" RESPONSE: A link has been inserted.

30162:8 The backscatter is mostly from the coarse particles. Do the smaller aerosol sizes probably have a similar mixing pattern? RESPONSE: It may be seen from Figure 15 that there were no substantial differences in the size distribution with altitude except in the nanoparticle region. The surface area distribution is probably that of most relevance to the lidar measurements and that varied very little with altitude as seen in Figure 15.

4.3 section: Is there major differences between weekdays? Do the measurements (roughly) agree with the size distribution flux measurements described later on the paper? Were the instruments intercalibrated? RESPONSE: Section 4.3 describes data collected with the MOUDI, SMPS and SMPS/APS. Significant differences are seen between weekdays and weekends in the traffic-generated pollutants which have been accounted for in our data analysis. The size distribution measurements are broadly consistent with the flux measurements described later on in the paper, but whereas all

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SMPS instruments were inter-calibrated in both campaigns, the optical instrument used in making size-resolved particle number flux measurements was not intercompared with the other sizing instruments as it works on a different physical principle and would not be expected to correlate in a consistent manner.

30171:24 CEH not explained (but can be cleared from the author list..) RESPONSE: This section is repetitive and has been edited.

Fig 15 (and 30170:15) I have really trouble seeing ANY accumulation mode in the number plots. Of course in the volume distribution such mode is visible, but this should be more clearly pointed out in the discussion of a "clear" accumulation mode in BT Tower. RESPONSE: The text does not say that the accumulation mode is visible in the number plots and it would be surprising if it were. The line number cited by the reviewer relates to Figure 16 (not Figure 15) in which the accumulation mode is very clear.

30170:21 Do the PMF methods agree with the cluster methods explained earlier, or are they not comparable? RESPONSE: The cluster methods referred to earlier were applied to SMPS data alone. Clustering is a technique for combining size distributions of similar form whereas PMF is a technique which breaks down a size distribution into its component parts. The two methods consequently achieve different objectives and do not therefore provide comparable results.

30172:5 Isn't the boundary layer mixing more dependent on temperature gradient, than on actual temperature levels? I can guess that the solar radiation was weaker in the colder period, was this the case? RESPONSE: The reviewer is of course correct that boundary layer mixing is more dependent on the temperature gradient than on actual temperature levels. We have made a slight adjustment to the text to avoid the implication that the temperature itself is the cause of the boundary layer mixing.

30172:28 How was the emission factor derived? RESPONSE: We have not added detail on this point in the interests of length, but have added a reference to the paper by Allan et al. (2010) in which the derivation of the emission factor is described.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

30173:15 Bonfire night? Was this explained in other analyzes? Could you then include it on the time-series plots. RESPONSE: Bonfire night was not a particularly prominent feature in the air pollution climate in 2007 compared to many years. It does, however, show up in the data, for example, in Figure 11.

30175:10 Are the EC fraction of the ATOFMS any way agreeing with the MAAP optical measurements on the sites? RESPONSE: Elemental carbon appears in three ATOFMS clusters (Ca-EC, Na-OC-EC and EC). These particle types are quantified in terms of particle number and we have not sought to estimate a mass of elemental carbon from the ATOFMS data which could then be compared with the MAAP measurements. 30175:14 I do not understand what a "coarse size distribution" means RESPONSE: The text actually refers to a relatively coarse size distribution which means that the distribution is shifted to larger sizes than those of other species. We did not feel that any modification is necessary.

30175 in general: Do the earlier mentions of the size distribution clustering agree with the LRT markers used in the ATOFMS and AMS? RESPONSE: The clustering of the size distributions combines distributions of a similar form into a much smaller number of average distributions referred to as clusters. Each cluster contains a wide range of chemical species from a range of sources and consequently these would not be expected to have the same size distribution as the individual particle types measured by the ATOFMS or the factors measured by the AMS.

30177 I would recommend trying to include the TEM analyzes somehow more integrated way in the big picture. Now they seem to be very separate from the other analyzes. Did they agree on the parts they could (understanding the limitations) with the other analyzes? RESPONSE: We had hoped that the TEM data would complement the overall data collected in the project but they provide a completely different view of the aerosol as we pointed out. Much as we would have liked to, it was very difficult to integrate the data any further than has been done. Additionally, due to limited sampling intervals and low overall particle numbers (hence poor statistics), it was not worthwhile

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to try to compare in detail those components measured by TEM and another technique.

