Appendix RIVM report 2025-0081

In the first part of this appendix, the sensitivity of the Operational Priority Substances model (OPS) to individual parameters which were perturbed in the main study is discussed in somewhat more detail. Besides a somewhat more detailed discussion on the five most influential parameters already highlighted in the main report, a discussion on the sensitivity to the other seven parameters is included as well.

To perform the sensitivity analyses, code adjustments were needed in OPS. The second part of this appendix discusses these adjustments in more detail.

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Contents

Introduction

In the study "Uncertainty in calculated nitrogen deposition from individual sources", a sensitivity analysis with the so-called long term version of the OPS (Operational Priority Substances) model is performed. This model version will henceforth simply be referred to as OPS. Model documentation can be found in Sauter et al. (2023).
The aim of the study is to increase insight in the model-specific uncertainty in the annual nitrogen deposition caused by a single source, up to several hundreds of kilometres. The present appendix provides additional background information on the sensitivity study.
As a part of the study a one-by-one sensitivity study was performed. A brief analysis of the impacts of the five most influential parameters on the OPS results was already presented in the main text. In Chapter 1 of the present appendix, the sensitivities of OPS to each of the perturbed parameters OPS are analyzed in somewhat more detail. This is done for the various source types considered in the study, i.e., NH_3 emission from a livestock farm and NO_x emissions from an industrial stack and a motorway, respectively. The latter represents a line source, which is modelled as an array of point sources which are henceforth called source segments. See main text for more details.
For each source type, the parameters discussed here were perturbed according to the methodology explained in Chapter 3 of the main report. Because the uncertainty distributions of the parameters differ, the perturbation factors applied in the one-by-one sensitivity study also differ among the parameters. The applied perturbation factors are indicated in the figures presented here.

For the sensitivity analyses performed in the study, several adjustments in OPS were needed. The adjustments made for the present study are described in Chapter 2.
References to figures and text parts will explicitly mention if these are to the main report. References to figures and text part in the present appendix will simply follow the numbering used here.

Parameter symbols:

Plume rise (livestock farm and industrial stack); source height (motorway)	- Δh
Vertical dispersion length	- σ_z
Mixing layer height (boundary layer height)	- z_i
Chemical conversion rate	- k_{chem}
Dry deposition velocity of primary compounds	- $v_{d,pri}$
Dry deposition velocity of secondary compounds	- $v_{d,sec}$
Washout scavenging rate, primary compounds	- $\Lambda_{w,pri}$
Washout scavenging rate, secondary compounds	- $\Lambda_{w,sec}$
Rainout scavenging rate, primary compounds	- $\Lambda_{r,pri}$
Rainout scavenging rate, secondary compounds	- $\Lambda_{r,sec}$
Meteorological conditions	- Meteo
Diurnal variation	- *dv*

1 Sensitivity to individual parameters

This appendix discusses results of a set of sensitivity studies based on perturbation of single, individual parameters (one-at-a-time sensitivity analysis, see Chapter 3 in the main text for a description of the method). The purpose is to gain an understanding of the model response to specific parameters. This may facilitate interpretation of the results based on the Monte Carlo approach (Sections 4.1-4.3 in the main text).
Here, we examine the model behavior as a function of distance. At a specific distance, responses of the model obtained by averaging the responses over the 12 spokes are shown (see main text, Section 3.4, Figure 7). Although averages from the 12 spokes are given as a function of distance, these results do actually represent variability among the spokes, and specific features along them. Examples are the spatial variability of k_{chem} along the spokes and between them (impact on concentration), variability in source depletion due to varying land use and roughness length and background concentrations (impact on dry deposition), and variability of meteorological conditions, like precipitation characteristics (impact on wet deposition). Not only will such variability lead to differences among the spokes, but it will impact results over a specific trajectory of a singular spoke as well. This may sometimes result in 'spikey' patterns of the relative difference, for instance in the case of dry deposition. Here, we focus on the more general trends in the patterns.
In the next Sections, results per parameter will be discussed. Each of the following Sections is started with a brief summary of the role of a process of parameter in the calculations. This is followed by a discussion of results for concentration of individual primary and secondary compounds, i.e., NH_3 and NH_4^+, and NO_x and NO_3^-+HNO_3. Furthermore, sensitivities of dry and wet deposition of NH_x and NO_y, respectively, are presented. The sum of wet and dry deposition of these compounds is the total deposition, of which the sensitivity is discussed in the final Section.

