Detailed response to the reviewer's comments: Deposition velocity concept does not apply to fluxes of ambient aerosols

Rostislav Kouznetsov[1], Mikhail Sofiev[1], Andreas Uppstu[1], and Risto Hänninen[1]
[1]Finnish Meteorological Institute, Helsinki, Finland
Correspondence: Rostislav Kouznetsov (Rostislav.Kouznetsov@fmi.fi)

We thank both reviewers for their constructive comments and pointing out omissions and ways to improve our manuscript. In order to address the comments, in the revised manuscript we implemented the following changes:

- Performed a simulation of the single-column case with a set of finite relaxation times to explore the effect of the kinetics of gas-particle conversion for ammonium nitrate on the vertical profiles of concentrations and depositions of ammonia and nitrates.
- Added a section describing the recommended way of simulating deposition of semi-volatile species in regional and global-scale chemistry-transport models using gas-particle equilibrium in combination with aerosol dry deposition, without involving the concept of "Effective Vd".
- Added requested clarifications to figures and simulation descriptions.
- Addressed all other specific points from the reviewer's comments.

Below we re-iterate our responses to the reviewer's comments from the discussion listing the changes made to the manuscript and addressing the comments that have not been responded during the discussion. The reviewer's statements in *blue italic* and our response in plain text.

1 Detailed response to RC1

1.1 General impression

The authors revisit a problem that has been known for a while, i.e. that the dynamic gas-aerosol partitioning of between gas-phase ammonia and nitric acid one the one hand and particulate ammonium nitrate on the other causes fluxes that are non-conserved with height for the interacting compounds and particulate matter as a whole. I fully agree that most ambient flux measurements are affected by this process and cannot and should not be used to derive the deposition velocity of particles at the ground, even at only small contributions from ammonium nitrate. Whilst it is worthwhile reiterating this message which is widely ignored, partially because it is difficult to deal with, unfortunately, this paper neither provides a full review of the *subject nor does it add many new insights to the discussion already in the literature, provide a quantification of the impact of aerosol evaporation or offer a solution to the problem. The main new angle it adds, is framing the problem as a non-linearity between flux and concentration. This I agree has not been done before, but I am also not sure it helps (see below).*

The message of the paper is indeed that the ambient particle fluxes are affected by gas-particle conversion and should not be used to derive the deposition velocity of particles. We very much agree that this phenomenon is known - and that it has been widely ignored. The latter was the main motivation for this paper. However, we respectfully disagree with the statement that the paper does not show a feasible way to address the problem. To the opposite, the paper demonstrates that this problem has been solved in the SILAM model since more than a decade ago. The revised paper now has a dedicated section explaining the model components and setup.

The paper was not intended to be a literature review of the effects of aerosol evaporation on particle deposition, but, responding to the critical comments, we expanded it in the revised version. For details, we refer the readers to comparatively recent reviews of particle flux measurements and deposition velocities, either published as separate studies (e.g. Farmer et al., 2021) or as parts of other studies (e.g Pleim et al., 2022). We have also added references to detailed size- and chemistry- resolving modelling studies of the Speuld and other relevant datasets (Ryder, 2010; Van Oss et al., 1998; Katata et al., 2020).

As highlighted in the revised paper, our goal is to address the two orders of magnitude discrepancies between the aerosol deposition velocities derived from the first principles and those calculated from the field observations - and to suggest a practically applicable way to handle it in atmospheric composition models. The practical goal was to object against the concept of "effective deposition velocity" and attempts to derive such velocity from experimental plots like (see e.g. Fig. 4 of Pleim et al., 2022).

The paper demonstrates a much more accurate and physics- and chemistry- based approach. We demonstrate that a model with a mechanistic deposition scheme and gas-particle partitioning is capable of reproducing the main features of observed fluxes. The revised version now includes recommendations on simulating these processes in global and regional-scale chemistry-transport models, and comparing such models with flux observations (section "Recommendations").

In addition, there are some omissions, questionable assumptions and inconsistencies in the paper that would need to be addressed prior to publication.

