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Biogeosciences Discussions

Interactive comment on "Aerosol fluxes and particle growth above managed grassland" *by* E. Nemitz et al.

E. Nemitz et al.

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We thanks the three anonymous reviewers for their thorough reading of the manuscript and their constructive and thoughtful comments that have helped to improve this paper. All three referees judge the manuscript to be interesting and original and worth of publication in Biogeosciences. Here we address the individual comments in turn and describe how we have taken them on board in the revised manuscript. The original text of the referees is printed in italics.

Anonymous Referee #1

1) The mass accommodation coefficient measured by Rudolf et al. for HNO3 is 0.5-1.0 not 0.3-0.8 (see page 352 in BGD version)

The stated range was the overlap in the ranges of the three studies quoted. However,

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we have now extended the cited range of possible accommodation coefficients to 0.3 – 1.0. Since a value of 0.5 is used, this does not affect the calculations.

2) In page 353 it is mentioned that ammonium nitrate is - in practice - the only condensing vapour. However, it is hard to see from the presented data. Therefore, I hope that authors could further clarify this point. Since, the growth over 11 nm detection limit could also be due to hygroscopic growth.

We are somewhat surprised by this comment, because the original manuscript already goes into some detail why we believe that the growth is not due to uptake of water vapour or organic gases. We believe we have dealt with the potential of VOCs contributing to the observed growth sufficiently in the manuscript. However, also in response to Referee #2 below, we have now added a statement comparing the latent heat fluxes before (49 W m-2) and after the cut (52 W m-2), which now numerically supports that no significant change in hygroscopic growth is to be expected.

3) In page 358, it is said that Aitken mode consists mainly of primary organic aerosol. I think that there is no justification for that statement. Hopefully authors could clarify or change this statement.

We agree that we do not have measurement evidence for this from this study and actually we did not mean to make this statement; our sentence was phrased the wrong way round: what matters for this argument is that much of the mass of primary organic aerosol (POA) falls mainly into the Aitken mode, rather than that the Aitken mode is dominated by POA. We have re-phrased the statement accordingly.

Anonymous Referee #2

This reviewer raises specific comments that should be used to further increase the quality of the manuscript. The most important one is the unclear presentation of some sections of the theoretical reasoning (chapter 2.4). Some Figures need editing as well. Most of the manuscript is densely written, thus a bit hard to read and follow.

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These general points will be addressed in the individual points raised by this Referee below. It is clear that this Referee raises concerns about the clarity of the manuscript which were not picked up by the other two referees, suggesting that the current readability depends on the scientific background of the reader. However, every effort was made to increase readability for a wider audience in response to the helpful, constructive comments of this Referee.

For example, one can follow the reasoning given in the sentence on page 354 (last line) and the first two lines of page 355. However, the authors require the reader to be very much familiar with the material. That is okay, just may result a negative feedback on the readability for some of the potential audience.

We have simplified this particular sentence, without changing its meaning.

p 350, lines 3-5: Ke is a good approximation for humidities below the relative humidity of deliquescence (about 80% for (NH4)2SO4 and 60% for NH4NO3): Why do you restrict that to the low humidities?

For solid aerosol the co-existence of other compounds does not affect Ke, while for aqueous aerosol it does. This was only implicitly but not explicitly stated in the original manuscript and the wording has been changed accordingly.

p 350, lines 14 - 15: Particle growth implies that some particles will grow across the lower cut-off of the particle counter (dp=11 nm or r=5.5 nm) as they deposit: This sentence comes out of the blue here. You probably want to say: If one plans to observe particle growth with a simple particle counter without size information, ... The following sentence is even less clear. Where do you assume particle growth (across the detection limit of the analyzer) to occur? What are the deposition velocities of particles of various sizes in relation to each other? Please rephrase this paragraph in a more clear and detailed fashion. After having read the entire paper, it is clear what is meant. However, a reader going through the manuscript does not have sufficient information yet at this point.

