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> Interactive Comment

Interactive comment on "Seasonal trends in concentrations and fluxes of volatile organic compounds above central London" *by* A. C. Valach et al.

A. C. Valach et al.

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The authors would like to thank the reviewers for their astute comments and suggestions, which have helped in improving the revised manuscript. Responses to the reviewer's comments are below with reviewers' comments followed by a response to each point with the respective revisions to the manuscript in "quotations" unless the changes included the reworking of whole sections. As well as in response to the reviewers' comments, changes have been made throughout to improve the clarity and readability. The manuscript and figures should now be much easier to read and follow. Furthermore, acronyms and formatting have been checked and are now clear and





consistent throughout. Section 3.2.2 has also been expanded.

Review 2

Major comment 1: The suitability of the King College site for monitoring turbulent fluxes needs further assessment. The land cover is very heterogeneous and the buildings morphology does not seem to contribute for measuring fluxes by eddy covariance. The street canyon formed by the own building where the measurements were conducted may enhance the accumulation of freshly emitted VOCs below the urban canopy, particularly during periods of stable atmospheric conditions at night and winter.

Response 1: The KCL site at King's College has been used as a long term CO2/H2O flux measurement site and thus site characteristics and suitability for micrometeorological flux measurements have been investigated and described extensively by Kotthaus and Grimmond (2012; 2014a; 2014b). Although the site is not ideal for flux measurements, the cited studies show that representative surface-atmosphere fluxes can be measured at the site without significant bias from the local morphology as long as data are filtered to remove contributions from local micro-sources, e.g. vents and windows, if present. Additionally, they show that results from flux footprint models at the site can provide reasonable information. Some considerations were highlighted such as instrument siting in complex urban areas requiring careful interpretation of measured and modelled data. The relatively low measurement height allows close coupling to the street canyon. The analyses presented here were based on averaged data to reduce some of the uncertainties and used to describe overarching trends. These trends agree with conclusions from previous urban VOC flux studies. This study is the first of its kind to present long term continuous VOC flux measurements by PTR-MS over an urban area and hence provides valuable information on VOC fluxes from central London, despite some minor limitations imposed by the less-than-ideal site morphology. Section 2.1 was expanded to include: "The sampling point (which we call KCL) is located 37 m west of a sampling point (KSS) that has been used for long-term energy and CO2 flux measurements (Kotthaus and Grimmond, 2012). Although the site is

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not optimal for micrometeorological flux measurements due to the heterogeneity of the urban canopy, its suitability has been assessed in detail by Kotthaus and Grimmond (2014a; 2014b). This study describes in detail the measurement area and investigates the influence of source area characteristics on long-term radiation and turbulent heat fluxes for the KSS site. They conclude that the site can yield realistic data on surface to atmosphere fluxes."

Kotthaus, S., and Grimmond, C.S.B.: Identification of Micro-scale Anthropogenic CO2, heat and moisture sources – Processing eddy covariance fluxes for a dense urban environment, Atmospheric Environment, 57, 301-316, http://dx.doi.org/10.1016/j.atmosenv.2012.04.024, 2012. Kotthaus, S., and Grimmond, C.S.B.: Energy exchange in a dense urban environment – Part I: Temporal variability of long-term observations in central London, Urban Climate, 10, 2, 261-280, http://dx.doi.org/10.1016/j.uclim.2013.10.002, 2014a. Kotthaus, S., and Grimmond, C.S.B.: Energy exchange in a dense urban environment – Part II: Temporal variability of spatial heterogeneity of the surface, Urban Climate, 10, 2, 281-307, http://dx.doi.org/10.1016/j.uclim.2013.10.001, 2014b.

Major comment 2: For eddy covariance flux measurements samples are usually collected at 10 Hz (15,000 samples in a period of 25 min). The sampling rate when using the disjunct eddy covariance method is slower. However, a sampling rate of 5.5 sec (273 samples in a period of 25 min) as that used here seems to be extremely slow. The statistical uncertainty of the fluxes caused by a longer time resolution needs to be evaluated. The CO2 flux data discussed in section 3.2.2 may help to assess this issue.

