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Atmospheric Chemistry and Physics Discussions

**ACPD**

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Interactive Comment

# *Interactive comment on* "Mixing ratios and eddy covariance flux measurements of volatileorganic compounds from an urban canopy (Manchester, UK)" *by* B. Langford et al.

**B. Langford et al.**

Received and published: 11 February 2009

Reviewer 2

RC: lack of detail on PTR-MS and misinterpretation of what the "transmission curve" is and means;

AR: We fully agree that the description of the PTR-MS setup was insufficient and have expanded this to include specific details of the instrument used, typical primary, reagent and target ion counts as well as measurement sensitivities. We also thank the reviewer for pointing out our misinterpretation of transmission which has been altered accordingly. Section 2.2 line 5 has been changed to "The PTR-MS used was a standard model, containing two turbo molecular pumps, a 9.6 cm long drift tube (stainless steel rings) and heated silico steel inlet which drew air at a rate of 0.15 l min-1" and section 2.2, paragraph 3 now states "During the measurement period drift tube parameters such as pressue, temperature and voltage were held constant at 2 mbar, 45 oC and 600 V respectively to give an E/N ratio of approximately 125 Td. The primary ion count ranged between 2.2 and 3.6 E6 with an average of 2.8 E6. The reagent ions ranged between 1.07 and 2.62 E5 with a mean of 1.69 E5 which represented 6 % of the primary ion signal. Average normalised counts ranged between 3 (benzene) and 72 (methanol) with instrument sensitivities in the range of 4.3 (isoprene) and 13.26 (benzene) ncps ppbv-1." Finally section 2.2, paragraph 4 was changed to "A VOC gas standard was not available for on site calibration of the PTR-MS, hence mixing ratios were calculated using the instrument specific transmission coefficients and reaction times taken from Zhao and Zhang (2004)."

RC: very short dwell time (20 ms) in the vDEC method and resulting potentially (very) high noise level. Even for commonly high fluxes measured under high parent ion abundance and dwell times (2E6 cps, 0.2 s) the reviewer has found that the noise level affects the high frequency end of the cospectrum (see also comment by referee #1);

AR: We would agree that the short dwell times chosen for the vDEC approach were perhaps not ideal and therefore our measurements may well have suffered from limited counting statistics. However, the issue is more complex than indicated by the referee. The amount of information on concentration that enters a 25-minute flux calculation is in fact very similar between DEC and vDEC. However, while the individual data point is less certain, there are more data points available. The main uncertainty, in our mind, is how well the time lag can be quantified. The effects of this on the results have been discussed in detail in the revised text and are outlined in our response to a similar point raised by reviewer 1.

RC: should it be m/z 37 on page 251 (not m/z 39), aka the first water cluster?

AR: Yes, this should refer to m/z 37 and has been changed accordingly. Section 2.2,



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paragraph 2, line 6 "In addition to these compounds, the primary ion count, measured at m/z 21 (H318O+) and two reagent cluster ions, m/z 37 (H316O+ (H216O+)) and m/z 55 (H316O+ (H216O+)2) were also recorded."

RC: measurement setup relative to roof environment (see also below); the rule of thumb is that the vertical distance from the rooftop should be at least two times the horizontal extension of the roof to avoid the worst;

AR: Regrettably, safety considerations and the need for planning permission meant the maximum height of the mast used on the tower was restricted to 15 m. We presume the "worst" refers to the wake effect of the building. We would refer the reviewer to our comments below which address the effects of wake turbulence at this site.

RC: m/z 69 is likely going to have other interferences in a city besides just furan . benzene

AR: It is true that other compounds may be detected at m/z 69, therefore we have suggested this in the revised manuscript. Section 2.2, paragraph 5, line 11 "Similarly m/z 69 may be isoprene and/or furan, although the latter is normally present at very low concentrations in ambient air (Christian et al., 2004). Despite this, in the urban atmosphere, contributions from unknown compounds such as alkenes cannot be entirely ruled out and therefore m/z 69 was tentatively attributed to isoprene."

