

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

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We thank both Referees for their careful reading and valuable comments.

The text in the manuscript was improved by a native English speaker. These corrections are not listed separately below. The text was also partly rewritten after the referee comments particularly in Sections 3.3-3.5. The order of Sections 3.4 and 3.5 was changed as in the current version also CO₂ emission factors were calculated with the aid of the footprint function. Section 3.6 was removed from the manuscript as suggested.

Below our responses are listed in more detail.

ANONYMOUS REFEREE #1

C5493

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

Authors: The language of the manuscript has been checked 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).

Authors: The effect of the hill and complex emission area are now mentioned on Page 7, lines 1-4. Stationary test according to Foken and Wichura (1996) was made on both particle and CO₂ flux data, but it was forgotten to mention in the manuscript. The averaged flux periods were divided into 5 sub-intervals and if the difference between

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those two was larger than 60%, the flux was omitted. Sentence has been added on Page 6, lines 29-33.

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.

Authors: The winter 2007-2008 was exceptionally warm in Helsinki, and weather during the typical winter months (Dec-Mar) was similar to typical fall period (Sep-Nov) as temperature ranged between 10 and -5°C and no permanent snow cover could be observed. Thus, there was no meteorological reason to separate winter from fall. We also studied the behavior of particle fluxes during different months and we found them similar in fall/winter.

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.

Authors: The referee is correct about the heat fluxes as the current version of the SCADIS model does not take the thermal effects of the urban boundary layer into account. A sentence about this has been added to Page 8, line 25. Fig. 1 represents one simulation and information about the assumptions are listed on Page 8, line 28-31 and on Fig. 1 label.

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

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are tightly linked to fuel consumption, they are much more tightly constrained.

Authors: The calculation of the particle emission rate was made with the aid the diurnal particle flux and traffic profiles without any reference to the meteorological conditions in the footprint simulation. In the current version, we searched for the particular conditions in which the footprint is valid and calculated both the particle number and CO₂ emission factors. Both of them were found to be similar to reported in previous studies. Due to this improved analysis text in Section 3.5 was largely modified.

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 parameterized, 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 if 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.

Authors: As suggested we plotted the diurnal behavior of emission ratio for different seasons and land use sectors (New Fig. 9). However, as the CO₂ emissions are representative for mixed fleet, which structure (amount of HDV and LDV) we do not know, the calculation of emission factors with the aid of fuel consumed is difficult to do. Thus, those were excluded from the manuscript. We also calculated the correlations between particle and CO₂ fluxes with the negative fluxes included and these were replaced to Table 2.

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

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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.

Authors: Due to the reasons explained in Introduction on Page 4, lines 6-17, we do not wish to use v_d in our analysis. We agree that deposition is relative to particle concentration and thus discussion concerning the strength of the deposition flux has been excluded from Page 10, lines 12-14.

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.

Authors: We added discussion about the missing two peaked behavior in Section 3.3. This Section was also largely re-written as suggested by Referee 2.

Minor Scientific Comments:

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

Authors: The smallest particles are usually formed as secondary particles from the exhaust gases during the dilution processes, while the larger UFP (e.g. soot) are generally primary origin (e.g. Kittelson et al. 1998). The sentence on page 2, line 32 has been modified to be more clear.

C5497

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

Authors: Yes, it is related to PM₁₀ and it has been added to the manuscript (Page 3, line 29).

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.

Authors: The cut-off size of the WCPC is somewhat composition dependent as studied by e.g. Hering et al. (2005). Sentence about this composition dependence has been added on page 5, lines 12-13.

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

Authors: The inlet was heated with a power of 4 W m⁻¹ and it has been added to the manuscript on page 5, line 27.

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?

Authors: The displacement heights were determined to be two-third times the mean canopy height at each sector (Grimmond and Oke, 1999). This was added to the manuscript on Page 7, lines 21-23. We have also tested to calculate the displacement heights from wind profile measurements. However, as the lowest measurement height was 8 meters (well below the mean height of the buildings), negative d resulted in the urban sector and occasionally in other land use sectors. In general, d got values between 3-7 m in the road and vegetation sector, which are similar to those used in this study.

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

Authors: It should read that the particle fluxes were higher in the road sector in fall/winter, which does not mean that emissions from traffic itself were higher. There

C5498

are several possible reasons to this like increased emissions from stationary combustion sources, changes in footprints, more heavy-duty and bus traffic in fall/winter than in other seasons. Discussion about this has been added on page 10, lines 8-10.

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

Authors: Yes, congestion would create non-linearity between particle emissions and traffic counts due to the effect of e.g. vehicle speeds on particle emissions. The Section 3.3 was largely re-written.

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.

Authors: In stable conditions, turbulent mixing is lower which itself explains the lower particle fluxes. Of course, decoupling between surface and measurement height is possible, but the author's think that it is too speculative. Thus, discussion concerning the stable fluxes was left out.

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

Authors: It is not actually as obvious since the relationship of particle emission factors from car speed changes according to car type and also according to driving cycles. Both Morawska et al. (2007) and Ristovski et al. (2005) reported a u-shaped dependence between particle emission and vehicle speed the minimum being approximately 50 km h⁻¹. The sentence has been moved due to the rewritten version of Section 3.5.