1.1 Plume rise Δh

The effect of a change in plume rise or source height (both indicated with Δh) is related to a combination of the following phenomena:

- The stack height (emission height) plus plume rise, henceforth simply referred to as "plume height", determines along with the atmospheric conditions where the plume touches the Earth's surface. Beyond that point, the concentration at the surface exceeds 0 µg/m3. This is also the distance at which dry deposition starts.
- Upon emission, the plume height may exceed zi. No, or only part, of the pollution is emitted into the mixing layer in that case. The fraction of pollutant assumed to be immediately incorporated into the mixing layer is based on the difference between the plume height and zi. At greater distances, part of the pollutant mass originally emitted above the mixing layer may be entrained into the mixing layer again, due to boundary layer growth. In either

case, if the emission occurs above zi, OPS sets the emission height of the fraction incorporated in the mixing layer to zi
- Some meteorological parameters used in the model can vary (slightly) due to the turning of the wind with height. Δh influences the height at which these parameters are evaluated, and hence their value. Wind direction and hence turning of the wind with height determines the transport direction of the plume and its actual location. These in their turn affect the classification in terms of wind sector, and interpolation of meteorological parameters between wind sectors. This also influences the frequency of occurrence of a particular meteorological class. Important parameters affected are, e.g., windspeed at 10 m, boundary layer height, the length and intensity of rain events and friction velocity.
- The calculation of σ_z (depth of the plume) and the wind speed depends on the plume height and hence on Δh.
- The ratio of the plume height and zi determines which parameterisation scheme (surface layer, convective or neutral boundary layer above the surface layer, interpolation between these) is used to compute σ_z and wind speed at the plume height.
- Wet deposition is affected in various ways. The impact of Δh on the effective windspeed at the effective transport height influences the travel time. This affects the average duration of a rainfall event and hence rainfall probability, such that increased wind speed, due to increased Δh, increases wet deposition, and vice versa. Notice that the rainfall probability is also influenced by the possibly altered wind direction (see 3rd bullet). Furthermore, the weighing between washout (most relevant nearby the source) and rainout (most relevant at greater distances) depends, amongst other things, on the distance between the plume height and zi.

1.1.1 Concentration – impact of Δh

For the livestock farm, relative effects of Δh on concentration (Figure A1) are quite small, up to 2% between 2 and 10 km, reducing to only a few tenths of a percent beyond about 50 km for NH_3. Effects on NH_4^+ concentration remain within about 5% and are largest within a few kilometres from the source. By contrast, the effects of Δh in the case of NO_x emissions from the industrial stack are much larger, ~ –35% to almost +50% for NO_x and ~ –50% to +100% for HNO_3+NO_3^-, at about 20 to 50 km from the source. In the case of the motorway, the maximum NO_x concentration changes range between –10% and +5% at 1 km from the source and between –10% and +8% at a few kilometres from the source for HNO_3+NO_3^-.