30178:20 Please also indicate the potential LRT period in the figure 19 RESPONSE: The LRT period is now marked in Figure 19.

30179: 13 on: Are these stations different from the main stations used? If not, why explain them again? RESPONSE: This paragraph is indeed largely repetitious with the introductory sections and has been heavily edited.

30181 On discussing the NO<sub>2</sub> emission patterns, some discussion on how typical the vehicle fleet in comparison to other European levels could be useful, especially in context of congestion charges affecting different vehicles differently. RESPONSE: There is insufficient detailed information on the emissions of primary NO<sub>2</sub> from vehicles of different technology to add a comprehensive discussion here. Such a discussion is probably not warranted as the text in question is designed to set in context the measurements which are analysed subsequently in this section. Discussion in the context of other cities or changes in vehicle fleet composition is a major task and beyond the scope of this paper.

30183:17 I do not see hourly aggregated values in the top panel of Fig 24. RESPONSE: The hourly figures are on the second panel of Figure 24. The text has been amended to reflect this.

30184:7-8 The "Positive correlation" for O<sub>3</sub> is not so clear from fig 25. Is the p-value for the correlation different from random noise? RESPONSE: The legend to Figure 15 has been amended to show the correlation coefficient and number of points. It indicates that correlations are significant at the 99% level.

30184:16 what is reaction R<sub>2</sub>? RESPONSE: Reaction R<sub>2</sub> refers to reaction 2 on page 30183 of the ACPD version. This wording was inserted by ACPD editorial staff. This has now been amended to improve clarity.

4.9: I could not directly see the connection of the perfluorocarbon experiments to other

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

parts of the paper, this should directly connected to main conclusions of the paper.  
RESPONSE: The following text has been introduced at the beginning of Section 4.9 in order to provide a better linkage with the remainder of the study.

"The main strands of work in REPARTEE were concerned with the atmospheric chemistry, aerosol processes and fluxes of trace components in the atmosphere of London. A key factor determining the spatial distribution of pollutants within the London atmosphere is the dispersion from ground level sources and perfluorocarbon tracer experiments were conducted in order to provide additional information on this topic. These took advantage of the substantial sampling infrastructure provided by REPARTEE and in particular the availability of the BT Tower as a sampling platform."

An additional link has also been created in the conclusions section as follows:

"The work with perfluorocarbon tracers has provided new knowledge on the timescales of both horizontal and vertical transfer of pollutants within the urban atmosphere relevant to ground level releases."

30188:11 which roof top measurements? RESPONSE: The roof top measurements are presented in the citation provided at the end of the sentence.

30189 last line: Equation 1? RESPONSE: This sentence should have referred to Eq. (5) and this has been corrected in the revised manuscript.

30190:13 U? RESPONSE: U has been defined in the revised manuscript as the local wind mean horizontal wind speed.

30193:9 for which time-resolution and data point amount was the R2 calculated for?  
RESPONSE: A reference has been added to Table 8, the caption of which states that the values of R2 were calculated for hourly data. The number of paired samples differs between each two time-series and it would thus require an additional matrix to represent them fully, which we do not believe is warranted. We have therefore added the following text to the caption of the correlation matrix of the revised manuscript: "Each

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)



correlation coefficient was calculated on between 188 and 587 hourly data points."

30193:18 NAEI? RESPONSE: NAEI is the National Atmospheric Emissions Inventory. This is now spelt out in full. 30195:24-on Are the particle flux time-series similar as the ones measured down below concentration? I.e. can the flux differences seen directly from concentration differences near emissions, or do the micrometeorology affect the diurnal variation strongly? RESPONSE: The flux shows a much stronger diurnal pattern than the concentration time-series, because in the early morning hours the emission is close to zero, while the concentration is not (due to the contribution of background concentrations and nocturnal build-up). The exchange velocity ( $V_e$ ) describes the ratio of the flux to concentration. Thus, the pattern in  $V_e$  (e.g. Figure 34e and also Martin et al., 2009) reflects the difference in changes in concentration and flux with time.