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

Authors: As suggested by Referee 2, the section 3.6 has been removed from the
C5499

current version.

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

Authors: Fig. 3 represents the peak frequency of the normalized co-variance of the heat flux. The calculation of nm is based on Horst (1997), where similarity between covariance of scalars is expected. Information was added on page 7, lines 15-17 and in Fig. 3 label.

Technical comments:

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

Authors: Was modified accordingly.

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

Authors: Was modified accordingly.

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

Authors: Sentence was removed due to the removed section 3.6.

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

Authors: Was modified accordingly.

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.

Authors: The language was corrected 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.

Authors: The text has been modified.

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

Authors: Was modified accordingly.

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

Authors: Was modified accordingly.

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

Authors: Was modified accordingly.

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

Authors: Was modified accordingly.

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

Authors: Was modified accordingly.

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

Authors: They have been changed as suggested.

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.

Authors: Text has been modified accordingly.

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

Authors: We added the horizontal zero line to Figure 7, but Figure 5 would have been too messy and we decided not add the zero line. Figure 10 was removed from the manuscript.

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

C5501

Authors: The quality of the figures has been 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.

Authors: The text concerning the CO₂ fluxes is still quite narrow and not the main subject of this paper. Therefore, the reference was left out.

REFEREE #2: James Dorsey

General Comments

The paper describes measurements and analysis of particle flux measurements conducted at the SMEAR III field station in Helsinki between July 2007 and July 2008. The investigation of the seasonal and land use controls on urban aerosol production is generally sound, and adds to the small but growing literature on urban aerosol flux measurements. The paper is of a quality suitable for publication in ACP, subject to the authors addressing the following scientific comments and correcting the numerous typographical and linguistic errors throughout the paper. It would be very useful to have the paper proof read and corrected by a native English speaker. Due to the scientifically useful nature of the paper, this reviewer is willing to assist with proof reading the manuscript if no suitable person can be found at the authors' institutions.

Specific Comments:

It would be very useful to have some information on the magnitude of the co-spectral correction to the measured flux. The use of the fit shown in figure 3 and equation 3 gives rise to some concern about the accuracy of the correction in stable cases, as the fit appears to be less than robust. The mean value and variation in the ratio of the measured to corrected fluxes (F_s/F) would be a suitable measure, and should be clearly stated in section 2.4.

C5502

The median correction factor with its 5 and 95 percentiles was included into Section 2.4 on page 8, lines 13-14.

The method used to derive the footprint shown in Figure 1 is not clearly described. It appears that the footprint is an average for one wind sector, and it should be made more explicit in the figure caption that it is an average rather than an example for a single half hour period. It appears that the authors have assumed a fixed wind direction for footprint calculations in the road sector. It would be useful to explain the reason for doing so rather than using measured wind direction.

Authors: The footprint in Figure 1 represents only one meteorological situation and not an average of footprints. The particular wind direction 117° was chosen as it is perpendicular to the road. The method to describe the footprint calculation has now been improved in Page 8, lines 28-31, and in Figure 1 and 10 captions.

Figure 4 is slightly difficult to read due to pixellation. Is it possible to replot this figure?

Authors: The quality of Figure 4 was improved and it was re-plotted.

The comments on figure 7 suggest that particle emissions should be linearly correlated with traffic counts. No sound basis is given for this assertion, and indeed, lower traffic speed at high traffic flow rates might be expected to give rise to an exponential relationship. This section should be reconsidered.

Authors: The Section 3.3 was re-written and incorrect text about linear relationship between emission and traffic rate was removed.

Section 3.6 on the relationship between particle diameter and particle number flux is somewhat weak, and could be removed from the paper without compromising the scientific quality. The authors seem to state that the correlation between geometric mean aerosol diameter is only observed from one of the three wind sectors, and even then only during one of the three measurement periods. Given the weakness of the correlation, it seems tenuous to use figure 10 as evidence of road traffic being a major

C5503

source of ultrafine aerosol. This is likely to be the case, but the argument is not strongly supported by the data presented.

Authors: The Section 3.6 has been removed from the manuscript according to Referees suggestion. Removing this also removed text from the measurements part as the DMPS data is no longer used.

It would be interesting to see section 3.5 substantially expanded. Given the fact that urban CO₂ and aerosol sources are both likely to be combustion related, it may be possible to distinguish between the effects of different types of combustion by comparing the fluxes in more detail (e.g. space heating vs. transport sources). This referee would suggest adding an extra figure with the CO₂ and aerosol fluxes plotted on the same axes as a diurnal average split by land use type and season. This may go some way towards explaining the differences between the diurnal patterns of traffic counts and aerosol number flux.

Authors: The emission ratio of particle number and CO₂ was studied on diurnal and annual scale and separately for different land use sectors (New Figure 9). We also calculated the emission factors for CO₂.

Technical Corrections:

There are several more areas of the paper which are slightly unclear, but it is difficult to tell which of these are caused by linguistic difficulties. They are, in any case, far too numerous to list here. It seems likely that the paper will benefit greatly from further proof reading.

Authors: We wish to thank the Referee for proof-reading the manuscript.

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

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