The authors should take note of the study of (Ryder, 2010) which applied a more sophisticated model to the Speuld dataset, amongst others. I admit that this is unfortunately not very accessible via literature searches. Also not mentioned is the study by Van Oss et al. (1998)who also simulated the Speuld dataset. There is a lot of other literature not cited in the paper (see also below), including a relatively recent study over forest (Katata et al., 2020).

The references have been added.

Overall, I feel this paper is not publishable in its current form. It would either need to be significantly extended to become a more complete review of the subject or the analysis of the modelling would need to be expanded to provide more guidance of *how to deal with the problem. In either case the modelling would need to be redone incorporating a kinetic constraint on the attainment of the equilibrium (see below) and consideration of temperature gradients (if not already done).*

We have added simulations of the single-column case including kinetics. For that we implemented a parameter for temporal scale of gas-particle equilibrium into the model and also implemented a smoother lookup table for the ammonium-nitrate partitioning as a function of temperature and relative humidity. As one can expect, the flux profiles of semi-volatile particles were comparatively sensitive to the kinetic constant, but the bulk depositions for ammonium and nitrates were not. The results have been added to the subsection that discusses the kinetics.

The effect of temperature gradient has already been in the model, since the gas-particle equilibrium has been calculated at every model grid cell separately, thus accounting for the local temperature.

1.2 Major comments

1. I am not sure the modelling and measurement communities have different definitions for Vd. The modelling community uses the form F = (-) Vd x C to derive the flux from the modelled concentration, the measurement community uses the equation Vd = (-) F/C to derive Vd from flux measurements, often for use in models. This Vd is meant to be the same, that is usually the rationale for making the measurements. There are a number of reasons why the flux measured at a certain height above the surface does not represent the processes at the surface, such as advection, storage and, for non-inert compounds, chemical conversion. These errors need to be corrected for or shown to be negligible. The measurement community measuring fluxes of NH3, HNO3, NH4+ or NO3- is getting increasingly aware of this, but the community measuring particle number fluxes less so. I do not think that this is due to a difference in definition; it is a problem of awareness and lack of solutions to deal with the problem.

The naming convention has been rephrased. There are multiple observational papers, which plainly report "deposition velocities" (actually apparent ones), and there are numerous modelling papers that apply these values as V_d in Eq. (1), which is the root cause of discrepancy. This is now highlighted in the revised paper.

The second problem is that, strictly speaking, derivation of V_d from observations following Eq. (2), indeed widely used in the observations-based studies, is not statistically correct. If one wants to use simultaneous flux and concentrations observations to identify V_d as a parameter of the proportionality model in Eq. (1), there are well established procedures for that. One should make a regression, accounting for error bars, check if the specific linear model is suitable for the specific data set, etc. It should be made separately for each set of conditions. Such processing explicitly assumes proportionality as a basis for linear regression, and also takes care for small values, data uncertainty, etc. Unfortunately, we are not aware of any experimental paper where such a processing has been done for fluxes and concentrations of ambient particles.

Finally, we very much agree that fluxes and concentrations measured at some height do not necessarily represent the processes at the surface. In particular, it means that the reported velocity, even if derived correctly via regression, is not the deposition velocity and should not be used as such in the models.

2. The authors specifically comment on Nemitz et al. (2009) explicitly mentioning the problem, but still calculating Vd. This is no different in this paper which also deals with the problem and then calculates a deposition velocity (Fig. 2f), and refers to the measured deposition velocities as Vds (Fig. 1). This entity is explained in the caption, but (as a symbol) also has a clearly defined meaning from which this paper diverges. It is difficult to get away from these inconsistencies. Papers that are aware of the problem often use the term "apparent deposition velocity" or "local deposition velocity". I agree that maybe a different symbol is needed by the community to distinguish this from the Vd that describes the deposition process at the ground. Here the authors use subscript "a" as an option. What might be more important, however, is that the associated text correctly describes how to interpret the presented values of Vd and what they really represent.

Thank you! We tried to resolve the inconsistency, in particular, the axis title in Fig. 2f has been corrected.