30195: Ref to fig 31: Are the weekday and weekend spectra actually statistically different? The variation is smaller in weekends, but so is the data amount.. RESPONSE: There are, as yet, insufficient data to test robustly whether the weekend spectral behaviour is statistically different.

30199:10 I do not understand why such effect on  $DF/d\log D_p$ ? RESPONSE: We agree that the bimodal structure in  $dF_m/d\log D_p$  is somewhat surprising. Above Edinburgh, we found a continuous decrease in  $dF_m$  from 0.1 to 0.8  $\mu\text{m}$ , measured with a different optical particle counter, and then an increase for super-micron particles as measured with an aerodynamic particle counter. The particle range of the UHSAS used above London was clearly not geared to capturing the super-micron particles, thus we cannot fully capture this rise here, but might have seen a tail of the super-micron mode to creep into the largest submicron size ranges. This was not the case, either because of poor counting statistics or because resuspended material did not make it to the measurement height, but re-deposited beforehand. More intriguing is the fact that there was a dip in  $dF/d\log D_p$  around 200 nm or, conversely, a second peak at 400 nm, once measurements were converted to mass or surface area scale. There are several potential

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



explanations: (i) there are distinct physical or chemical processes that generate these two modes of particles e.g. the chemical production of accumulation mode particulate matter mentioned in the text; (ii) particles in the 200-400 nm range evaporate particularly efficiently before they reach the measurement height; or (iii) the UHSAS has a slightly higher sensitivity for particles > 400 nm than for the smaller particles. More measurements, ideally with collocated different instruments, are required to validate this behaviour.

## FIGURES

Overall, please check the figure legends and axes for better readability. I had to guess many of them due poor resolution. RESPONSE: These have been checked and improved where appropriate.

Scaling is a major problem in many figures, e.g. 3, 5(second panel), 12, 19, 20, 24. Also, these figures would greatly benefit from some sort of indication of a) weekends and b) periods of LRT. RESPONSE: The figures have been enhanced in the way recommended by the reviewer.

Fig 7. Units of vertical wind velocity show poorly. RESPONSE: We find the figure perfectly readable. There is no requirement to extract exact values.

Fig10 and other concentration figures: Please indicate weekends also on the figure time-axes. This is useful to see if the local emission patterns change significantly the measured concentrations. RESPONSE: Figure 10 and many others have been enhanced in the way recommended.

Figure 16 is too low resolution. The legend is almost impossible to read. I guess that the bottom part of the legend is "<105 m"? RESPONSE: The reviewer has interpreted the legend correctly. This has now been enhanced.

Figure 26: The figure is poorly explained in the text. What are the circled areas? RESPONSE: The circles are now clearly referred to in the revised manuscript and the

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

figure caption has been expanded.

Fig 29: What are the individual lines? RESPONSE: We have made the caption more explicit: "Figure 29: Normalised particle number flux as a function of flux averaging period for both REPARTEE experiments on the BT Tower (London Autumn 2006 and London Autumn 2007). The value of these Ogives at the averaging times of 15, 30 and 60 minutes (green, red and black line, respectively), reflects the fraction of the total flux that would be captured if these averaging times were used."

Fig 30: Colour scale is confusing: I recommend to change so that the hour of the day is cyclical (for example, night time dark, day time light) RESPONSE: We have already tried to make the time-scale somewhat cyclic, returning to black in the evening. However, we would like the colours for AM and PM to stay distinct and prefer the reader to spend some more time on absorbing the figure rather than losing the information on the difference in emission ratios in the early morning and late afternoon.

Fig 31: Y axes confusing RESPONSE: The outer labels for each pair of graphs refer to the particle number flux (weekends only) in units of particle number per cm squared per second. The inner label refers to the experimental period/experiment in each case. "L-AUT06" refers to the London Repartee experiment in Autumn 2006, whereas the "L-AUT 07" label refers to data from the London Repartee experiment in Autumn 2007. We believe this is quite clearly stated in the legend.

Figure 32. This interesting plot would need to be explained more in detail. However, the caption (last line) is confusing, and I am not sure from the text what can be understood from this. RESPONSE: In response to the comment for Referee #2, this Figure has been removed to Supplementary Information and the legend has been improved.

