

Interactive comment on “Annual particle flux observations over a heterogeneous urban area” by L. Järvi et al.

Anonymous Referee #1

Received and published: 30 June 2009

General Impression:

Particulates in urban areas are of concern to human health. The understanding of the sources of particles is limited and emission factors are uncertain. Emission estimates are almost entirely based on emission factors from test stands, rather than measurements in the real environment. Although several studies have recently been presented on micrometeorological measurements of particle fluxes in the urban environment, the paper by Järvi et al presents the first annual dataset to date and therefore makes a welcome addition to the sparse literature on this subject. The paper is concise, all material is useful. There are a few minor scientific points the authors should address prior to acceptance for publication in ACP. In addition, there are a large number of minor improvements to the English (in particular the addition of omitted articles) that should be

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made prior to publication. These are too numerous to be listed here and this Reviewer recommends checking by a native speaker.

Major Scientific Comments:

The measurement and data analysis approach seems generally sound. However, some uncertainties can be expected due to effects from the hill and advection across the heterogeneous emission area. Although these effects are difficult to quantify, they should at least be mentioned. The authors do not appear to conduct a stationarity test on their data. While this may be more difficult to achieve for particle number fluxes (due to statistical limitations), the authors may want to consider basing the stationarity test on the CO₂ data. Possibly, they may find that some of the negative flux values are associated with non-stationarities as observed by Nemitz et al. (2002).

It is somewhat disappointing that the authors did not distinguish between fall and winter seasons on a calendar month basis. Surely, temperatures in winter were still lower than in fall, providing contrasting conditions.

Insufficient detail is provided regarding the footprint calculations. As far as I understand, the footprint calculations take account of topography and changes in surface roughness, but not of heterogeneity in the heat fluxes, which leads to limitations in the prediction and should be pointed out. Also, Fig. 1 presumably shows an example calculation for a single flux measurement, rather than the average footprint of the entire dataset. This needs to be pointed out, because the limitation in the footprint calculation has implications for the emission factors derived in the manuscript.

At present, the calculation of the traffic emission rate is unconvincing. Clearly, there are other traffic sources in the footprint, not just the single road, even if it is the largest. At a minimum, the authors should validate the approach by deriving the CO₂ emission rate in the same way to see if this derives reasonable numbers. Since CO₂ emissions are tightly linked to fuel consumption, they are much more tightly constrained.

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Indeed, the CO₂ data are under-used in the current version of the paper and Section 3.5 should be expanded. This reviewer believes that the derivation of relative emission factors of particulars (i.e. relative to CO₂) would derive robust emission factors. For this purpose, the terrestrial source/sink may need to be parameterised, rather than just omitting deposition fluxes of CO₂. Emission fluxes of CO₂ may also be reduced by photosynthesis or enhanced by respiration, thus the exclusion of negative net fluxes is somewhat arbitrary. Some information may be gained by investigating changes in the emission ratio of particles vs. CO₂ as a function of season and time-of-day. Derived emission factors of particles relative to CO₂ could then be combined with typical fuel consumption to derive an alternative estimate of particle emission rates per km.

The particle deposition to vegetation estimated in P13416, L16 appears to extrapolate flux measurement results from a forest environment to the city environment. Clearly, the deposition flux depends on the air concentration. A more robust estimate of the deposition to vegetation would be derived by combining V_d to vegetation (e.g. from Pryor et al., 2007) with actual concentrations measured at the urban site.

Interestingly, the rush-hour traffic pattern of Fig. 6a is not reflected in the diurnal cycle of the emission from the road sector. This is consistent with observations elsewhere (e.g. Dorsey et al., 2002; Martin et al., 2008), but has not really satisfactorily been explained in the literature. One explanation could be that traffic counts are often taken on commuter roads, while traffic activity on other streets may not show these rush-hour peaks. Another explanation could be that other emissions, e.g. from cooking, are highest between the rush-hour peaks and therefore smooth out the double-peak pattern. Finally, it could be that during midday conditions are more unstable and the flux measurement reflects more the busy road in the vicinity, while during the rush-hour periods conditions are (on average) less unstable and the footprint is larger. I would like to challenge the authors to add their view to the discussion.

Minor Scientific Comments:

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P13408, L29. The ultrafine particles associated with traffic sources are usually primary in nature.

P13409, L28. Does this figure relate to PM₁₀? Please specify.

P13411, L15. The authors should note that the cut-off of the WCPC is somewhat composition dependent and can be larger for pure soot particles.

P13411, L28. What temperature was the inlet heated to?

P13413, L23. Explain how the values of d were chosen. The standard relationships between d and z_0 only apply for closed vegetation canopies. Have you looked at what d may linearise vertical wind profiles?

P13416, L10. Why were the fluxes from road traffic larger in fall/winter?

P13417, L20. Surely, congestion, leading to stationary traffic would also lead to a non-linearity in the relationship between traffic counts and emissions.

P13418, L1. Maybe the lower flux/traffic ratio in stable conditions could indicate that the measurement tower becomes decoupled from the surface due to inversion conditions? Or, possibly, the flux footprint changes to an area with less surface activity? The authors should expand their discussion here.

P13419, L7. Is it that obvious that particle emission factors (per km) should be lower at 97 than at 40 km/h?

P13420, L13. The causality is probably the other way round than implied: the smallest GMD results when particle number fluxes are largest?

Fig. 3. Please clarify whether this figure represents the peak frequencies for the covariance of vertical wind component with particle concentration or CO₂.

Technical comments:

P13408, L3: better: '... over an urban area in Helsinki, ...'

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P13408,L6: '... with a daytime median flux of 0.8 x ...'

P13408,L8: '... the direction of the road ...'

P13408,L12: '... corresponds to an average emission ...'

There are many minor deficiencies of the English of this type throughout the manuscript, too numerous to list here fully. The manuscript should be checked over by a native speaker.

P13409,L20 & 21. Maybe 'in the city of Munster in Germany' and 'from Boulder, Colorado' should be set in parentheses, because the authors do not really want to say that these are the first measurements in Munster and Boulder.

P13411,L1. '... heterogeneous, consisting of buildings ...'

P13413,L6. 's' should be printed in italics.

P13415, L16. '... with single family houses ...'

P13416, L28. '... were low at night-time, ranging ...'

P13419, L27. '... resulting in negative fluxes.'

P13420, L4 & L6. Use 'R²' rather than 'squared R'.

P13420, L13 & 17. I feel it would help to talk explicitly about 'particle number fluxes' here, since conclusions for mass fluxes would be very different.

Please add horizontal zero lines to Figs. 5, 7 and 10.

Figs. 3-8 are very pixilated. The quality of the embedded graphs should be improved.

Additional Literature Referred to in this Report:

Nemitz, E., Hargreaves, K.J., McDonald, A.G., Dorsey, J.R. and Fowler, D., 2002. Micrometeorological measurements of the urban heat budget and CO₂ emissions on a city scale. *Environmental Science and Technology*, 36: 3139-3146.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 13407, 2009.

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