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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Direct determination of highly size-resolved turbulent particle fluxes withthe disjunct eddy covariance method and a 12 – stage electrical low pressureimpactor" *by* A. Schmidt and O. Klemm

A. Schmidt and O. Klemm

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Response to the comments of Referee #1:

"The paper addresses an important issue of particle exchange between surface and the atmosphere. It also presents a new application of disjunct eddy covariance method for size resolved particle fluxes. The instrumentation presented seems reliable and is clearly described. The data presented shows the capability of the instrument for field measurements. The paper is well written and the subject is suitable for ACP. However, there are a few errors in the manuscript which should be straightened prior publishing in ACP."

Thanks for the overall positive statement.

MAJOR COMMENTS: "In the Results and discussion part when comparing the EC and simulated DEC fluxes the authors seem to confuse the effects of analyzer response time, or sampling tome of the DE sampler, and sampling interval in the disjunct eddy sampling methods. This is obvious from the sentence "The deviations show a slight underestimation of the DEC fluxes probably due to the high frequent turbulence parts which can not be resolved using the applied measurement interval ∆t" (Page 9010, lines 6-8). Also lines 8-9 on page 9009 are similarly misleading. The long interval between subsequent samples does not cause high frequency loss due to the aliasing of the frequencies higher than sampling frequency to the lower frequencies. On the other hand, the non-instantaneous response time, or the sampling time in the case of DES methods, causes higher frequencies to be lost from the fluxes. Thus in the case of the DEC system described in the paper the high frequency limit is defined by the sampling time of 0.4 s, not by sampling interval of 5 s. Thus the correct high frequency limit is 1.25 Hz."

Indeed, the description of the time different measurement intervals was somewhat misleading. It is correct, that the measurement interval of 5 seconds itself, does not lead to a lower spectral resolution. The idea of the DEC method is that the time series is only thinned out by taking fewer samples during one averaging interval. But, due to the fact that she single samples are taken fast (within 0.4 seconds in our case) every 5 seconds, the record frequency is 0.4 Hz but the Nyquist frequency is 1.25 Hz. Therefore, the loss of spectral information is acceptable when using the DEC method. The authors agree that the wording (Page 9010, lines 6-8, page 9009 lines 8-9) has to be replaced and adapted to the actual course of action. The respective part of the text was corrected and enhanced to clear the situation. Thanks for the helpful hint.

"The authors state on page 9012, lines 11-14, that the main diurnal cycle of the particle fluxes would be related to diurnal cycle of surface layer turbulence. Also, on page 9013, lines 4-5, they claim that the mean fluxes of smallest particle size classes are depen-

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dent on turbulent regime. However, comparing the particle fluxes on weekdays and to fluxes on Sundays (Figure 9a) one can clearly see that the driver of the diurnal cycle is not the diurnal cycle of turbulence, but that of emissions. This is also how it should be based on our theoretical knowledge on turbulent transport. Unless the turbulence is not for example stirring up material from surface, the flux should be independent on the strength of the turbulence, provided that turbulence exceeds some minimum value. This is the reason to filter the data using a suitable u* threshold value."

Concerning the comment on the text passage on page 9012 lines 11-14: This is not really what we state. We wrote: “The total particle number flux exhibits a pattern with 3 peaks (about 7:30, 13:00, and 16:00 local time) that are embedded in the daily main peak that is obviously related to the known diurnal cycle of atmospheric turbulence development" Hence, of course the daily run of the fluxes is always a consequence of both influences – (traffic) emissions and development of the boundary layer (i.e. turbulence regime). The threshold value of the friction velocity was obtained to be 0.15 m/s and was used for filtering the data with respect to the strength of turbulence (p 9005, lines 6-8). But the flux is not independent from the strength of turbulence. The development of turbulence exhibits a diurnal pattern and has an important effect on the fluxes. This superposition of meteorological and anthropogenic causes, both having an effect on the particle flux values, has also been observed and stated in other studies about particle fluxes above urban areas e.g.:

Mårtensson, E. M., Nilsson, E. D., Buzorius, G., and Johansson C.: Eddy covariance measurements and parameterisation of traffic related particle emissions in an urban environment, Atmos. Chem. Phys., 6, 769–785, 2006.

Dorsey, J. R., Nemitz, E., Gallagher, M. W., Fowler, D., Williams, P. I., Bower, K. N., and Beswick, K. M.: Direct measurements and parameterisation of aerosol flux, concentration and emission velocity above a city, Atmos. Environ., 36, 791–800, 2002.

In order to clear the situation, we enhanced the text by the explanation of this superpo-

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sition.