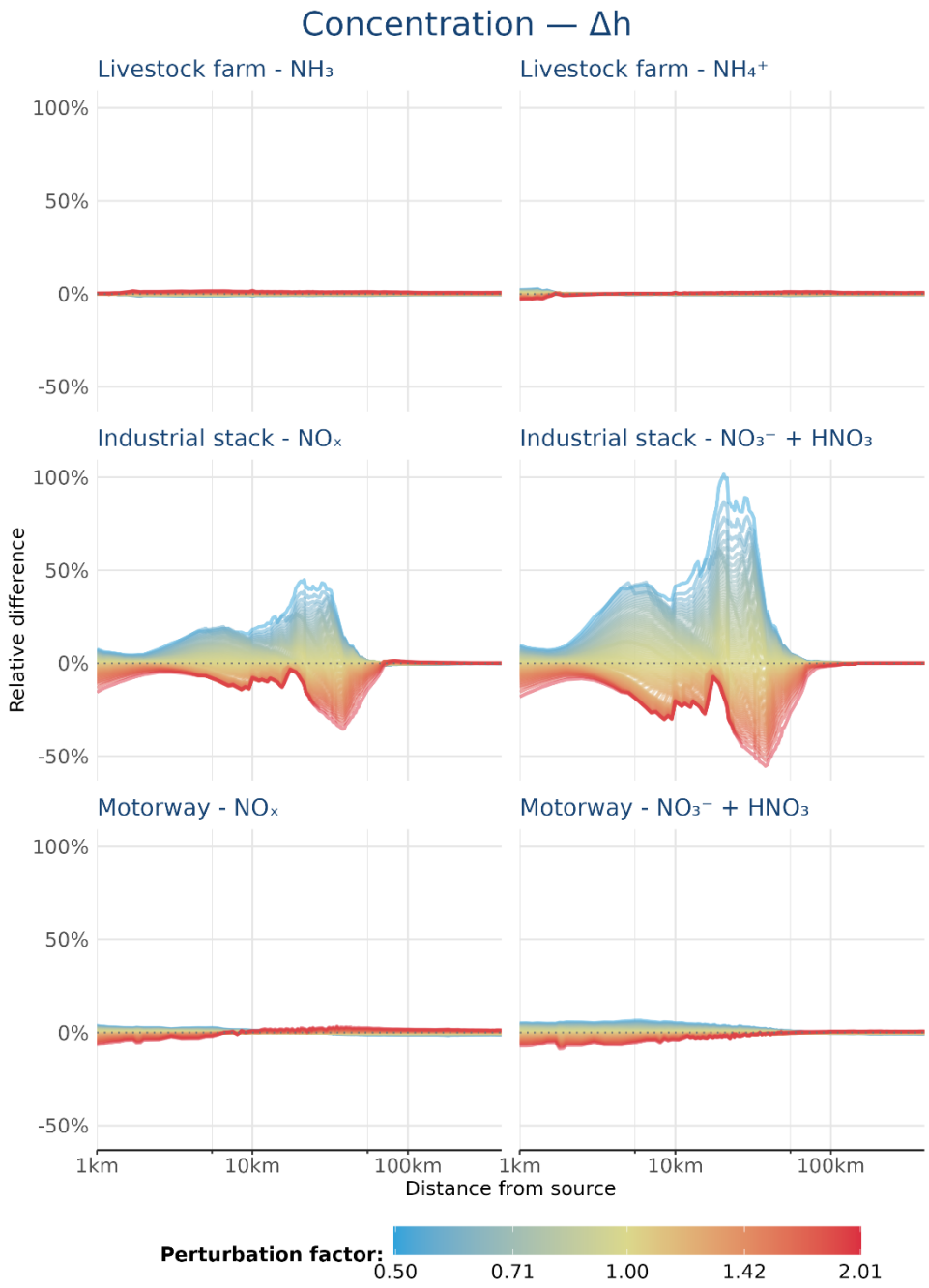

Figure A1 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying the plume rise (livestock farm and industrial stack) or emission height (motorway), both called Δh. Top row: livestock farm NH_3 and NH_4^+; middle row: industrial stack NO_x and NO_3^- +HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

Generally, an increase of Δh in the case of the livestock farm leads to a somewhat increased concentration, and vice versa. With increased Δh, a plume reaches the ground at a greater distance from the source. Where this happens the concentrations are somewhat higher than in the reference case, because there has been less deposition over the