3. The paper is not framed in the context of what the models are being used for. Total aerosol deposition is not normally a metric of interest. The main interest is in surface PM2.5 concentrations (for health impacts) and in total nitrogen deposition (for ecosystem impacts). For the former, deposition is considered a loss process for the PM2.5 (and/or its chemical components) from the atmosphere. For this purpose it is of secondary importance whether the removal of the ammonium nitrate is due to actual deposition to the surface or evaporation, and the concept of an "apparent" or "effective" deposition velocity may suffice and is arguably the appropriate loss term to use (except for the fact that the non-mechanistic treatment of the process then does not allow for tracking of the gas phase compounds and their potential to reform NH4NO3 further aloft). If the goal is to quantify nitrogen deposition, the evaporation matters and as does the fate of the ammonia and nitric acid. How much of it is deposited and how much is transported upwards (line 188) and how can this be estimated?

We agree that aerosol deposition itself may be not a metric of interest for some applications, but we disagree that it is an excuse to simulate it wrongly.

From a model development standpoint, surface $\mathrm{PM}_{2.5}$ concentration can be "tuned" to offset the error in V_d by tweaking other parts of the model. The orders-of-magnitude discrepancies between the V_d schemes mentioned in the first paragraph are often not evident from the surface PM skill scores in regional applications, as all AQ models that implement these schemes have been tuned to reproduce measured $\mathrm{PM}_{2.5}$ concentrations. However, reproducing PM concentrations through error compensation is not a correct approach. From practical standpoint, it fails at global scales, even if deposition is not the reported parameter: long-range transport is sensitive to dry deposition, and concentrations in remote areas will be gravely wrong if it is miscalculated.

Eutrophication and acidification problems, indeed, require the deposited amounts of nitrogen regardless of its gaseous or particular phase. An "effective" deposition velocity can be tuned to obtain roughly correct total deposition amounts at a specific site, for example in the Netherlands, but this approach would certainly break down under substantially different conditions, since the proportionality wrongly enforced by this concept does not hold for particles, as well as for gases in the presence of particles, as explained at the end of the revised Sec 4. The bi-directional fluxes of ammonia and its different deposition rate as compared to nitric acid add more complexity. To quantify deposition vs upward transport of ammonia and nitric acid, one would need detailed information about in-soil processes of ammonia (a.k.a. compensation point) and a detailed description of the behavior of deposited nitric acid in the soil.

In addition, accurate dry deposition of particles is important, e.g., for long-range transport of particular pollutants and deposition of hazardous substances in a particular form, such as radioactive fallout. We added a paragraph discussing the need for accurate dry-deposition schemes.

4. The paper needs to distinguish from the start between two related but different effects: the impact of aerosol evaporation on (a) bulk deposition fluxes, e.g. of nitrate mass or total PM mass, and (b) the impact on size segregated particle flux measurements measured over a specified size bin. The authors acknowledge the second effect in the paragraph starting in line 260, but ignore it in their attempt to reproduce the fluxes measured at Speuld (Fig. 1), which is impacted by exactly that effect as previously shown (Ryder, 2010). In this context, I am not convinced the (size-segregated) Speuld dataset can be reproduced with a bulk model. It needs a model that explicitly simulates the change in the particle size distribution.

We agree that details of the size-segregated dataset could not be reproduced with a bulk model, neither did we attempt to do that. We address the problem of particle deposition in large-scale chemistry-transport models that can neither afford detailed particle size information nor have sufficient input to constrain it. However, our simulations showed that bulk models can be strikingly good in catching the overall effect if aerosol evaporation is included. The temporal variability of the apparent deposition velocity $V_{d,a}$, shown in Fig. 1 of the paper and in Fig. 1 of (Gallagher et al., 1997), has been reproduced with a simple bulk model and instant gas-particle equilibrium. The corresponding discussion has been extended. The size-segregated fluxes and opposite-sign fluxes in different size bins have been discussed in Sec 5.3, where a reference to (Ryder, 2010) has been added.