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We appreciate this criticism. The challenge here is to introduce the idea of using the apparent emission fluxes for the estimation for growth rates, before the observation of emission fluxes is actually described. We have expanded the introduction to this section to read: "As shown in the Results Section below, the measured time-series of particle fluxes changed from mainly small deposition prior to fertilization, to clear diurnal cycles with consistent and significant apparent upward fluxes after fertilization. As discussed below, this behaviour is attributed to particle growth during deposition. Where particles grow during the deposition process, e.g. due to uptake of secondary aerosol products or due to water uptake, this particle growth implies that some particles will grow across the lower cut-off of the particle counter (d_p = 11 nm) as they deposit. As a result the concentration of particles $>$ 11 nm may increase towards the ground at the same time as the total number of particles decreases due to deposition. In this Section we show that this growth can result in a situation where eddy-covariance may derive an apparent upward flux of particles $>$ 11 nm. We also develop the mathematical concepts to relate the apparent emission flux to the growth rate of the particles."

page 350, lines 24-26: Please be more specific and clear what delta_chi/delta_t and Q_chi mean, and what exactly is meant with storage in this context. This also refers to page 351, lines 2-4, and line 5 as well. It is clear to this reviewer what is meant with flux divergence here, but the authors are requested to help a reader to follow their reasoning more easily.

We have significantly expanded the explanation of Eq. (3) in response to this referee and the derivation of Eq. (4) is explained in more detail now.

page 352, line 12: Start a new paragraph here. Tell the reader what you are doing and why. Plus you start to become unclear here. A lot of stuff in just a few lines! What, all over sudden, is the flux expected in the absence of chemical reactions in terms of equation (3)? What is the average deposition velocity? Is it average over time, over

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size classes, over height? What is Ni in equation (10)? Is i an index, if yes, what does it indicate? The latter is a very important point that must be made clear here! (Later in the maunscript, it is said, what it is. But that is not good enough here. Plus, the later explanation is just vague as well). Is the measured flux (line 13) identical to F_N in line 15 and equation (10)? If yes, the r.h.s of equation (10) can be transformed into: F_N(1 - chi_Ni / chi_N) / z_m. Without knowing what Ni means, the meaning (and potential validity, limitations, ...) of the entire equation (10) remains hidden. Further, what is the specifics of a measurement before fertilization?

Again we have significantly expanded on the derivation of Eq. (10) (now Eq. 11) and have introduced a new term, the surface flux, to aid the discussion.

page 352, line 18: Please indicate in a more detailed fashion what you mean with the growth rate across the size cut-offs. What is the term in the preceding equations, what is the unit?

We have added the following parentheses: "(i.e. dd_p/dt at d_p = 11 nm, in nm hr^{-1})".

page 352, line 21: 11nm: Are you talking about diameters now? If yes, why do you jump between diameters and radius?

Typically sizes of particles are given in diameter. However, for the mathematical derivative it is more appropriate to work with radii. This was the reasoning why we used both in the manuscript. On reflection, however, we agree with the referee and have everything transformed into diameter space. This has changed the growth rates which are now ddp/dt rather than dr/dt.

page 353, lines 18-20: This sentence is unclear. What do the frequencies indicate exactly?

They are the fraction of time during which emission or deposition was observed. This has now been made clearer.

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Table 1: It seems a little awkward to indicate negative deposition velocities. Would it not be better to use the term transfer velocity instead?

For deposition velocity the sign convention is clearer than for 'transfer velocities' which could be defined with either sign. We therefore decided to stick with the original concept.

Fig. 2: Printed on a s/w printer, there is no chance to distinguish between the lines properly. How it is on a (high quality) color printer, this reviewer is not sure. On the screen it is okay as long as you spend enough time deciphering the lines and coding. In the opinion of this reviewer, the quality of this Figure is unacceptable. This also applies to Fig. 10.

The advantage of an online journal such as BG is that figures can be set in colour and can thus be more complicated. The Reviewer cannot reasonably expect that all colour figures are readable in black and white. The colour figures (including the online version) are, in our mind, very easy to read. Having said this, we did make an attempt to make them suitable for b/w printing and agree that their readability decreased during transfer into the BG format. In the revised manuscript we have tried to increase the spacing of the dotted line to make them easier to tell apart.

Fig. 2, lower panel: There is a red line around the zero line that is not explained. The caption section: Also indicated are the periods of the MOUDI impactor runs (B to D) is unclear. If that is the mysterious line, it is presented with too little detail.

The sentence on the MOUDI run was erroneously left from an earlier version of the manuscript and has been removed. All other lines are described in the legend.

Equation (12): Isn't that estimate affected by the compensation point concentration? page 354, lines 20-21: Isn't that a contradiction to the simplifications that lead to equa-

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tion (12)?