Response 2: The disjunct sampling increases the random error of the flux but, provided the sampling intervals are less than the integral timescale, this should not introduce a systematic bias. We tested this assumption by simulating disjunct sampling on sensible heat flux data, which were calculated from the continuous data and then compared with the sensible heat fluxes calculated from a disjunct series with a sampling rate of 2 Hz and a sampling interval of 5.5 s. The overall difference between the EC and

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DEC sensible heat fluxes over the entire measurement period was minimal (0.01 %), therefore no additional corrections have been made to the VOC fluxes. This information was added to Section 2.3: "The error due to the disjunct sampling was estimated by comparing the sensible heat fluxes calculated from the continuous data series with those calculated from a disjunct data series using a set sampling interval of 5.5 s. The continuous data were averaged to match the sampling frequency of the disjunct data (i.e. 2 Hz). The difference between the eddy covariance and DEC sensible heat fluxes was minimal (0.01 %) and thus no additional corrections were applied."

Specific comments:

Comment 1: P6602, L17. G95 algorithm?

Response 1: p. 6602 l. 17: expanded to "Guenther et al., (1995)".

Comment 2: P6603, L12. ...use a "bottom-up" approach based on activity data and emission factors....

Response 2: p. 6603 l. 12: changed to "... "bottom-up" approach based on activity data and emission factors".

Comment 3: P6603, L22. This reviewer has serious concerns on the methodology used by Park et al., 2010 & 2011.

Response 3: p. 6603, l. 22: There are very few urban VOC flux measurements and even fewer that use DEC based techniques. Therefore to expand the comparison of VOC fluxes in this study with those in the literature, additional studies using relaxed eddy accumulation (REA) were consulted. REA can be associated with large errors due to the loss of reactive compounds on canister surfaces, but also the potential error from a bias in the vertical wind velocity, as shown by the normalised bias (w $l\check{E}/\sigma_w$). A sentence has been added for further clarification: "Unlike the other studies cited, Park et al. (2010) use relaxed eddy accumulation to measure VOC fluxes and hence the data obtained are not directly comparable with measurements made by EC-based

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methods."

Comment 4: P6603, L29. Define PTR-MS.

Response 4: p. 6603 l. 29: expanded to "proton transfer reaction-mass spectrometry".

Comment 5: P6604, L11. Check symbols of seconds, minutes, inches, etc. throughout the text.

Response 5: p. 6604 l. 11: changed to decimal degrees: "51.511667 N 0.116667 W".

Comment 6: P6604, L16. Update classification based on Stewart & Oke (2012).

Response 6: p. 6604 I. 16: Updated site classification, added the reference and changed text to: "This site is classified as Local Climate Zone (LCZ) Class 2 Compact Midrise according to Stewart and Oke (2012) (i.e. dense mix of midrise buildings (3–9 stories), few or no trees, land cover mostly paved, stone, brick, tile, and concrete construction materials."

Comment 7: P6604, Section. 2.1. Add fractions of the plan area cover (i.e., building, roads, vegetation, water bodies, etc.).

Response 7: p. 6604, l. 18: Added: "Land cover types (in %) were calculated based on the Ordinance Survey map for the 9 km2 area (Figure 1) encompassing the site and are: roads (37 %), buildings (31 %), other paved areas (14 %), unpaved/ vegetation (11 %), and water bodies (7 %)."

Comment 8: P6605, L13. Although in following paragraphs the averaging process is described, in a few words mention why periods of 25 min were used instead of periods of 30 min. Periods of 30 min are usually used when measuring fluxes over urban surfaces.

Response 8: As mentioned in Section 2.2 p. 6606 l. 9-11, the hourly duty cycle of the PTR-MS consisted of 5 min zero air measurements, followed by 25 min MID used to calculated fluxes, then a further 5 min mass scan and finally another 25 min MID mode.

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We present 25 min fluxes as we have no measurements for the initial 5 min of each 30 min period.

Comment 9: P6605, L16-18. Note that emissions in cities respond strongly to human activities, and the behaviour of these follows the local time and not the UTC.

Response 9: Section 2.2, p. 6605, l. 16-18: The time axes of figures all state that local time was used, which has also been added in the text as: "However, all analyses used local time."

Comment 10: P6606, L-15-20. Why were data of m/z 33 and m/z 121 not included?