5. The analysis in the paper assumes chemical equilibrium to be instantaneous, but concedes that this is probably not the case and later discuss potential kinetic constraints and time-scales. This is critical. If there were no constraint on evaporation then there would never be any NH4NO3 near the ground where concentrations of NH3 and HNO3 converge towards 0. Without a kinetic constraint the effect is overestimated. The authors omit to mention that kinetic constraints have been used in virtually all other model studies of this effect, either deploying rate constants (Kramm and Dlugi, 1994) or a relaxation time scale towards equilibrium (Brost et al., 1988; Van Oss et al., 1998; Nemitz and Sutton, 2004, e.g.) So why is this neither used here? At a time scale of 1000s evaporation likely starts to become unimportant given that transport timescales are smaller; e.g. see timescale analysis of Wolff et al. (2010) In fact, equilibrium is not usually maintained even at the measurement height with concentrations affected by conditions further aloft (e.g. Aan De Brugh et al., 2012)

We agree that the effects of finite time scale are worth considering. We have added a set of single-column simulations with finite equilibration times and a discussion to Sec. 5.2. The corresponding plots are included in the supplementary materials. The overall conclusion, however, appeared not so dramatic. The above-surface particle flux profiles are sensitive to the relaxation time and vertical discretization, but the bulk fluxes of ammonium and nitrate are not. This justifies using instant equilibrium in regional models, and reiterates the statement that particle fluxes above the surface are a rather poor constraint for deposition fluxes at the surface.

6. From a modelling perspective, the problem is that (a) aerosol evaporation is an unresolved subgrid process in atmospheric chemistry and transport models which do not have the vertical resolution to calculate the thermodynamic partitioning according to the vertical concentration and temperature (and RH) gradients near the ground and (b) these models assume aforementioned equilibrium. Maybe it should be stated in these terms.

This is rather a matter of resources and efficiency of implementation. One can explicitly account for kinetics of evaporation for vertical profiles inside and above the canopy in limited-scale simulations used for research, where computational costs are of secondary importance. However, applying the effective V_d concept for such particles would be wrong at any scale, regardless the model complexity. The revised paper stresses that, while details of in-canopy processes are too much to ask from regional models, evaporation of ammonium nitrate in the vicinity of the surface can well be reproduced with minimum, if any, additional costs. Capturing this effect would resolve most of the problem.

7. The paper identifies a problem, but does not offer a solution. If the deposition concept is not applicable, what should be used in its stead? For example, the EMEP model acknowledges aerosol evaporation as a non-resolved process within the model and uses a parameterisation of an "effective deposition velocity" for the constituents of ammonium and nitrate (Simpson et al., 2012; Eq. (68) and associated text). This has been developed by fit to nitrate flux measurements (apparent deposition velocity). This is a first approximation that should be improved with time. In terms of nitrogen deposition it implicitly makes the assumption that the ammonia and nitric acid that are liberated during evaporation are in fact deposited.

We respectfully disagree. SILAM is an operational model which has successfully implemented the solution without excessive costs. The gas-particle partition scheme for ammonium nitrate was published in 2000, whereas the dry deposition scheme was presented in 2012. Since then, this approach is default in all SILAM computations.

The "Recommendations" section has been added to the revised manuscript to summarize this very point.

8. The text suggests that the reduction in gas-phase concentrations towards the ground, caused by their deposition (and in particular of HNO3; line 166), is the main driver of evaporation. The model results of (Ryder, 2010) suggest that at Speuld this effect is only responsible for about half of the evaporation, the remainder being driven by increases in temperature (and equilibrium vapour pressures) near the ground and inside the canopy. In other words, even if NH3 and HNO3 did not deposit at all there would still be a driver for evaporation. Clearly, this varies over the day as the temperature gradients change. Are the temperature gradient accounted for in the model?

SILAM does account for the temperature and humidity dependence of $\mathrm{NH_4NO_3}$ saturation, but not for in-canopy temperature and humidity profiles. More detailed canopy-resolving models are needed for the latter. The plots in the supplementary material added to the revised version show the additional fluxes caused by this effect.