Figure 34d is repeated in the last panel (there are two same spectra). RESPONSE: This has been corrected, together with one of the units which was incorrect in the Discussions paper.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



The study gives an overview and conclusions of the REPARTEE experiment carried out in London, UK. The two measurement campaigns have been comprehensive and have brought new information about the behaviour of urban climate in a mid-latitude city. Particularly comprehensive turbulent flux measurements have been carried out above a city and for the first time ozone and carbon monoxide fluxes above a city have been reported. However, there are some issues that the authors need to address before the paper can be published in Atmospheric Chemistry and Physics.

First of all, the manuscript does not address the downsides of the experiments, which are the very short measurement periods only in autumn time in 2006 and 2007. How general conclusions can be drawn from such a short measurement period and how representative the results are for London itself and can the results be applied in other cities? RESPONSE: This point is very similar to that raised by Reviewer #1 and was addressed in response to his/her first comment. New text has been added.

The paper is an overview, but lacks to utilize the comprehensive dataset. For example, the authors have shown a case study for LIDAR data but no other variables were simultaneously analyzed with respect to the observed mixing and aerosol layers. Though, LIDAR data was successfully combined with the measured CO<sub>2</sub> flux. Also reasons to the observed differences between the campaigns were not properly studied. RESPONSE: It is not true that no other variables were simultaneously analysed with respect to the observed mixing and aerosol layers. Figure 16 presents precisely this kind of analysis and the work in the REPARTEE paper by Dall'Osto et al. (2011) uses the lidar data in interpreting the aerosol size distributions in a more detailed manner. In some of the data analyses (e.g. that in Dall'Osto et al., 2011) we have considered data from both campaigns. However, it should be pointed out that although it was not our plan, the available measurements in the two campaigns were rather different. For example, the lidar was available only in the second campaign and the MOUDI and Partisols only in the first. The PMF analysis of the AMS data is an example of where

Full Screen / Esc

Printer-friendly Version

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Discussion Paper



data from two campaigns were examined and significant differences resulting from the different climates over the campaigns were identified.

The manuscript is very long and there are too many figures (34). The length of the paper should be shortened, particularly Introduction is too long. The long historic overview on P30148 – 30149 between lines 21 – 26 could be shorter. Also there is too detailed information about the past PM10 measurements on lines 27 (30149) - 22 (30150). PM measurements are systematically measured in various cities so less focus could be given to them. RESPONSE: The two sections referred to by the reviewer have been shortened as suggested. Additionally, we have reduced the number of figures.

The rareness of flux measurements of aerosol particles and gaseous compounds were not addressed in introduction at all. RESPONSE: Some text has been added to the manuscript to review the existing information on flux measurements over urban areas.

Some results presented in the paper have been published in previous papers so text related to these could be shortened (e.g. in Sections 4.4 and 4.5). Also the number of figures should be reduced: some of them could be removed or moved to Appendixes. Many of the figures have poor quality and reading them was difficult (See detailed comments below). RESPONSE: We have carried out additional shortening as suggested and have reduced the number of figures. The quality of figures has been improved where possible.

Section 2 should be made more consistent. Now for some measurements lot of details are given under “Section 2.1 Sampling sites” while these would be better under instrumentation. Either the authors should give minimal information and refer to previous publications or give more information. RESPONSE: This has been resolved by increased reference to other publications in cases where the information is available from elsewhere.

For example, on P30155, L18 – P30156, L3 tube details and flow rates are given, but important information including: did all instruments use the same inlet, was there a

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



filter, what was the distance between the anemometer and the inlet, how long measurement tube was and what are particle losses in the tube, is missing. Authors mention that they made 2-dimensional rotation to fluxes but do not give other information about the flux analysis (wpl-correction, spectral corrections, etc.). It is also unclear that was same procedures used for all fluxes? RESPONSE: The reviewer appears to have overlooked some of the information on the flux measurements that was already provided in Section 2.1, including the fact that different inlet tubes were used for the aerosol and gas-phase flux measurements. The inlet length was 50 m. Given the long inlet line, temperature fluctuations will have dampened out by the time the instruments are reached, the part of the WPL correction that relates to water vapour fluctuations was applied to those instruments which do not already correct for this effect. Due to the slow turbulence associated with high measurement height, frequency losses are negligible. By contrast, low frequency losses may be as large as 15% (cf. Section 4.10.2). However, these are variable and difficult to predict and have not been corrected for. This detail has been added to the manuscript.