Response 10: m/z 33 is included, however m/z 42 and 121 were not included as the signal was too low and during the measurements their places in the limited duty cycle were subsequently used to investigate other less typically measured masses, none of which showed any useful information.

Comment 11: P6607, L2. Check that all variables are written with italic fonts.

Response 11: p. 6607 l. 2: variables have been checked and are now written with italic font throughout.

Comment 12: P6607, Eq. 1. Fix the fluctuations' symbols.

Response 12: p. 6607 Eq. 1: fluctuation symbols have been checked and are now correct and consistent throughout.

Comment 13: P6609, Eq. 1. This equation is unreadable.

Response 13: p. 6609, Eq. 1: This equation is clearly presented. It may be that the reviewer is using a non-compatible PDF-viewer. We will check this at the proof-reading stage.

Comment 14: P6609, L20. Ergodicity is a rare/exotic term to indicate that the buildings height and morphology in the monitored district were quite variable. From Wikipedia:

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"The term ergodic is used to describe a dynamical system which has the same behaviour averaged over time as averaged over the space of all the system's states. In physics the term is used to imply that a system satisfies the ergodic hypothesis of thermodynamics."

Response 14: p. 6609, l. 20-22: The sentence has been changed to: "The high number of files rejected in the stationarity test is to be expected for eddy covariance measurements over highly heterogeneous canopies, although horizontally averaged canopy morphology recovers some surface homogeneity."

Comment 15: P6610, L7. Do not begin sentences with numbers or acronyms.

Response 15: p. 6610, l. 7: added "Exactly".

Comment 16: P6612, L5-10. If daily mean fluxes are presented, it would be better to use units of kg km-2 day-1.

Response 16: p. 6610, l. 3-12: The figures 2a and b show the diurnal profiles of VOC fluxes and mixing ratios, whereas the values represent hourly average fluxes, which are typically given in units of mg m-2 h-1 (Karl et al., 2004; 2007; 2009; Langford et al., 2009; 2010a; 2010b; Misztal et al., 2011; Rinne et al., 2001; 2002). Both fluxes and concentrations include data from the entire measurement period. For clarification purposes the words "average diurnal cycles" are now used throughout to refer to diurnal profiles of VOC emissions and mixing ratios. The units mg m-2 h-1 are used in table 2 to show the overall average of diurnal profiles, which are in hours of the day. However values are now cited in the main text body in Section 3.1 in units of kg km-2 d-1 as suggested by the reviewer, as follows: "Largest median (interquartile range in parenthesis) fluxes per day were from C2-benzenes and toluene with 7.86 (0.92-21.8) kg km-2 d-1 and 7.26 (1.83-15.3) kg km-2 d-1 respectively, followed by oxygenated compounds, i.e. methanol with 6.37 (2.99-10.0) kg km-2 d-1. Isoprene and benzene showed smallest median fluxes with 2.14 (0.56-4.85) kg km-2 d-1 and 1.78 (0.06-4.34) **ACPD** 15, C3916–C3939, 2015

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kg km-2 d-1 respectively."

Comment 17: P6612, L14-15. ... lifetimes and widespread origin including anthropogenic and biogenic sources and photochemistry ...

Response 17: p. 6612 l. 14/15: changed to "lifetimes and widespread origin including anthropogenic and biogenic sources and photochemistry".

Comment 18: P6613, L5-7. It may only be true for London and other UK cities.

Response 18: p. 6613, I 5-7: changed to "central urban areas in UK cities".

Comment 19: P6614, L23-24. Explain how advected air masses rich in methanol and acetone might affect the local boundary layer meteorology.

Response 19: p. 6614, l. 23-24: This was poorly worded and has been changed to: "De Gouw et al. (2005) reported that changes in boundary layer meteorology could result in greater effects on observed concentrations of methanol and acetone due to their high background values. The mixing ratios of these compounds are, therefore, dominated by advected pollution rather than the local flux."

Comment 20: P6615, L11-12. Is there an important potential emission source (e.g. petrol station) at the west of the flux tower?

Response 20: p. 6615, l. 11-12: As the Congestion Charge Zone in London limits the number of private vehicles in this area, there are only few petrol stations in the vicinity. The nearest is 1 km to the north and the closest westerly station is 2.5 km away. These petrol stations are outside of the 90 % flux contribution distance and are not likely to have contributed to fluxes. No other significant point sources of this type were identified within the footprint area.