trajectory up to that point. The increase in concentration levels off slightly at greater distances.
Higher concentrations near the source for decreased Δh occur if the plume axis is closer to the receptor. At small distances, this effect dominates over the increased deposition over the trajectory. Enhanced deposition leads to a crossover point because concentrations are lower as compared to the reference run. With increased Δh, for NH_3 this crossover point has already occurred at distances smaller than 1 km (not shown), for NH_4^+ it occurs just after 1 km.
The relative change nearby the source is somewhat larger for ammonium than for ammonia. NH_4 is the formed secondary aerosol and concentrations at close distances are still low. Hence, the impact of changed NH_3 concentrations on chemical production of NH_4^+ is comparatively strong. The crossover point is seen at greater distances compared to NH_3. This is likely caused by the less efficient dry deposition of NH_4^+ compared to that of NH_3. This renders the source depletion less efficient and it takes longer to compensate the effect of plume rise on concentration.
For the livestock farm, most of the impact of Δh on the model results appear to be related to the changed distance at which the plume reaches surface and to the changes in transport speed of the plume. The other points mentioned at the start of Section 1.1 are less relevant due to the low source height. The low source causes emitted material to remain inside the mixing layer, and turning of the wind has hardly any impact. An analysis of the underlying components of the model results indicated that the impact of Δh on σ_z is also minimal in this case (not shown).
For the industrial stack, the sensitivity to Δh is considerably larger than for the livestock farm case and the motorway. Variations with distance are quite strong up to about 50 km. Slightly increased concentrations for increased Δh are obtained after about 50 km. Especially for high sources and for high Δh, plume rise impacts many variables in OPS. Due to the many non-linear interactions, it is impossible to pinpoint the exact cause of variability in the sensitivity to plume rise. Yet, an analysis of the main patterns is attempted here.
Up to about 50 km from the source concentrations tend to decrease with increasing Δh and vice versa. This is likely due to a combination of higher plumes not touching the ground yet, and part of the emission being released above the boundary layer. Entrainment of the pollutant into the boundary layer at some greater distance can cause an interruption in the decrease of concentrations. If plumes touch the ground at greater distances from the ground, concentrations with increased plume rise may be higher compared to the reference run, because there has been less deposition over the trajectory. This is also the case if they become well mixed at greater distances. The interaction between deposition over the trajectory and the concentration causes a crossover point. Beyond this point, maximum differences with the reference run are at first about one percent, but become nearly negligible when approaching 400 km. For the industrial stack the crossover point of found at distances much greater that for the livestock farm. This is probably because plumes from the industrial stack are higher and differences in Δh are stronger than for the livestock farm. Furthermore, dry deposition of NO_x is less efficient than that of NH_3.

The sensitivity to plume rise is somewhat larger for the sum of NO_3^- and HNO_3 than for NO_x. Similar to NH_3 from the livestock farm, the explanation is, that both NO_3^- and HNO_3 are formed by chemical conversion in the atmosphere, so that their concentrations are still low near the source.
For the motorway, in general a similar impact is seen as for the livestock farm. However, the crossover point is reached at greater distances from the source due to the less efficient dry deposition of NO_x compared to that of NH_3. The impact is more evenly distributed over the distances because of the contribution from multiple source segments with varying distance to the receptor. For NO_3^-+HNO_3 the impact is again somewhat larger than for the primary compounds. Also, the crossover point is at a greater distance than for NO_x, presumably because of the less efficient source depletion of NO_3^- and HNO_3.

1.1.2 Dry deposition – impact of Δh

Impacts of Δh on dry deposition are shown in Figure A2. For the livestock farm case the sensitivity to Δh is rather small, with relative dry deposition differences between –1% and 2% nearby the source, and only less than a percent beyond about 50 km. The relative difference of the dry deposition of NH_x is the combined effect of the relative differences for NH_3 and NH_4^+, respectively, which tend to follow their concentration (see 1.1.1). Close to the source, the variation of NH_3 dominates the effect on NH_x because NH_3 is more abundant than NH_4^+. With increasing distance the contribution from NH_4^+ is expected to increase, but this is hardly visible. The impact of Δh is of similar sign for both compounds far from the source.
For the industrial case the sensitivity of dry deposition to Δh is also affected by the aforementioned modelled interaction between Δh, plume height and transport characteristics, as influenced by meteorological conditions. In spite of such complex interactions, the dry deposition of NO_x and of the sum of NO_3^- and HNO_3 generally also follow the concentration patterns of these compounds (see 1.1.1). However, the relative difference in dry deposition of NO_y is smaller than for concentration, with peaks varying between 10% to 25% (also for the opposite sign) up to about 50 km, decreasing again with reversed sign toward negligible impacts further away.
For the motorway, the relative impact of Δh on the dry deposition of the individual compounds also follows the patterns of the relative concentration differences (see 1.1.1). Hence, the dry deposition of the total NO_y follows the pattern of the primary material nearby the source, while the impact of dry deposition of the secondary material becomes more important at greater distances. This also explains that the crossover point is now found between the crossover points for NO_x and for NO_3^- + HNO_3, respectively (Figure A1). The impact is highest near the source, ranging between about –7% and 4%, and reduces to between about plus and minus 2% at 400 km.