### SPECIFIC COMMENTS

In Section 3 there should be some information given what is the typical climatology of autumn period in London to get some idea how representative the measurement periods are. RESPONSE: This has now been included in the synthesis and conclusions section.

The text related to turbulent transport scales on P30190, L13-20 is unclear. Could the authors explain more in detail what do they mean by “The normalization had little effect on the pattern observed, indicating that  $z/U$  is not appropriate. . .”. What pattern do the authors mean and how does this indicate improper scaling? RESPONSE: The spectral pattern referred to is the clear diurnal sinusoidal variation in the peak of the power spectral density due to changing atmospheric stability.

There are many different spectral models of turbulence in the atmospheric boundary

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

and surface layers and without significantly extending the length of the manuscript we cannot justify a detailed discussion in this manuscript. The relevant scaling factors and functional analysis of turbulent quantities for urban environments is discussed in the cited reference, Roth (2000) and references therein, and in the discussion of figure 27. To shorten the manuscript as requested and to avoid confusion the sentence, “The normalization had little effect on the pattern observed, indicating that  $z/U$  is not appropriate. . “ will be removed as it is not germane to the discussion.

Also from Figure 28a, it is not clear that in stagnant conditions the peak maxima would shift to higher frequencies. To me it looks like opposite. RESPONSE: This was a typographical error. It should indeed have read “to lower frequencies” and has been amended. During stable nocturnal flow periods an increase in energy at low frequencies can occur if gravity wave activity is present, Hunt et al. (1985), Some observations of turbulence structure in stable layers, Hunt, J.C.R., et al. Quart. J. R. Met. SOC. (1985), 111, pp. 793-815. This spectral enhancement typically scales according to the local distance scale  $\bar{\sigma}_{sw}/N$ , where  $\bar{\sigma}_{sw}$  is the vertical velocity standard deviation and  $N$  is the Brunt-Vaisala frequency. These low frequency turbulent motions will contribute to transfer of both heat and contaminants from the surface layer but these do not appear common within the REPARTEE period described here.

P30192, L20 – 24: The authors say that a slight curvature is observed in midday and early morning hours in CO vs. CO<sub>2</sub> plot, but looking Figure 30 there seem to be only few data points “curving” in midday and in early morning the curvature is not clear at all. Is there any statistical proof for this reported behaviour? If not, the conclusions drawn out of this seem to be over interpretation. RESPONSE: There is some evidence of curvature throughout the dataset, which is somewhat masked by the large number of measurement points. We tried to make the data points smaller, but then their colour becomes difficult to interpret. Moving from a linear to a second order fit improves the R<sup>2</sup> slightly, but we agree that evidence is not very strong and have therefore down-tuned the statement.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

P30165, L1 – 5: It is concluded that during REPARTEE I there was less exceedences than during REPARTEE II but no discussions for the reasons are given. RESPONSE: The following text has been added in explanation: “The explanation for this difference lies with the street canyon nature of the Marylebone Road sampling site. This is located on the southern side of the canyon such that as a result of the vortex set up within the canyon, the on-road traffic emissions are sampled in addition to the local background when winds have a southerly component, but when winds are from the northerly sector, the sampling site is exposed predominantly to background air from outside of the canyon. During the REPARTEE I campaign, there was a much higher predominance of winds from the southerly sector than during REPARTEE II.”.

P30198, L25 – 29: This bimodal behaviour was observed on 26th October with low wind speed conditions. How representative the fluxes are for the surface in this case? In general, how often this bimodal pattern was observed as in Figure 34 we see only four days. RESPONSE: A full in-depth analysis is beyond the scope of this overview paper and may follow in a separate paper. A closer look at the overall dataset suggests that in general the accumulation mode flux does appear to be larger (relative to the Aitken mode) at lower wind speeds, which may support the hypothesis that it is due to SOA production below the measurement height, but as discussed also in the response to Reviewer 1 we cannot fully understand the behaviour with the measurements available.

P 30212, L5 – 9. Aerosol particle fluxes were not compared with emission inventories. RESPONSE: Correct, we could not make this particular comparison because there is currently no official UK emission inventory for particle number emissions.