RC: One of my main criticisms is the lack of detail on the site description. An urban site is by nature heterogeneous, and the urban fluxnet community (http://www.indiana.edu/˜muhd/, or Oke, 2006) has developed criteria to describe the environment, and to locate an appropriate measurement site (the building used in this study is far from ideal). Very important is the characterization of the surrounding urban morphology, which determines air flow and turbulence parameters, such as displacement height and roughness length, z0. I see no efforts the authors made in that respect although at least one of them has previously published such. A quick look at the internet (e.g. http://www.aidan.co.uk/photo4688.htm) shows that the



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Portland tower has an extensive (rectangular!) footprint and is obviously in the neighborhood of at least one taller building (the Beetham Hilton Tower, to the N). Aside from that, using Google Earth for viewing, it appears to me that the tower height extends significantly past the surrounding buildings ("2 times?). These are important observations: The former suggest that there is at least one extensive wind direction sector that is influenced by a wake from a neighboring building. The latter suggests that the wake produced from the Portland tower itself is going to be large and varying with wind direction due to its rectangular shape. Even though the authors made an effort to escape the building wake effect, an up to 15 degree effect was observed (page 255). I think the stated dependence should be shown and explained together with an analysis of the urban morphology of the surrounding areas. Otherwise, no confidence can be created in the flux data presented. In that respect the statement that "although the mean airflow at the anemometer is affected by the building, the influence can be compensated by standard rotational corrections" (page 255) is misleading, because it suggests that the measured flux under these conditions is representative of the urban areas surrounding the tower (as calculated with a footprint model), which is not the case. Rather it may be representative of the immediate surroundings of the Portland tower, or not at all (see below).

AR: We would agree with the reviewer that our description of the measurement site could have been more detailed. To this end we have expanded the site description to include a classification of the urban morphology based of the criteria described by Oke (2006), details of the surface area of Portland tower as well as details of the surrounding buildings which exceed the height of Portland tower. Section 2.1, line 9. ".The VOC flux measurements were taken from the roof of Portland Tower, an 80 m tall, rectangular (46 m by 16 m) office block, which is located in central Manchester. The building is situated on Portland Street, which is approximately 600 m distance from the Arndale centre, (the citys principal shopping district) 475 m from Piccadilly railway station, (the north-wests busiest station), and 100 m from the China Town district (a concentrated area of restaurants). The office block is surrounded by trafficked streets on three sides



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and a multi-storey car park on the other. The buildings in the immediate vicinity of the tower are predominately commercial thus giving the site an urban classification of 2 according to the criteria described by Oke (2006) (intensively developed high density urban area with 2-5 storey, attached or very close-set buildings often of brick or stone, e.g. old city core)."

The reviewer suggests that no confidence can be put in the data collected at this site due to the wake affect generated from the building itself, as well as from the surrounding buildings. To investigate this claim we performed an analysis of the turbulent statistics at the site which is now included in the body of the manuscript. This analysis, which compares measured values of u*/sdw to values obtained for a set of ideal conditions and is used routinely by the FLUXNET community (Foken, 2004), showed 85% of the data to rank category 6 or better (suitable for general use) and less than 3% of the data qualified for rejection, which would suggest Portland tower to be a suitable (but not ideal) location for urban flux measurements. In addition we would like to draw the referees attention to a previous study made at this location which found Portland Tower to be a reasonable location for flux measurements (Martin et al 2007).

Section 2.3.3, paragraph 7 now reads "Flux measurements made in the urban environment can be subject to additional uncertainty due to the extremely heterogeneous nature of the canopy. It is generally assumed that, provided the measurement location is sufficiently elevated above the street canyon, small scale heterogeneities merge to form one stationary net-flux above the city (Nemitz et al., 2002). However, individual buildings that exceed the average canopy height, including the measurement tower, may generate additional wake effects which affect the flux measurements. Within the footprint of Portland Tower several buildings were identified as potential sources of wake turbulence, including the City Tower (107 m tall) located 0.23 km to the north, Arndale House (90 m) located 0.7 km to the north-west and Beetham Tower (169 m) which was situated 0.9 km to the south-west. In addition, a fourth, shorter building, 111 Piccadilly (64 m) was located to the north-east of Portland Tower. During the measure-



