

Interactive comment on “Gas-particle interactions above a Dutch heathland: II. Concentrations and surface exchange fluxes of atmospheric particles” by E. Nemitz et al.

E. Nemitz et al.

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We are very grateful to Üllar Rannik for his insightful, helpful and detailed comments, which have helped to improve the manuscript and have therefore been acknowledged in the acknowledgement section. Firstly, many thanks for pointing out the problem with Eq. (10) and the legend of Table 4. Clearly, u_* in Eq. (10) should not have been in the power of two. We have double-checked the calculation and no power of two was used for any of the calculation of the turbulent time-scales presented in Table 4 and Fig. 5. In addition, the labels in Table 4 should indeed have been minutes instead of hours. Since the calculations are correct, the conclusions do not change.

Specific comments

1. The ASASP-x optical particle counter deployed during this study has been used

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for eddy covariance flux measurements in several studies. The sampling volume of this instrument is very small indeed and tests indicate that the 95% response of our setup was better than 3 Hz, sufficient for eddy covariance at the measurement height used. We have added the following text: "Tests revealed that this setup had a 95% response time of < 0.3 s, sufficient for eddy-covariance flux measurements even over short vegetation."

2. We accept Üllar's comment that the detrending exercise would have been redundant when spectral high-pass filtering is applied afterwards anyway. However, fluxes were also calculated using just the detrending algorithm to provide a reference for the assessment of the effect of the spectral high-pass filter. As mentioned in the text, the difference was insignificant, suggesting that the high-pass filter did not reduce the fluxes any further than the detrending algorithm. The cut-off of the high-pass filter follows the suggestions of Fontan et al. (1994), which has presented the best (co-)spectral density functions for aerosol fluxes so far. Incidentally, we here derive some of the fasted deposition rates that have been measured for short vegetation, while Üllar implicitly suggests we may have underestimated the flux.

3. Üllar is absolutely right in saying that u_* should not have been in the power of 2 and this has been corrected. The coefficient of 1.75 is taken from Brost et al. (1988). It derives from the relationship between u_* and σ_w under neutral conditions. The eddy diffusivity is given by $K_H = \tau_d \sigma_w^2$ and, alternatively, as $K_H = \kappa(z-d)u_*$. From this follows $\tau_d = \kappa(z-d)u_*\sigma_w^2$. Brost and Wyngaard (1987) found $\sigma_w = 1.32u_*$ and substitution yields (the corrected) Eq. (10).

4. The units of the size-distribution function ($dm / d\log d_p$) would also be kg m^{-3} as the logarithm is non-dimensional. However, the wording here was indeed ambiguous. What is meant is that m_i is the mass loading in size interval i . This has been reworded in the revised manuscript: "... from measurements of the mass contained in discrete size bins (m_i) ..."

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5. In general, the word 'correlation' is used both in a statistical sense and in a more qualitative sense. In order not to confuse the two, we have therefore changed the word 'correlation' into 'similarity', which has the correct geometrical meaning.

6. We are obviously aware of the general v-shape of $V_d(D_p)$. However, we do not feel that an extensive discussion of this shape and the underlying processes would be of value at this point in the results section. We have, however, changed the wording to make it more explicit for this size range: "..., which is in agreement with other measurements and model predictions for this size range ($D_p > 0.1\mu\text{m}$) (Gallagher et al., 1997)."

7. In the text, to which Üllar refers in his comment, we justify why, for the generation of Fig. 3, no filtering was necessary to exclude samples with poor counting statistics. Poor counting statistics lead to a random rather than systematic error, which is reduced by averaging several sampling periods. The reader is not meant to derive the uncertainty from Fig. 3 as implied by Üllar's comment. We have changed the wording somewhat to make this clearer. We also tried to include error bars in Fig. 3 and decided that they make the figure much harder to read without adding more information.

8. Eq. (12) is a purely empirical equation, which was derived from the measurement data. The value of V_{ds}/u_* for stable conditions is set to the value found at neutral conditions, which did not appear to differ between sizes (cf Fig. 3 at $1/L = 0$). The current theoretical understanding of the particle deposition process is incomplete and the response of V_{ds} to atmospheric stability is not captured by current models. Fig. 3 (and Wesley et al., 1985) suggests, however, that this effect can be very significant. Since we do not understand how stability affects the deposition process, it is difficult to speculate how V_{ds} should change with particle size under (very) stable conditions. We have added some discussion to this effect in the discussion section rather than the results section: "Interestingly, in neutral conditions ($1/L = 0$ in Fig. 3), V_d appeared to be insensitive to particle diameter over the size-range of the measurements."

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9. The value of d (0.54 m) has been added to Section 2.2.2, where d is defined.

10. See introductory comment.

11. A brief physical explanation has been added: "Particles that evaporate during deposition contribute to a larger number concentration of particles of a smaller size class close to the surface, leading to apparent emission gradients in this smaller size class. This reconciles the ..."

12. Only small gas-particle conversion fluxes are needed to significantly modify surface / atmosphere exchange fluxes and vertical concentration gradients. By contrast, much larger conversion is needed to significantly alter the total concentration. We hypothesise that, if we could measure horizontal concentration gradients accurately, we would probably see a small difference.

13. We are not aware of anybody having demonstrated that there is an effect of w on the inlet sampling efficiency for such small particles, but agree that it is theoretically feasible. We have therefore removed statement (i) from the text.

14. See reply to comment 1.

15. See reply to introductory comment.

16. As before.

17. A caveat has been added to the text: "By contrast, flux measurements provide a powerful tool for estimating particle growth or evaporation rates within the uncertainties of the flux measurements themselves."

18. These time-scales are consistent with Fig. 5, but inconsistent with the old Table 4, where the labels have now been corrected.

Technical corrections

1. K_e has now been introduced directly following Eq. (3).

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2. h has now been introduced as relative humidity at its first occurrence.
3. The text talked about 2-hour filter-pack runs, while Table 1 listed the duration as 1-3 hours. This has now been made consistent.
4. This has been corrected.
5. The period of the measurement campaign has been added to Section 1.3.
6. This has already been corrected at pre-review stage.
7. The text has been made more explicit: "... the comparison of measured vapour phase concentration products with their theoretical equilibrium values, ..."
8. It should actually have been Figure 4. This was corrected at pre-review stage.

Additional changes

The Vong et al. (2004) reference which had been substituted with a pers. commun. due to delays with the publication of this work has been re-inserted.

Brost, R.A. and Wyngaard, J.C.: A model study of the stably stratified planetary boundary layer. *J. Atmos. Sci.*, 35, 1427-1440, 1978.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 4, 1519, 2004.

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