#### SPECIFIC MINOR COMMENTS

P30145: In title atmosphere is mentioned two times. Maybe leave “atmospheric” out? RESPONSE: We considered this point but the title has been associated with the special issue of ACP for such a long time, we do not feel that change is now possible.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)



P30155, L12: In what coordinate system the coordinates are given. Why there is two times BT Tower? RESPONSE: The coordinates are given as degrees, minutes and seconds of latitude and longitude. The title BT Tower is cited twice to show the full name and the abbreviation used in the text (see also Regents Park (R. Park, etc.)).

Section 2.3: There is no need for this section. These dates could be given under Section 2 after the first sentence. RESPONSE: This section has been deleted and the information transferred as suggested.

Section 2.4: Are the details related to tracer experiments explained in Martin et al. 2011a? If yes, text in this Section should be shortened. RESPONSE: It is appreciated that there is a rather excessive level of detail here. Unfortunately, this is not described adequately in Martin et al. (2011a) but has nonetheless been shortened.

P30159, L14: Are the limits standard deviations or something else? RESPONSE: Yes, these are standard deviations of daily averages and the point has been clarified.

P30161, L23 – 24: Urban heat island is not only caused by this delayed cooling. It is also observed in daytime but is just typically stronger in night-time. Anthropogenic heat emissions play an important role. RESPONSE: The reviewer is correct and this point has now been acknowledged.

P30163, L16 – 21: Are these values calculated from the daily data or directly from high resolution data? Also is it necessary to list minima and maxima here as they are also visible from Figure 10. Or have them in a table rather than scattered in the text. RESPONSE: These values are calculated from daily data. We do not feel that it is possible to read the maxima and minima easily from Figure 10 and are reluctant to include another table. They take up very little space in the text and are easily available there to the reader.

P30170, L5: Looking Figure 15 the peak at the Park seems to be also in the nucleation mode rather than accumulation mode despite the shift of the mode. RESPONSE:

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

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Our text does not state that the peak at the park is in the accumulation mode. Rather, it states that the spectrum shows an increase in the accumulation mode relative to the nucleation mode which is clear in the number size distribution and far more obvious when taking into account the area size distribution (taking account of the different scales in the figures).

P30171, L16 – 28. This text would be more suitable under Methods. RESPONSE: This section has been shortened with additional reference to the methods section. P30177, L20 – 23. This text could be removed to Methods. RESPONSE: The majority of this information is already in the methods section and a cross reference has been included.

P30178, L1 – 3. I think there is some over interpretation of the differences between the Park and NK as in error limits the concentrations are nearly equal. RESPONSE: The error bars represent standard deviations of hourly measurements and not measurement errors. If standard errors are considered, the ranges do not overlap and the means are seen to be significantly different. The text has been amended to clarify the point.

P30182, L19 - P30183, L14. This text would be more suitable in methods section. RESPONSE: We feel that this information is specific to this section and probably best retained here rather than referring the reader back to the methods sections.

P30184, L6 – 8. What were the correlation equations and statistics (rmse, r) and were the correlations significant. Looking only Figure 25 the stated correlations are not clear. RESPONSE: The correlation statistics are now included in the legend to Figure 25.

P30189, L13: What does “\_ < ws” stand for? RESPONSE: This is a typesetting error, which we will check for in the final version. It should read “<0”.

P20190, L5 – 6: Why weaker correlation for particles and CO<sub>2</sub> are expected? RESPONSE: Depending on the efficiencies of the multiple individual source processes contributing to CO<sub>2</sub> and particle emissions the two may or may not be correlated per-

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fectly. Theoretically the correlation will also depend on the inertial linkage between aerosol mass and fluid shear forces and the relative importance of the different diffusion processes as we move from the molecular to condensed phase regime. In this case the poorer correlation is likely a combination of all of these.

P30194, L11 – 14. This information is not relevant here and should be given in the Introduction. RESPONSE: This quantitative statement is relative to a number of sections of the report including this one. It has now been relocated to the introduction.