"The authors do not provide any estimates of uncertainty for the flux values they present. The fluxes shown in Figure 4 d seem to be below detection limit of the system. The authors should clearly state if this is the case or not instead of a vague sentence "...the exchange of coarse particles appears to be more or less balanced within a diurnal cycle". There are also a few other parts in the paper, in which the uncertainty estimates would help interpretation of the data. The discussion on the Figure 9b, on page 9015 lines 7-9, should include estimation of the significance of these differences. Also the fluxes of the coarsest particles (Figures 9c) may not differ significantly from zero, which means that their fluxes are below detection limit of the system. Therefore the discussion on their differences between weekdays and Sundays (page 9015, lines 9-12 is not meaningful. In the case that the flux values of the coarsest particles are not significantly different from zero, its value in the figure 10 shows not the significance of this size class to the total particle exchange, but instrumental background noise."

This comment, concerning the significance of some results, is not objectively comprehensible. We do provide estimates of uncertainty for the turbulent flux values. The errors were calculated and are given in the text (page 9007, line 10 to page 9008, line 8), according to Buzorius et al. (2003). Also the used equations and approaches to obtain the uncertainties were described (page 9007, Eq. 4). The statement: “The fluxes shown in Figure 4 d seem to be below detection limit of the system.“ is indeed a vague sentence. Measurements below the detection limits of the used devices were not analysed as described on page 9005 in the discussion paper. In fact, we used a very high signal-to-noise ratio of at least 3 (page 9005, lines 14-16). Thus, insecure low quality data was not respected for presentation at all, as described in the text. That’s why our data amount shrunk from about 4800 to 3500 half-hourly values (page 9005, lines 19-24). The data availability of such reliable measurements decreases for the coarser particles, but this was also mentioned on page 9008, lines 3-8. The fact, that the flux values in Figure 9c are very low does not mean that the values are below any detection

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limit. The detection limits refer to the measurement of the respective concentrations. The calculated fluxes can be close to zero even if the concentration values are high. As we used the covariance of (preliminary quality tested) concentrations and vertical wind velocities for the flux calculations, the detection limit constraints do not affect the significance of low flux values in the way as mentioned by referee #1. Thus, the values in Figure 4d and 9c are surely no instrument noise but reliable, albeit small flux values. The same applies to the values shown in Figure 10 and the derived conclusions. Nevertheless, with respect to the current comment of referee #1 and the corresponding minor comment on Table 3 at the end “Table 3: Uncertainties on the net fluxes would be useful..“ we added the uncertainties in Table 3, with respect to the measurement errors as well as the sampling frequency reduction, according to Eq. 4 on page 9007. In addition we added the important information about the value of the electrometer noise level to the text. For the used range of 100.000 fA the maximum noise of the electrometers is only ś15 fA (Dekati Ltd., 2006). This information was added to the caption of Figure 2, where the applied border for acceptance of measurement values (which is at least three times the electrometer noise level (page 9005, lines 14-16)) is marked. The range for this electrometer noise level was also added to the text on page 9005, lines 14-18.

MINOR COMMENTS:

"Page 8999, lines 4-5: "Also the formation of clouds is driven by aerosols that function as condensation nuclei..." I would rather say that the driver for the formation of clouds is the flow dynamics of the atmosphere. This defines where the clouds form and where not. The aerosol particles of course are important in serving as CCN, but their scarcity rarely suppresses the clouds from forming when other conditions are met."

The wording was changed into the accurate expression: “Furthermore, aerosol particles have an effect on the formation and characteristics, e.g. droplet number, of clouds by functioning as condensation nuclei“

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"Page 8999, lines 26-28: "Considering... ...needed". This sentence seems somehow muddled."

This was corrected. The wording in the text is improved now.

"Page 9000, line 3: "...EC method has to be replaced by other methods" The authors could mention some of these methods here. At least relaxed eddy accumulation has been applied to aerosol particle flux measurements (Gaman et al., 2004)."

We mentioned some other approaches. With respect to the hint of referee #1 and referee #2 we added also the REA method and referred also to the work of Gaman et al. (2004). An additional hint to these approaches and related studies at this text passage appears to be misplaced since we are going to introduce the capability of the DEC method right afterwards.

"Page 9001, lines 14-17. "With respect... ...behind the measurement region of the 3D ultrasonic anemometer". Why was the inlet placed behind the anemometer? According to Kristenssen et al. (1997), the placement of inlet below the sonic anemometer causes a minimal loss of flux with no wind direction dependent lag time. How close was the inlet from the sonic, and was there any detectable lag-time due to this sensor displacement."