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ment period the prominent wind vectors were from the north, west and south, which meant flux measurements may have been influenced by wake turbulence from the City Tower, Arndale House and 111 Piccadilly, as well as other smaller buildings not listed here. In order to assess the effect of this on flux measurements the integral turbulence statistics of the vertical wind velocity (sdw/u*) were analysed and compared to modelled values which were derived for a set of ideal conditions (Foken et al., 2004). Classification of data based on this test was outlined by Foken et al. (2004) as part of the FLUXNET program. According to their criteria, over 85% of the current data were rated category 6 or better (suitable for general use) and less than 3% of the data qualified for rejection which suggests Portland Tower a suitable location for flux measurement as described here."

RC: As there is no detailed description of the surface and activities from which the measured fluxes are expected to come from, and there are no vertical measurements of wind speed and turbulent structure, one cannot assess what depth and therefore influence the roughness sublayer has on the measured fluxes. The review of Roth (2000) suggests that the roughness sublayer depth over cities may be at least 2, likely closer to 4 times the average roughness element height. Without at least some estimate of the latter, the given measurement height of 95 m and its effective reduction by the buildings wake effect tells little about the flux measurement validity.

AR: see above comment on the effects of wake turbulence at this site.

RC: Meteorological data is given and I assume it was coming from met instrument installations on the Portland tower. If so, the authors need to realize that low wind speeds at such height probably occurred alongside a decoupling from the surface emissions, and high wind speeds may be biased due to the wake effect, as are the wind speeds statistics as a whole. Similarly, measured temperatures should be viewed with caution.

AR: We would agree that low wind speeds observed at the tower are likely to result in a decoupling of the measurement site from the street canyon below. Nevertheless, we would strongly argue that the use of average diurnal flux cycles should provide a robust estimate of the typical day-to-day surface exchange. In order to recognise the reviewers point we have added the following paragraph to Section 3.2 paragraph 4 "Regardless of these factors it is still possible that some of the flux is missed by the measurement systems, as during stable night-time conditions and on calm mornings / evenings the measurement site may become decoupled from the street canyon below. Although this may result in damping of some temporal features, the effect is somewhat reduced through nocturnal heat output from the city and the relatively windy location and therefore the average daily fluxes presented here should provide a robust estimate of the total exchange occurring throughout the day."

RC: Related to the above are the footprint calculations presented on page 264 and in Figure 9. It is unclear whether the input values were taken from typical onsite measurements. The assumed roughness length is most likely an underestimate in these conditions (Roth, 2000). If the authors want to analyze their data properly, they ought to make a better z0 estimate, possibly as a function of direction, and also present the traffic counts collected, assuming traffic is the major contributor to the emissions. It is unclear to me how the footprint analysis was used to compare to the emissions inventory, as the latter does not normally come in the shape of a footprint. Hence, some spatial extrapolation must have occurred aside from the temporal one discussed in the manuscript.

AR: Reviewer raised concerns over the footprint analysis and in particular the low value chosen for z0. In this instance we fully agree that this was an underestimate and have changed this value from 0.4 m to 1.5 m which represents 1/10th of the average building height.

Section 3.3, paragraph 2, line 7 "The following parameters were used in the model: standard deviation of vertical wind velocity sdw = 0.3 m s1; friction velocity u* = 0.3 m s1 (average during measurement period); measurement height zm = 95 m; roughness length z0 = 1.5 m (estimated as 1/10th of the average building height (15 m)); and



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boundary layer height h = 2000 m"

In addition, we have repeated the footprint analysis for the range and average u* conditions observed at the tower. Finally, as suggested we have provided a more detailed description of how this footprint analysis was used to generate an emission estimate for the NAEI and in this new estimate we include a weighting factor for wind direction as suggested by reviewer 1.

Section 3.3, paragraph 2 line 13 now includes "A circular flux footprint (radius = Xr) was then superimposed over a map of NAEI grid squares and the entrained grids averaged using a weighting factor to account for the wind direction during the measurement period."