Related to the text in P30196, L14 – 20, how wide the footprint of the flux measurements typically is? RESPONSE: The horizontal angle of the footprint plume will depend on the sampling height, wind and in particular the atmospheric stability. In the case for REPARTÉE the flux footprint width, in terms of the plume length to width angle will range from approximately 18 to 30 degrees for typical neutral to unstable conditions encountered here.

P30199, L5 – 8: From figure 34 it is not clear that the times when deposition fluxes were observed, the air was more polluted than on other times. RESPONSE: We agree and have rephrased this sentence to put the emphasis on the fact that the northerly wind sector is less densely urban, now also referring to the CO<sub>2</sub> flux of Helfter et al. (2010), which was smaller for N than for SW wind directions.

P30220, L16: Helfter et al. has already been published in ACP so maybe use rather that reference RESPONSE: The reference has been updated in the revised manuscript. P30205, L9 – 10 and L21 – 22: Do these sentences refer to the same “remarkable change”? RESPONSE: These statements referred predominantly to different parts of the size distribution and both statements have been edited to clarify the situation.

## TABLES AND FIGURES

Table 1: Used abbreviations should be explained (e.g. KCL). Turbulence and heat flux have a temporal resolution of 10 Hz here while in the text is said to be 20 Hz

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(P30155, L25). Why the measurement uncertainties are left out in the case of turbulent fluxes? These can be calculated (both systematic and unsystematic) RESPONSE: The abbreviations have now been explained as a footnote. The anemometer was recorded at 20 Hz (which has been corrected), while the gas analysers were recorded at 10 Hz or lower as indicated. The uncertainty of a turbulent flux measurement depends on the atmospheric conditions and pollutant concentration and therefore varies with time. Thus, to include a blanket value of the uncertainty is therefore not appropriate and would be highly misleading.

Figure 1: The area marked in the larger map is not clear. Using another colour for the box would help. RESPONSE: We have taken up this helpful suggestion.

Figure 2. Is this figure necessary? RESPONSE: We feel that this figure is valuable for two reasons. Firstly, the BT Tower is a major London landmark and many readers of the article will be familiar with it and will be interested to know where the sampling instruments were sited. The other reason is that we wish to demonstrate the good exposure of the instruments which should be made clear by the figure.

Figure 4: This figure is not giving any extra information and should be left out. RESPONSE: We have moved Figure 4 to Supplementary Information.

Figure 3 and 5: The quality of these figures is poor. The x-axis labels should be improved. Also the labels should explain the used abbreviations and what are the plotted resolutions (10-min, 30-min etc.). RESPONSE: The quality of these figures has been improved and the legend now includes information on the temporal resolution.

Figure 7: x-axis label is missing from the lowest panel RESPONSE: The label has been added.

Figures 10, 11: What are the black vertical lines in the figures. Labels should be with bigger font. RESPONSE: Both points have now been addressed in revising the figures and legend.

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Figures 12, 13: Are poor quality. Labels are not clear and reading the figures is difficult. RESPONSE: These diagrams summarise a great deal of information very concisely. We have attempted to improve the labelling.

Figure 14: Could be removed. The strong morning peak between 6 and 10 can be described in the text only. RESPONSE: We have moved this figure to Supplementary Information.

Figure 17: The different x-axis is subplots should be emphasized in the figure label. RESPONSE: The figure legend has been modified accordingly.

Figure 18: This was published already in Allan et al. (2010) paper. Is this necessary here as there are quite many figures anyway? If yes, please, add the reference to the figure label. RESPONSE: We feel that this is key information and have retained it with a reference to Allan et al. (2010). Figure 26: What are the black circles in the figure? RESPONSE: This has been addressed in response to Reviewer 1.

Figure 27: The quality of this figure needs to be improved: different text's are not readable. RESPONSE: This figure should be more readable once it is set in portrait format of Atmos. Chem. Phys. We will be assessing this at the proof stage.

Figure 29. Could be removed (or added to appendixes if needed) and explained in the text. RESPONSE: This has been removed to the Supplementary Information.

Figure 32. Again, is this necessary figure in the main text? RESPONSE: This has been removed to the Supplementary Information.

Figure 34. The figure needs to be bigger. In its current form it is not possible to see in size A4. RESPONSE: Again, this figure should be more readable once it is set in portrait format of Atmos. Chem. Phys. We will be assessing this at the proof stage.

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 30145, 2011.

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