Since we used a small, downward directed cone on top of the inlet tubing to avoid that rain water gets into the valve box or the ELPI, the inlet will cause a (probably small) flow distortion if wind comes from behind the inlet. Besides, the reference mentioned by referee #2 (Kristensen et al., 1997) does not give a general conclusion but an experience with a small thermometer as measurement device for the scalar in combination with a simple model. In fact, the best placement depends on the current construction of the setup and the geometric properties of the inlet. Thus, in our case, a placing of the inlet below the sonic anemometer would certainly among the worst possible options when measuring vertical particle fluxes. The position of the inlet was about 15 cm downwind (with respect to the main wind direction) of the anemometer. This detailed information (15 cm) was added to the text. Significant lags were detectable for the H2Og and CO2

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concentrations measured with 10 Hz and had to be corrected. The lag between the aerosol concentrations and the vertical wind velocity were negligible because of the 1 Hz resolution of the concentration records. For the synchronisation we used the flow through the ELPI (25 l min-1) and the valve-open signal, recorded with 10 Hz (page 9006, lines 7-12 in the discussion paper).

"Page 9005, line 2: "...WPL-correction..." Is this correction needed? The inlet is likely to damp most of the temperature fluctuations before the air gets into the ELPI (see e.g. Rannik et al., 1997)."

Yes, the correction is definitely needed in order to get reliable values, at least for the carbon dioxide fluxes but we think also for the particle fluxes, because of the two terms in the WPL equation (Webb et al., 1980). One term of this equation respects the density fluctuations caused by water vapour transport. In order to avoid this error it is recommended to apply the WPL correction, and the temperature fluctuations are not negligible for the determination of the CO2 fluxes. Further, Rannik et al. (1997) recommend a ratio between the length of the inlet tubing and its inner diameter of about 1000 in order to damp these fluctuations enough to disregard it. This ratio is not reached with our setup (see page 9001, lines 14-28 in the discussion paper). Thus we prefer to respect the correction because it can be done automatically in our analysis software package anyway and there is no reason to ignore or accept this (probably small) error.

"Page 9007, Equation (3): This equation is not immediately clear. Maybe a bit more explicit description could be included below it"

We added some text below the equation (3). In addition, we enhanced the equation to make it more clearly by showing all implemented steps of calculation.

"Page 9007, lines 13-15: "This was found by Bosveld and Beljaars (2001)". Similar results have also been obtained by Haugen (1974), Kaimal and Gaynor (1983), Lenschow et al. (1994), Rinne et al. (2000; 2008), and Hendriks et al. (2008), using

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simulations and field observations. The reference to Hendriks et al. is made again on page 9009, lines 10-12. Thus there seems to be some redundancy here."

Yes, there are also other studies which obtained that. We extended the list to respect these contributions in an appropriate way. As a consequence of this correction, we deleted the following reference to Hendriks et al. (2008) on page 9009, lines 10-12 (which was not redundant here in the former (discussion) version of the manuscript).

"Page 9010, lines 12-19: The Figure 4 should be referred already in this paragraph."

Yes, the reference to Figure 4 was placed here in the revised manuscript. Thanks for the hint.

"Page 9014, line 25: I would write "...biological net uptake of CO2 by the vegetation..." "

This was corrected to improve the accuracy of wording.

"Page 9015, line 2: "...small particles have no meaningful negative flux intervals..." Do the authors mean "small particles have no significant negative flux intervals..." "

No, that is not what we meant. All flux values that were further analysed are significant in statistical terms. Meaningful means, that there are no negative fluxes that can be related to any daily assimilation course (i.e. biological uptake) which was discussed in the context on page 9014 line 22 to page 9015 line 3. The fluxes do not show the high values as on weekdays but, due to the increased turbulence during daytime which is independent from the day of the week, increase from around 0 up to ≈ 8 x 106 particles m-2 s-1 (Stages 1-3).

"Page 9017, line 4: "The different size bins show different turbulent dynamics during daytime" I do not understand the meaning of this sentence."

The sentence was replaced. It is surely better understandable now.

"Table 3: Uncertainties on the net fluxes would be useful. Also the formulation of

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the figures in the last column is confusing. I suspect the authors mean 1013 by the expression E+13. The formulation must be changed to a more conventional one."

As suggested by referee #1, the notation of numbers in Table 3 has been changed into a more conventional one which was used in the text. A Column with uncertainties for the total flux values was added to the Table 3.

The authors would like to thank referee #1 for his time invested and for the very thorough comments and hints which helped to improve the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 8997, 2008.