We are not sure we fully understand what this question is aiming at. The value of X_NH3(z0') depends on emission potential at the surface. It is here derived through a top down process from the measurements of air concentration and the net flux. This approach is valid independent of what regulates X_NH3 physiologically. In a bottom up approach, X_NH3(z0') could also be modelled from the compensation point (Personne et al., 2009). If the model parametrisation is correct, both estimates should agree.

Fig. 5: Particle radius and particle diameters are now mixed up in one single figure. Why don't you use just one of them?

See above.

page 357, line 2 and other spots in the manuscript: What do you mean with deposition rate? Is it deposition flux, or deposition velocity? If something else is meant, please specify!

We used deposition rate analogously to deposition velocity and have replaced it to make it clearer.

page 359, line 9: Please indicate which of the three proposed mechanisms you exactly mean with the notion nucleation. It seems that you mean No. 1, but the notion is also used in connection with the other two in the literature.

Nucleation is in our mind clearly defined. Nucleation is needed for both processes (i) and (ii). We have clarified this in the revised manuscript by (a) replacing 'formation of new particles' by 'formation of new particles through nucleation' and (b) referring explicitly to processes (i) and (ii).

p 343, line 23: delete but

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Done.

p 347, line 1: replace annual by annular

Done.

p 347, line 14: delete runs

Done.

page 350, line 23: Replace Q_c with Q_chi (using the greek letter chi)

Done.

page 355, line 10: Financial constraints ... Well, this is not an argument that should appear in a scientific journal contribution such as this one.

Changed.

page 356, line 13 and Fig. 6: Please say either Damkoehler number or Damkoehler ratio, but don't use a mixture of the two.

Unified to 'ratio'.

Fig. 7: Only one line can be seen. As one seems to overlay the other one, please indicate that in the Figure caption.

Since they are so close to each other, we have decided to omit this graph from the revised manuscript. Because the NH3 flux is still important background information it has been added as a third panel to Fig. 2.

page 358, line 5: replace micro m s-1 by mm s-1

Done.

Anonymous Referee #3

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Aerosol fluxes were recorded at 10 Hz for an instrument (TSI CPC 3760) with about a 1 L/min flow rate. How are these fluxes affected by the slow response of the instrument relative to the sampling rate? What portion of the flux could be lost in this situation for sampling at 2 m above the tall grass? Were the instruments still 2 m above the vegetation after it was cut? Does this change in displacement height and surface roughness change the fluxes and, if so, how will this affect the use of pre-cutting deposition velocities to represent surface removal during and after fertilization? The authors briefly address this on p.363, lines 18-21, but offer no bounds on its importance.

Similar setups to that used during this study have been used to measure surface atmosphere exchange above short vegetation before (e.g. Nemitz et al., 2002). The response time of the CPC used here is a little faster than 1 Hz. Dorsey (2002) investigated the average flux co-spectral density functions in some detail and found no appreciable flux loss, by comparing the gradient in the inertial sub-range with the expected -5/3 slope. On the basis of other measurements using instrument with similar response time in similar conditions, we conclude that the flux loss is likely to be $<$30%. We have added a statement to this extent to the manuscript.

Accurate correction for flux losses is difficult on a 30-minute basis: in our experience, simple empirical correction functions over-estimate the correction. In addition, corrections based on co-spectral analysis (e.g. shape of ogives) are uncertain for aerosol flux measurements because spectra tend to be noisy.

The measurement height remained the same above ground, before and after the cut. Before the cut, the measurement height above zero-plane displacement height (zm-d) was therefore somewhat lower than after the cut. While the lower measurement height above the tall grass canopy would have led to smaller and faster eddies (associated with larger loss of high frequency contributions), the higher surface roughness would have had the opposite effect. The fluxes show large variability in the fluxes (as demonstrated by the large standard deviations, compared with the means), which are typical

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for aerosol fluxes as alluded to by this Referee in their next comment. Thus the main uncertainty in Vd is likely not associated with flux losses, but with statistical uncertainties. As stated in the manuscript a difference in Vd of 1 mm s-1 translates into a change in the derived growth rate of 3 nm h-1. In response to this referee, we have added additional text to this statement: “From the range of results found in the other studies cited above, it appears that the potential range of deposition velocities lies within the range 0 to 2 mm s^{-1} for the size-range 11 to 200 nm, which dominated the number size-distribution that would have been detected during the experiment presented here. It follows that the error range in the growth rate lies between 0.18 to +6 mm hr^{-1}. In addition, the potential underestimation of the flux due to the limited frequency response of the particle counter, would have resulted in under-estimation of both the V_d to the unfertilized grassland and apparent emission fluxes above the fertilized grassland. Thus, the growth rates derived here appear to be conservative and are more likely an underthan an overestimate.”