Comment 21: P6617, L3-6. Was turfgrass considered?

Response 21: p. 6617, l. 3-6: added "total tree leaf area". Turfgrass was not considered as turf grass species used in the UK do not emit isoprene or emit it at undetectable

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levels (e.g. Stewart et al., 2003).

Comment 22: P6617, L21. ... for fluxes indicated (What?) ...

Response 22: p. 6617 l. 21-22: This section was poorly phrased and has been changed to improve clarification and consistency: "Correlations of VOC/VOC fluxes (R2 = 0.40-0.62, p<0.001) indicated two groups of compounds with good correlations within each group, i.e. compounds related to traffic sources such as aromatics, and oxygenated and biogenic compounds, such as methanol, acetone and isoprene. Correlations of VOC/VOC concentrations (R2 = 0.13-0.84, p<0.001) showed highest correlations between traffic-related compounds (R2 = 0.45-0.84, p<0.001) and good correlations between the oxygenated and biogenic compounds (R2 = 0.55-0.69, p<0.001) (Figure 6)."

Comment 23: P6617, L22. Provide examples of such species.

Response 23: p. 6617, l. 22: See part of previous response: "i.e. compounds related to traffic sources such as aromatics, and oxygenated and biogenic compounds, such as methanol, acetone and isoprene."

Comment 24: P6617, L21-25. This paragraph is difficult to read.

Response 24: p. 6617, l. 21-25: See responses 22 and 23.

Comment 25: P6618, Section 3.2.1. A figure showing scatter plots of benzene versus toluene would be helpful.

Response 25: p. 6618, Section 3.2.1: Phrasing has been improved and two panels have been added to figure 7 showing scatterplots of benzene versus toluene concentrations for the 9th and 12th August respectively. The figure caption now reads: "Bottom: Scatterplots showing benzene to toluene concentration ratios during the 9th August 2012 (left) and 12th August 2012 (right) with linear regression with 95th confidence interval, regression equation and coefficient (R2)."

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Comment 26: P6618, L10. IQR?

Response 26: p. 6618, I.10: added "and interquartile range".

Comment 27: P6618, L14-24. Zavala et al. (2006), Rogers et al. (2006), Velasco et al. (2007) and Karl et al. (2009) reported benzene to toluene ratios for Mexico City. The vehicular fleet and industry from both cities are expected to be considerably different, as well as the benzene to toluene ratio.

Response 27: p. 6618, l. 14-24: The detailed comparison with Mexico City has been removed and only the b/t flux ratios from Karl et al. (2009) have been included to help explain a possible reason for the low observed flux ratios in this study. The comparison with other cities now focuses on European and UK cities. The section has also been rephrased to improve the clarity as follows: "The observed ratios compared well with those of other European cities, which showed b/t concentration ratios of 0.35 in Zurich (Heeb et al., 2000), 0.57 in Manchester (Langford et al., 2009), 0.57-0.63 in London (Valach et al., 2014), and 0.1 at 190 m above London (Langford et al., 2010b). Traffic related emissions are considered to be an important source of benzene and toluene in London. B/t exhaust emission ratios based on derived yearly emissions in other megacities, such as Mexico City, were found to be 0.4 (Zavala et al., 2006), which agreed well with observed b/t concentration ratios in this study. Airborne flux measurements over Mexico City have shown average b/t flux ratios of 0.31 with lower ratios of 0.07 to 0.1 over industrial areas due to increased toluene emissions from industrial processes (Karl et al., 2009; Velasco et al., 2007). Evaporative emissions from gasoline or direct industrial toluene emissions may have contributed to the lower b/t flux ratios in London. Furthermore, low b/t concentration ratios of 0.26 from diesel emissions have been reported (Corrêa and Arbilla, 2006). The widespread use of diesel fuel in London (buses, taxis and some cars and trains) and diesel emissions from roads which exclude passenger cars, such as Oxford Street (approx. 1.3 km W from the measurement site) or central railway nodes, such as Waterloo Railway Station (1 km to the S), may have affected b/t ratios."

