

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

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General comment

The manuscript by Nemitz et al. 'Gas-particle interactions above a Dutch heathland: II. Concentrations and surface exchange fluxes of atmospheric particles' presents a lot of interesting material, measurements and analysis results on particle flux measurements as well as measurements of relevant gas concentrations and their gradients. In general, the subject of the study is of high scientific interest because 1. Particle fluxes are difficult to measure experimentally, in particular size-resolved measurements are difficult to perform with sufficient accuracy, 2. The particle flux measurement results are often difficult to interpret. Current paper copes with both of these challenging issues

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and is therefore very valuable, presenting new insights into transformation processes occurring during vertical turbulent transport of aerosol particles. The research methods used in the study are appropriate as well as the analysis methods. The manuscript is clearly presented, however the minor deficiency of the manuscript is its length, which is acceptable concerning the amount of material presented (I do not have good suggestions how to shorten the paper). The paper is suitable for the journal and deserves definitely publication.

Agreeing generally with measurement results and interpretation, I do have also a major concern related to the time-scale analysis of chemical reactions and turbulent transport and discussion and conclusions based on the time-scale analysis. There are inconsistencies/errors in the manuscript related to time-scale analysis, which have to be clarified. First, the equation (10) of diffusive time scale is incorrect dimensionally ( $u^*$  should not be in second power). Second, the chemical time-scales presented in Table 4 are far too long (in the order of an hour) for being important in turbulent transfer processes at about 3 m height level. The order of magnitude of turbulent transport at such heights is in seconds. It is probable that the units of chemical time scales are incorrect, being minutes instead of hours as presented in Table 4. Even then the turbulent transport time (seconds) tends to be much shorter than the chemical time scale. Concerning eq. (10), there can be misprint and the analysis is actually correctly done. Both errors mentioned here have a significant influence on the conclusions based on time-scale analysis. Therefore this issue needs to be clarified before the manuscript can be accepted for final publication.

Thus, I expect that the authors clarify the major issue and wait for publication of this interesting paper.

#### Specific comments

1. P. 8, l. 20, the authors mention poor counting statistics as a limitation for determination of high-frequency part of the particle (Fourier) spectrum. What is the frequency

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response time of the instrument (OPC) and could that be also limiting as the instrument is not possibly able to register high-frequency fluctuations? Since measurements are performed at low height with relatively high-frequency turbulence, the frequency response characteristics of OPC are important to mention.

2. P. 8 last lines and p. 9 first two, the cut-off frequency 0.01 Hz corresponds to 100 s as a filtering 'window', this is rather short time for calculation of turbulent fluxes even at such low height as 3 m. How was linear detrending (mentioned in p. 8, l. 15) combined with this frequency-filtering and what was the point in linear detrending if frequencies lower than 0.01 Hz were omitted anyway?

3. P. 10, eq. (10), dimensionally not correct.  $u^*$  should not be in second power and where the coefficient 1.75 comes from?

4. P. 10, eq. (11),  $m_i$  is the mass-based size distribution, dimensionality shows that the units of  $m_i$  are  $\text{kg}/\text{m}^3$ , i.e. directly mass in size interval?

5. P. 11, l. 13 and l. 17, I think it is not proper to talk about 'high correlation', one cannot conclude really about correlation based on Fig. 1. It is more about the 'same pattern' and loosely speaking I understand what the authors' mean, but the correlation in statistical sense could be very low.

6. P. 11, last line, the authors mention that  $v_d$  increases with particle size. This is correct for sizes 100 nm and larger, but it would be also good to mention that  $v_d$  increases for smaller sizes as well in Brownian-diffusion-controlled deposition regime (100 nm is roughly a deposition minimum).

7. P. 12, l. 12, it is not evident from Fig. 3 that the statistical variation is averaged out. One should either present uncertainty intervals of averages (for example, as standard error of the average) or restate the sentence.

8. P. 12, eq. (12) and Fig. 3, is it a common parameterisation that normalised dep. velocity ( $v_d/u^*$ ) is not dependent on particle size under neutral conditions? This is not

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certainly true for all particle sizes (at least for below 100 nm particles) and therefore the applicability (size) range of the parameterisation should be stressed.

9. P. 14, l. 5,  $z-d = 3$ , it is not mentioned anywhere what is the value of  $d$ . It should be probably presented earlier in a relevant place.

10. P. 14, l. 5, Table 4 and Fig. 5. Is it correct that the units of chemical time scales in Table 4 are hrs? If yes, then the critical  $u^*$  values are incorrect. The turbulent time scale should be in the order of  $\kappa z/u^* \sim 0.4^3/0.4 = 3$  s, i.e. very small. Corresponding critical  $u^*$  values would be also very small. Is the conclusion in l. 16 correct?

11. P. 16, sentence starting with l. 9, a reference is given to statement that 'apparent emission of small particles may also result from aerosol evaporation'. This is in contrary to what is argued in bottom of p. 4 ('By contrast...'). Although the reference is given, I miss here as short as possible physical explanation for the mechanism.

12. P. 17, l.2, it is argued that the horizontal transport time over 1.5 km distance was possibly too short for gas-particle conversion to be effective. On the other hand, the vertical transport time in the order of seconds is long enough for the gpic to be important. There is at least apparent contradiction in the presumption presented here.

13. P. 19, l. 9, the statement i) does not look quite valid to me. If the inlet artefacts exist, then it is quite likely that such errors are correlated with  $w$  because a) if inlet is positioned vertically, then it is not 'symmetric' to  $w$ , b) if it is positioned horizontally, horizontal wind speed is correlated with  $w$  (momentum flux), and there can be again 'asymmetry' to  $w$ . I do not want to claim that there are artefacts because of 'sample inlet imperfections' in measurements, but the statement itself does not sound quite correct to me.

14. P. 19, l. 15, see specific comment 1.

15. P. 22, l. 17, l. 18, and l. 22, time scale analysis, check if the conclusion based on Table 4 and 5 is correct, if the time-scale range 1-8 minutes is correct, and if critical  $u^*$

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value 0.12 m/s is correctly estimated.

16. P. 23, l. 14, need for modified gradient-technique relies on the correct time-scale analysis. Reconsider the statement, if needed.

17. P. 24, l. 1, the measurements involve frequently large uncertainties and estimation of particle growth or evaporation rates from flux measurements is therefore subject to large uncertainties, too. This should be mentioned.

18. P. 25, l. 19, chemical time-scale, inconsistent with Table 4 and also large uncertainty coming from  $\alpha$  (100 times variation range as presented in p. 10, last line).

#### Technical corrections

1. P. 3, eqs. (1)-(3), coefficient  $K_e$  is not defined nor mentioned in its first appearance.

2. P. 6, l. 5,  $h$  is not defined.

3. P. 7, l. 28, there is no measurements reported in Table 1 with '2-hour filter-pack run', which I interpret from the text as the measurements with 2 hour averaging.

4. P. 8, l. 7, reference should be probably Nemitz at al. [2004a].

5. P. 11, Beginning of results. I think nowhere is mentioned what is the period of measurements (except later the dates are presented in Table 4).

6. P. 12, l. 21, Section number 3.3 appears twice.

7. P. 14, previous to last line, expression 'the investigations into the equilibrium' is loose.

8. P. 18, l. 7, should be Fig. 6.

9. Figure 4, lines for SJAC  $\text{NH}_4^+$  and Filter Pack  $\text{NH}_4^+$  are difficult to separate in the figure.

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