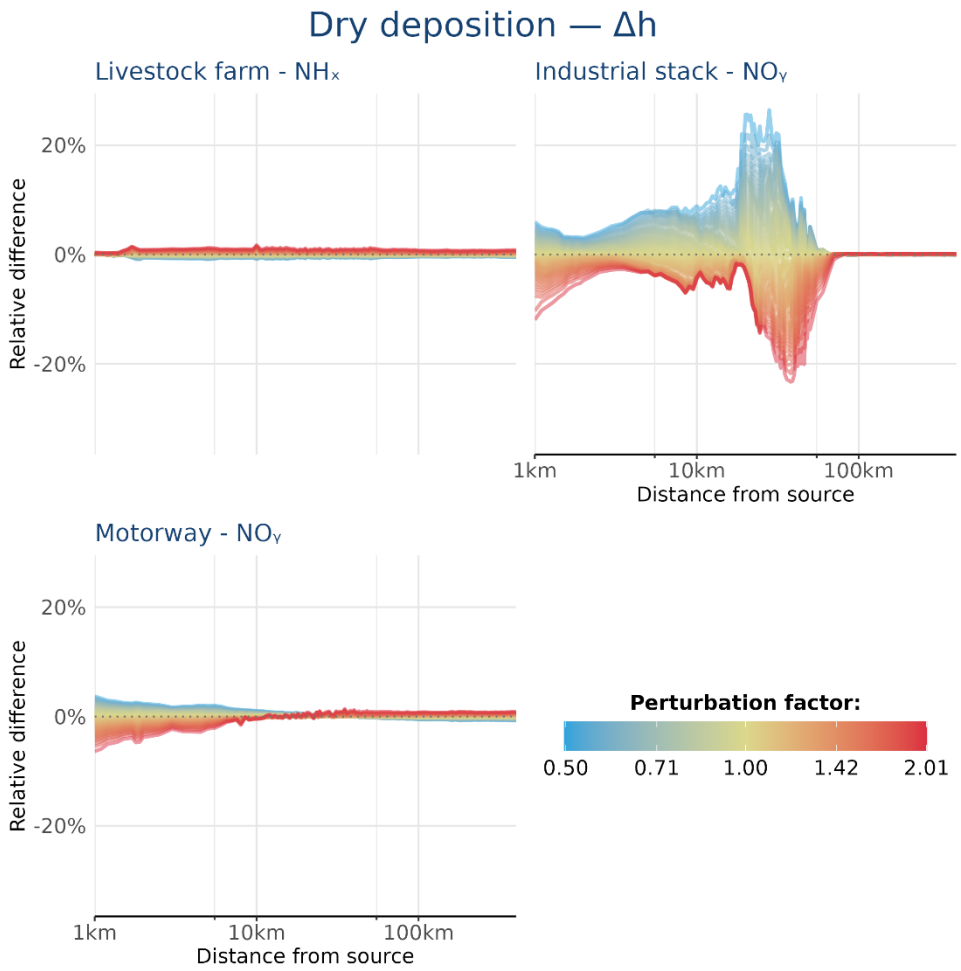

Figure A2 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying the plume rise (livestock farm and industrial stack) or emission height (motorway) Δh. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.1.3 *Wet deposition – impact of Δh*

Relative differences in wet deposition of NH_x and NO_y are shown in Figure A3. For the livestock farm they range between –1% and 2%. In the case of the industrial stack they vary from almost –20% up to about 8% for distances up to ~50 km. Beyond that distance, differences decrease to near-zero. For the motorway, the relative differences in wet deposition of NO_y remain within about ± 4%. Maximum differences are found around a distance of 10 km from the source.