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Comment 28: P6621, L20-22. If this was true, the reported fluxes would not be representative of the monitored district. The measurement height together with the data quality assurance suggests that the flux measurements were properly conducted at the inertial sublayer, where the turbulence and fluxes are relatively homogenous.

Response 28: p. 6621, l. 20-22: The section was poorly worded and has been rewritten as: "Due to the relatively low measurement height, flux measurements were always closely coupled with the surface layer, unlike measurements by Langford et al. (2010b), which were at times disconnected from the surface layer during stable night time conditions. The flux footprint in this study was relatively small compared with that of measurements previously made a 190 m height from the BT Tower in central London (Langford et al., 2010b)."

Comment 29: P6622, L7-27. This discussion is long and difficult to follow.

Response 29: p. 6622, l. 7-27: This section was poorly phrased. The structure and language have been improved. "Green areas, as defined on the OS map, comprised 9 % of the total grid area and were evenly distributed across the 9 km2. Only grid square 1 included a large green area of 23 ha (St. James' Park). The National Forest Inventory England only included 4.4 % green areas within the grid selection (NFI, 2012)." And: "The River Thames to the S may have caused the low fluxes associated with S winds (i.e. squares 1, 2 and 3). Contributions of traffic related compound fluxes were statistically significant from the W (i.e. squares 4, 5, and 7), followed by the N (square 8) and E (squares 6 and 9) likely from the nearby heavily trafficked roads (Kingsway, Charing Cross, Strand and Blackfriars areas, respectively). Biogenic compound fluxes were highest from the W and E which coincides with significant nearby green areas within the flux footprint."

Comment 30: P6623, L11. The London Atmospheric Emissions Inventory (LAEI) and the Atmospheric Emissions Inventories (NAEI) ...

Response 30: p. 6623 I. 11: changed to "London Atmospheric Emissions Inventory

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(LAEI) and National Atmospheric Emissions Inventories (NAEI)".

Comment 31: P6623, Section 3.4. Do NAEI and LAEI provide data on the spatial and temporal distribution of the estimated emissions?

Response 31: p. 6623, Section 3.4: Estimates are produced on an annual basis and over a 1 km2 grid system which coincides with the Ordinance Survey grid. This information was added to the first sentence: "The London Atmospheric Emissions Inventory (LAEI) and National Atmospheric Emissions Inventories (NAEI) produce yearly emission estimates over the 1 km2 OS grid for a range of pollutants and emission sources."

Comment 32: P6624, L6. SNAP?

Response 32: p. 6624 I. 6: added "(Selected Nomenclature for sources of Air Pollution)".

Comment 33: P6625, L15-17. The article does not discuss the suitability of the King College for turbulent flux measurements. If its suitability has been previously analysed, include the corresponding references.

Response 33: p. 6625, l. 15-17: This issue is addressed in the response to Major comment 1 by this reviewer and references have been added.

Comment 34: P6636, Fig. 1. The green marker is difficult to find. Make it larger.

Response 34: p. 6636, Fig. 1: The size of the green marker has been increased and a label of the site name added.

Comment 35: P6636, Fig. 1. It would be helpful to see the estimated footprint overlaid on the map.

Response 35: p. 6636, Fig. 1: Outlines for Xmax, 75 %, 90 %, and 99 % of the area contributing to the flux footprint are shown with respective labels. Changes in the figure caption have been made accordingly: "Map of central London overlaid with the Ordinance Survey grid including the measurement site (KCL) at King's College

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(green point) with references to the geography of Greater London and Great Britain. Outlines of the areas that contribute the maximum (Xmax), 75%, 90%, and 99% to the flux footprint using overall median meteorological values are shown in black with their respective labels laid out according to the median wind direction."

Comment 36: P6637, Fig. 2. There is no need of mixing weekdays and weekend's fluxes in one profile. For some species, such as C2-benzenes and toluene, the difference is considerable. Show only the variability (i.e. confidence interval) of weekdays or weekends.

Response 36: p. 6637, Fig. 2: The format used in figure 2 has previously been widely used to show diurnal profiles of pollutant concentrations and fluxes, including by Bigi and Harrison (2010), Langford et al. (2010b), Park et al. (2010), Park et al. (2011), Velasco et al. (2005), Velasco et al. (2009). However, to aid clarity weekend and weekday lines are now in colour (blue and red respectively) to improve the readability and the figure caption has been updated: "weekdays (red dashed line) and weekends (blue dotted line)".