9. Whilst I fully agree that the evaporation complicates the relationship between the surface (and also the measured) flux and concentration, field observations are unlikely to show a linear relationship for a number of reasons. Vd is not a simple function of u (as implied by some of the parameterisations including that reflected in Fig. 4), but also atmospheric stability (as discussed in the paper), size distribution, surface roughness (which might change with footprint and wind direction) etc. In* *addition, measurement uncertainty complicates any relationship. As the paper correctly describes, the traditional deposition velocity concept relies on fluxes being constant with height and therefore does not apply to reactive compounds. Because Vd is effectively the slope between flux and concentration, it automatically follows that the relationship also becomes less meaningful and proportionality breaks down. I am not sure this is an independent observation / statement.*

Yes, this describes the situation very well. Indeed, there are various ways to handle the observation uncertainty, atmospheric stability, footprint differences, etc. However, if one wants to apply experimental results to constrain the function in Eq. (1), V_d should be determined as the slope valid only for those very specific conditions. As a by-product one can obtain the uncertainty of the slope and information on the applicability of V_d for the considered case. Otherwise, the resulting V_d is just a ratio of two weakly connected parameters, both having noticeable uncertainties.

If the proportionality is not reproducible in the experiments, enforcing the measured ratio of flux to concentration in a chemistry-transport model as an effective deposition velocity is obviously not a good choice.

This point has been added to the "Recommendations ..." section.

1.3 Other Scientific Comments

10. Eq. (1) – Even in the standard use of the deposition velocity, it is a function of height (because the Ra component changes with height), whilst the flux is constant with height. For clarity it might be worth indicating explicit height dependencies where they apply, i.e. F = Vd(z) / C(z).

Thank you! We have added the mention of height as a possible argument of the function Eq (1). We are not sure if it is worth indicating z as an argument of Eq (2), since in this case Vd depends on literally everything, including height, concentrations of other compounds etc., and this list is quite uncertain.

11. It doesn't seem right that in the model NH3 deposits faster than HNO3 (line 186) and the deposition parameterisation needs checking. The opposite should be the case as stated in line 219-220.

Thank you for spotting it! It was an issue in the model setup for the single-column case with outdated and no longer consistent routine activated for surface resistance of gases. The simulations have been re-run with the currently used EMEP-type surface resistance. The updated simulations show faster deposition for $\mathrm{HNO_3}$, and the imbalance between the two gases is much smaller but still visible. The plot have been updated and the text changed accordingly.

12. The Wesely et al. (1985) parameterisation was derived for sulfate which is non-volatile, so this probably not the best reference to back up the fact that the stability-dependence is due to evaporation. In fact it highlights that at least a component of this dependence is not due to evaporation, unless the sulfate measurements are somehow affected by the presence of nitrate or the changing particle size.

Thank you! The paragraph has been dropped. The Wesely et al. (1985) measured sulphates and got two orders of magnitude smaller deposition velocities than those reported by later outdoor studies with ambient particles, so those are consistent with the lab measurements. It would be interesting to explore why the parameter V_{ds} that has been already compensated for aerodynamic resistance still depends on thermal stratification, but it is beyond the scope of the manuscript.

13. Line 301 – How is this performance quantified? Often models are evaluated mainly for O3 and total PM2.5.

The paragraph has references to regular evaluations, which indeed are done mostly for the regulated pollutants. However, there were dedicated projects for evaluation of depositions and specific aerosol compounds. We added references to those as well.

14. Line 71. Whilst the data of Nemitz et al. (2002) could be made to agree with wind tunnel studies by tuning the parameters in the Slinn (1982) model, in hindsight it is likely that they were also affected by ammonium nitrate evaporation not explicitly accounted for in the analysis.

Yes. Though it is, probably, not too important for the introduction where we formulate the problem. Schemes with many fitting parameters indeed can be made to match data, see e.g. Pleim et al. (2022).

15. Fig 1 legend: the description of Vds as the "apparent deposition velocity compensated for aerodynamic resistance" does not seem right. Vd is height dependent and Vds is the surface value, more accurately at the aerodynamic roughness height, zo, i.e. Vds = Vd(zo).

We would disagree here. The plotted quantity is derived from local observations at a certain height, and it is derived exactly as the legend says. Obviously, the intention was to get something that is not height-dependent, but such a compensation implies a constant-flux assumption, which is clearly violated. We added clarifications on that.