RC: One thing about the data that confuses me is the lack of a morning rush hour peak, both in the concentration data and the flux data. Unfortunately, Figure 3 does not have enough detail to evaluate this properly. What it does show is a buildup of concentrations under low wind speed conditions (e.g. Thu 8), a small weekend effect for toluene, and lower OVOC abundance after the frontal passage. But this is trivial, and the low wind speed conditions are generally conducive of low u* values, making flux calculations invalid. What this may show though is that the measurement location was far from ideal for the purposes of this study (see above). If the morning rush hour was not picked up due to too low wind speeds, and emitted VOCs are vented quickly instead (Wed 14?), and/or bypass the sensor due to the buildings wake effect (most of the time?), measured fluxes are invalid for comparison to emission inventories. Even if emission rates are low due to a presumably cleaner carfleet (page 264/265) a morning rush-hour peak ought to be observable in a city. Unless the measured toluene (and benzene) fluxes correspond to the collected traffic counts (showing a rush-hour at 11-12 UTC (10 to 11 local time?)), that is, in my opinion, a clear sign of a fundamental flaw in the measurement design (see above), or simply of an insufficient length of data collection (more time needed to observe situations of sufficient turbulence during rush-hour times). Alas, I suggest the authors show the traffic counts and plot u* alongside the measured VOC fluxes to remove the guesswork (note that the afternoon rush hour appears to be observed for toluene.

AR: Reviewer #2 suggests that the absence of a morning rush hour peak is a clear sign of a fundamental flaw in the measurement design. As advised, we have added a plot showing traffic counts (obtained from Oxford Road, which is a busy bus route adjacent to Portland Street) which clearly showed a morning rush hour peak (Revised Figure 7). Reviewer 2 suggests that this rush hour peak was not observed in VOC fluxes as the measurement site was "non ideal" meaning VOC emissions bypassed the sensor due to the wake effects of the building. However, the morning rush hour peaks were seen in particle number fluxes (Martin et al., 2007) which were measured over the same time period. As to why we do not see the same in the VOC fluxes, we believe there are several reasons which we have stated in the revised manuscript.

Section 3.2, paragraph 3, line 7. "The absence of this peak in the flux measurements is not unique to this site (Nemitz et al., 2002) and is thought to relate to a number of factors: firstly, flux measurements represent the average surface exchange occurring over a large area, thus rush hour trends which may be more apparent on some roads (e.g. bus routes such as Oxford road) than others, become smeared. Secondly, during the day there is more stationary traffic, due to loading / unloading and congestion. Thirdly, for measurements of VOCs which might partially be under temperature control, fugitive emission may become an important source at midday when temperatures are warmest and consequently any rush hour trend within the data becomes smoothed. Furthermore the use of products (e.g. cleaning products, solvents) is not likely to be linked to traffic counts. Finally, the fluxes presented here are an average of both weekday and weekend fluxes, which, as shown by Fig. 7, may also dampen any rush hour trend in the data. "

RC: Much of the spread in correlations shown in Figure 4 is trivial which has been pointed to by my fellow referees. I suggest the authors compare these data to existing urban measurements (e.g. Munich), particularly with respect to the range observed,



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which appears quite small to me, and may be the result of the duration of this study and/or the instrument sensitivity.

AR: We feel that the main focus of the paper is on the flux measurements and the comparison of the two techniques, therefore we do not feel it is necessary to compare these concentration measurements with other European cities, especially as we have already placed the observed ranges in context of previous studies from both European (Barcelona, Innsbruck) and U.K cities (Bristol and London).

RC: The parts about isoprene and the B/T ratio are superfluous. They contribute little to the manuscript and contain speculative discussion. I suggest referring to Holzinger et al (1999) on the "isoprene" data. There is probably an interference at m/z 69, and isoprene is a tailpipe emission (cited in Table 2; not evaporative because not a significant component of gasoline).

AR: We agree that perhaps too much attention was given to the discussion of isoprene mixing ratios and the B/T ratios. Therefore we have shortened this section considerably and removed completely the discussion of biogenic isoprene.

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