Comment 37: P6637, Fig. 2. For panels in section (b) select scales that help to visualize the diurnal characteristics. For example, the scale for benzene should go from 0.20 to 0.40 ppb, instead from 0.00 to 0.45 ppb.

Response 37: p. 6637, Fig. 2: It is conventionally accepted that it is good practice to plot figures with both axes beginning at zero. We retain this format in order to aid comparison with previously published data on VOC mixing ratios, e.g. Bon et al. (2011), Davison et al. (2009), Fraser et al. (1998), Heeb et al. (2000), Karl et al. (2007), Kato et al. (2004), Kim et al. (2001), Langford et al. (2010a), Liu et al. (2015), Misztal et al. (2011), Park et al. (2010), Park et al. (2011), Velasco et al. (2007), von Schneidemesser et al. (2011), Wang et al. (2014), Warneke et al. (2014). The figure caption was updated to include: "The mixing ratio axes start from zero apart from that of methanol, which begins at 6.4 ppb due to the high atmospheric background."

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Comment 38: P6638, Fig. 3. Check the linear regression of panel (E).

Response 38: p. 6638, Fig. 3: The regression line in panel E has been corrected.

Comment 39: P6639, Fig. 4. What do the bar charts represent? Do they show the mean daily flux/mixing ratio for each monitored month?

Response 39: p. 6639, Fig. 4: Bar charts show hourly fluxes averaged over each month in mg m-2 h-1. The figure caption has been expanded with: "Diurnal profiles by month with confidence intervals and bar charts showing hourly averages for the respective month and representative compound (top) fluxes (mg m-2 h-1) (m/z 45, 69 and 79) and (bottom) mixing ratios (ppb) (m/z 59, 69 and 79)."

Comment 40: P6640, Fig. 5. Too many dashed lines in the scatter plots. They are confusing.

Response 40: p. 6640, Fig. 5b: 1:2 and 2:1 lines have been removed.

Comment 41: P6640, Fig. 5. OLS?

Response 41: p. 6640 Fig. 5: caption changed to "Ordinary Least Squares (OLS)".

Comment 42: P6640, Fig. 5. Describe first the panels at the left and then the panels at the right.

Response 42: p. 6640, Fig. 5: The caption has been adjusted: "Figure 5a. Time series of both measured (grey) and modelled (black) fluxes, as well as PAR and temperature measurements for August and September 2012. Figure 5b. Correlation between modelled and measured isoprene fluxes (mg m-2 h-1) by wind direction using the G95 algorithm with temperature as a third variable, Ordinary Least Squares (OLS) regression lines, 99th confidence intervals, formulae, and R2-value."

Comment 43: P6641, Fig. 6. Scatter plots between fluxes would be more interesting.

Response 43: p. 6641, Fig. 6: Scatterplots between the fluxes show no clear trends, as

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many fluxes were below the LoD and therefore individual flux points have high uncertainties, hence averaged data have been used throughout in the analyses. However, flux correlations between the same compound pairs as used in the concentration correlations have been added and the figure caption updated: "Figure 6. Selected scatter plots of representative correlations of VOC/VOC fluxes (top) and mixing ratio (bottom) with temperature as a third variable showing an example of bimodal, strong linear and medium linear correlations as commonly seen in the mixing ratio correlations with R2values, 1:1 line, 1:2 and 2:1 lines for the bimodal example in the bottom left panel."

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Fig. 1. Map of measurement site in central London

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Fig. 2. Average diurnal profiles in local time for selected VOC fluxes



Fig. 3. Average diurnal profiles in local time for selected VOC mixing ratios

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Fig. 4. Examples, using isoprene, of averaged VOC fluxes (left) and mixing ratios (right) as a function of PAR, temperature, traffic density and boundary layer mixing height

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Fig. 5. Diurnal profiles by month with confidence intervals and bar charts showing hourly averages

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Fig. 6. Time series of both measured (grey) and modelled (black) fluxes, as well as PAR and

temperature





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Fig. 9. Top: 24h back trajectories from the NOAA HYSPLIT trajectory model. Bottom: Scatterplots showing benzene to toluene concentration ratios



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