16. Line 113. I am not familiar with the thermodynamic module used in SILAM and the references here do not shed light on it. The reference to Mozurkewich (1993) and discussion throughout the paper suggest that ammonium nitrate is treated as a pure compound, whilst in reality NH3 and HNO3 will form their individual vapor pressures above (usually aqueous) mixed aerosols. Whilst this will not change the qualitative results, ignoring the interaction with sulfate and chloride will over-estimate the vapor pressures. This simplification and its implications should at least be mentioned.

We have added the issue to the methods section. However, formation of sulphates and chlorides is included in the model, i.e., sulphuric acid consumes ammonia, and sea-salt consumes nitric acid.

17. Line 116. Maybe clarify that it is the upward flux FROM THE SURFACE (or emission) the model cannot represent. It can likely represent upward fluxes at higher heights.

Done.

18. Line 120. How closely did the model represent measured concentrations of individual compounds during the Speuld campaign? Some concentrations were published (Erisman et al., 1996; Wyers and Duyzer, 1997).

Thank you! The Erisman et al. (1997) paper ("Particle deposition...", mistakenly listed as 1996 in the review) has the data processed down to deposition velocities, without clear distinction between apparent and "real" one, so it is not straightforward to find a matching model variable. Another Erisman$*$ et al. (1997) paper ("The aerosol project...", Table 1 there) indeed lists the concentration measurements with few references. One of them, the TNO report by Duyzer (1994) is not available. Another reference (Wyers and Duyzer, 1997) has some typical values for observations for $\mathrm{SO_4}$ and $\mathrm{NO_3}$, one per campaign. For the considered June campaign Table 1 there gives $\mathrm{SO_4} = 5(3-10)\,\mathrm{\mu g\,m^{-3}}$, and $\mathrm{NO_3} = 5(1-7)\,\mathrm{\mu g\,m^{-3}}$. These values are consistent with our model simulations.

19. Line 156. This sounds like the conditions favoured active production whilst they actually favoured evaporation. Maybe rephrase "high enough for particulate NH4NO3 to be present".

Done.

20. Line 186. Several studies have found apparent reduced deposition of HNO3 (in fact this observation prompted some of the early investigations in the subject). The more extreme observation of upward flux of HNO3 is likely also the impact of evaporation being instantaneous in this particular model.

In this case it is a bug in the setup that has been fixed. "apparent reduced deposition of HNO3" would be properly named as "reduced downwards flux".

1.4 Technical Corrections

- *Throughout the paper there are spaces lacking between numbers and their units as well as different units, e.g. 1ms-1 should read 1 m s-1; the meaning is different.*

Fixed and homogenised.

- *Units for Vd vary between mm/s, cm s-1 and 10-2 m s-1*

Fixed. All Vd should be expressed in $\mathrm{cm\,s^{-1}}$

- *Line 32. Missing "of" in "regardless of the presence"*

Fixed

Line 89 should probably better read "The role of ammonium nitrate formation in modifying particle fluxes has been studied . . . "

Fixed

- *Fig. 1 – the style of the units and parentheses in the y-axis labels varies across subplots.*

Fixed

2 Detailed response to RC2

2.1 Overall evaluation

The authors have done a good job formulating, demonstrating and discussing the issue, but unfortunately there is rather little practical outcome and recommendations on how to deal with the identified problems (given the paper estimates quite an abundance of ammonium nitrate in Europe and indicates its potentially frequent presence in PM2.5?

Could the authors suggest better solutions for the application of Vd in the case of semi-volatile (ammonium nitrate) aerosols i.e. whether and how the "apparent Vd" from field measurements could be used in a sound way? Alternatively, could a model representation of the apparent Vd be proposed? Actually the paper anticipates that the authors would (L. 105) "..suggest an approach to bridge the gap between observed apparent deposition velocities and deposition parametrisations based on Eq. (1)".

The "Recommendations" section has been added.

In practice, we recommend using deposition schemes designed for passive particles only for passive particles. For semi-volatile particles, the particle deposition scheme should be applied in combination with gas–particle partitioning. This approach reproduces observed fluxes without invoking any apparent or 'effective' Vd.