Each sampling period was relatively short to fully characterize aerosol deposition velocities (about: 8 days over long grass, 7 days over short grass after it was cut, 11

days for short grass after fertilized); does each period have the same wind speeds,

stability, and relative humidity gradients? If not how could these differences affect the calculated production rates by changing the actual deposition from that estimated from the long grass period? (page 352, eq. 10).

We agree that the measurement database is too sparse to investigate the effects of parameters such as size-distribution, u* and atmospheric stability (the main drivers of aerosol deposition velocities) on the variability of Vd within each measurement period. As mentioned in a response to Referee 1 above, latent heat fluxes did not differ significantly before and after the cut. We have added average values of u* and H for the different measurement periods to Table 1, and discuss there influence in the revised manuscript: “Particle deposition velocities are known to increase with increasing sur-

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face roughness, increasing u_* and under unstable conditions (e.g. Wesley et al., 1985; Nemitz et al., 2004). The measurement periods were insufficiently long to derive robust relationships of V_d on these parameters from our measurements. However, values of u_* were very similar between the different measurement periods and H was similar during the later periods, but slightly reduced during the measurements above the long grass canopy, reflecting lower solar input (Table 1). In summary, the differences in deposition velocities are more likely linked to changes in surface roughness than meteorological differences."

p.350, line 26: Why can you assume stationarity and homogeneous conditions for simplifying the budget equation here ? The method is dependent on neglecting three terms in the RHS of eq(3) but there is no analysis presented that this is appropriate here. Were there no time changes /storage or advection? On p.354 (lines 16-19) it is stated that there were certain observed changes in regional air masses or regional chemistry in some of the observations. What is the sensitivity of the calculated production rates to these assumptions for eq. 3?

Eq. (3) is only used in this manuscript to introduce the source/sink term, in response to a Referee comment during the pre-submission stage, who found the introduction in the original manuscript to short. Eddy-covariance measurements are usually designed to minimise the effects of storage and advection by measuring over a large homogenous field away from point sources. While the different elements have not been quantified and studied in detail here, it is common practice in flux measurements of this kind to assume that they are small. A detailed quantification of these factors is not the subject of this study. In addition, the clear change from mainly deposition to consistent emission after fertilisation is linked to a process which changes abruptly the moment the fertiliser was applied. Storage and advection do not fulfil this condition.

p.351, line 6 Could not flux divergence also be related to the flow field besides just chemical production?

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This comes back to the same question. Here we have replaced 'flux divergence' with 'Q_N', which is really what we mean in this sentence.

p.351, line 21 The particle growth rate is calculated from theory and an assumed value of the sticking coefficient (p.352, line 6) which itself has a large degree of variation. How do the results depend on the assumed value of alpha = 0.5?

The derived growth rate at dp=11 nm is independent of alpha, but alpha affects beta and thus the calculation of the total mass flux (J_NH4NO3), but this is a secondary effect.

p.352-353 Does this method assume that the particle growth rate is not a function of diameter? If the surface area of advected aerosol varies with diameter over time will that affect your calculated growth rates?

The proposed method derives the growth rate at a single size (dp = 11 nm). The number of particles at 11 nm (dN/dr | r=5.5 nm) enters the calculation of the growth rate. This is derived from the size-distribution which was measured continuously. The growth is also size dependent as described by Eq. (6) and the growth rates quoted here only apply to 11 nm particles.

p.358, line 23-25 The paper states that the effect of non-zero water vapour fluxes can also cause artifacts but ought to be the similar before and after fertilization; but there is no support offered for this supposition. Were the soil moisture and ambient humidity profiles the same during both periods ? (the paper states that significant precipitation was reported only for June 6-7, just after fertilization). Were water vapor fluxes the same before and after fertilization?

See reply to Referee 1 above.

p.361, line 2, line 10/ Fig. 10 The concept of a maximum deposition velocity permitted

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by turbulence is mentioned and calculated without explanation or reference. Please define this term and show how it was calculated from these data.

A new Eq. (14) has been added and a short description of how Vmax was calculated was added to the manuscript, with reference to the companion paper of Nemitz et al. (2009a).

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