It would also be advisable to show the effect of accounting for gas-aerosol partitioning during aerosol deposition on model results on a regional (European) scale. What is the seasonality of this effect? Does it help to improve the model performance.

The gas–aerosol partitioning plays a major role for semi-volatile species, but it is hardly possible to isolate the effect of partitioning during deposition from other processes occurring throughout the aerosols/precursors life cycle (emissions, chemical transformation, and transport). A simulation with partitioning entirely turned off can be done with moderate effort. However, the results would have no meaning because the formation of ammonium nitrate is the result of the very same partitioning process that is responsible for its disintegration near the surface. So, in the simulation with partitioning off, there will be no ammonium nitrate in aerosols at all.

2.2 Detailed comments

L. 96 - "The goal… explore the discrepancy between different experimental studies of Vd" sounds like exaggeration.

After looking at the figures in (Pleim et al., 2022), especially Fig. 4 or Fig. 6 there that show the scatter of two orders of magnitude, the statement does not seem exaggeration.

L. 120-124 - was emissions for 1993 used for these simulations? If not, please comment on the uncertainties

It was 2007 emissions from TNO, the reference to the corresponding report added. Despite the differences in annual total emissions of Netherlands between 1993 and 2007 are some -25% in NOx and -50% in NH_3 (Staats et al., 2025) are quite notable, we would argue that summertime ammonia emission reduction should be smaller since substantial fraction of the reduction is due to the change in spring-time manure application. Since we are mostly interested in qualitative behaviour of particle fluxes, we do not consider the remaining difference important. If we performed the detailed evaluation of the simulation against time series of hourly or daily concentrations, more consistent setup would be needed.

A note added.

L. 135 - what season this simulation was representative of?’

Summer. The simulation uses the ERA5 meteorology starting on 2005-06-01. No specific reason for that date. A note added to the text.

Fig. 1 (b) and L. 138 - is that so that different diameters of NH4NO3 were assumed in the simulations in sec. 3 and 4: 0.3 um and 0.7 um? Any particular reason? Does SILAM model simulate size resolved Vd?

Indeed the sizes differ. The simulations for Speuld were made to match the measured size range. Currently the only mode used for ammonium nitrate in regular simulations is 0.7 um. The difference should be insignificant, since the settling and deposition velocity for these sizes are very low, kinetics is prescribed, and optical properties are not considered.

SILAM assigns V_d depending on particle properties, considering aerodynamic size, geometrical size and mobility size separately (see Kouznetsov and Sofiev, 2012).

In the revised version a note has been added.

Fig. 1 (a, b) - I assume that 0.3 um for NH4NO3 from SILAM’s was assumed in order to correspond with particle sizes of 0.1-0.4um in deposition timeseries from Gallagher et. al (1997). The latter is actually for particle number, where the smallest particles are said to probably be sulphate (and the smallest particles determine Vd due to their large number) - does that inconsistency matter for your findings/conclusions?

Due to a huge difference between deposition rates of gases and particles, the flux is determined by the size that is actually transported due to evaporation at the surface, rather than by the size that is most abundant at the observation height. The basic conclusion is that the particle flux is modulated by the presence of ammonium nitrate. It would not change.

What height the SILAM results in Fig. 1 are representative of?

As the top of the lowermost layer is 25 meters, it should approximately match both observation heights, that actually show very similar fluxes.

A clarifying sentence added.

6. L. 284 (Fig. 4) - Do you have model results showing that the PM in the lower cluster does not contain any ammonium nitrate indeed?

Yes. The figure has been changed so that different markers are used for cases with and without ammonium nitrate.

2.3 Minor comments

L. 3 - " A brief overview" instead of We analyse

The sentence rephrased.

L. 39 - define Vd,a

Done.

L. 162 - why not NH3?

Yes. Indeed both of them. Corrected.

L. 186 and 220 - explain the different (contrary) assumptions with respect to deposition efficiency?

A typo in the model setup for the single-column case. Fixed in the revised version.

3 Concluding remarks

In the end we would like to thank both reviewers for a good job on reading the manuscript and making their valuable comments that helped to make the manuscript more sound and consistent.

We hope that the revised manuscript can be accepted for the publication in GMD.

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