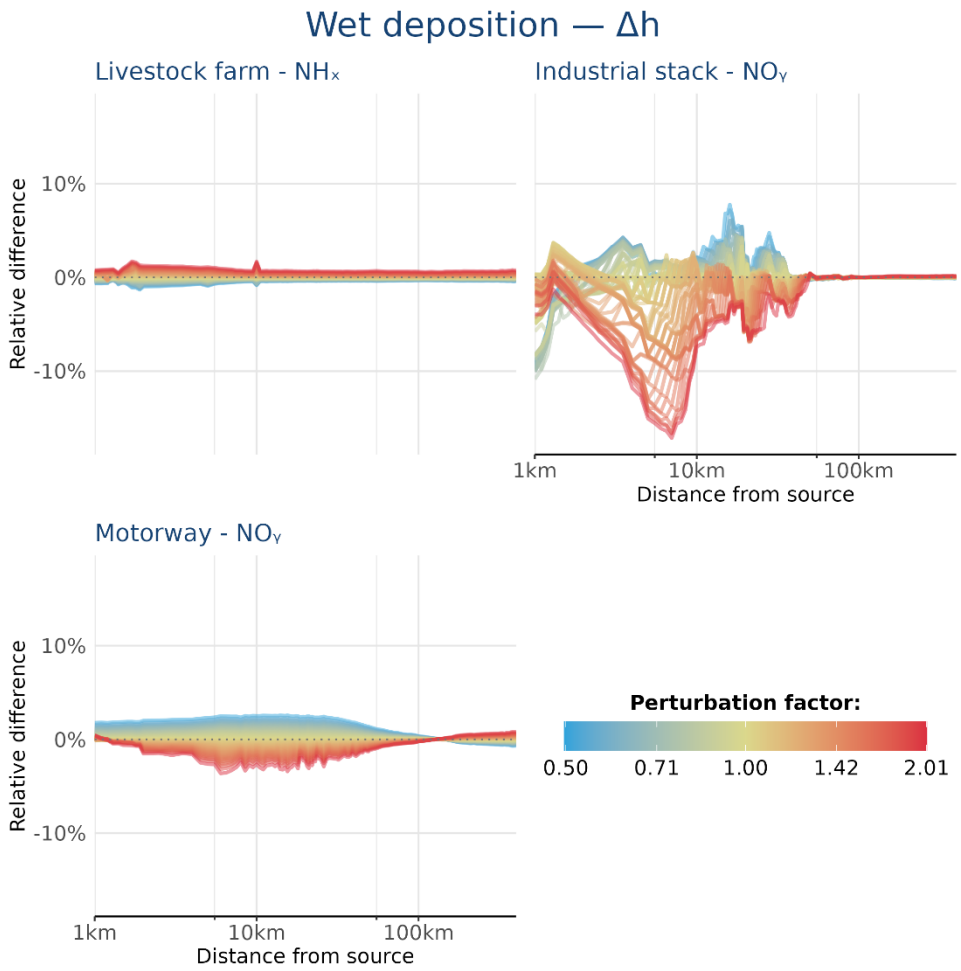

Figure A3 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying the plume rise (livestock farm and industrial stack) or emission height (motorway) Δh. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

The relative difference in wet deposition of NH_3 from the livestock farm generally increases with increasing Δh and vice versa. Differences more or less follow the concentration differences of the individual compounds after 2 km. The impact of Δh is expected to be less for wet deposition than for concentration at the receptor height. This is because wet deposition is related more to the concentration over the entire column than to the concentration at a specific height. For NH_4^+ results are rather different. Upon increasing Δh, an increased wet deposition is obtained up to about 2 km from the source, followed by a decrease between about 2 and 30 km, and by an increase again after about 30 km. The opposite pattern emerges when decreasing Δh. The difference between patters for NH_3 and NH_4^+ might be related to differences in scavenging ratio. The efficiency of washout depends on the rainfall intensity. The scavenging ratio for NH_3 is larger than for NH_4^+ when rain intensity is below 1.1 mm/h, but it is larger for NH_4^+ than for NH_3 when rain intensity exceeds 1.1 mm/h. For rainout, NH_4^+ has a higher scavenging ratio ($1.4 \cdot 10^7$ compared to $1.4 \cdot 10^6$ for NH_3). Since the relative contribution of washout

and rainout to wet deposition changes over distance, the relative efficiency of either compound may also change with distance.
The impact of varying Δh on the wet deposition of NO_y for the industrial stack shows more switches between positive and negative effects, than the impact on dry deposition and concentration. This could be related to different interpolation factors between wind sectors, which result in varying length and intensity of rainfall events for the separate OPS runs. This is confirmed by studying the underlying component model results in Easterly direction (not shown). For other receptor spokes the meteorological conditions will also differ between runs with differing Δh, which may lead to a more 'random' variation in sensitivity with distance. When averaging over the spokes, the sensitivity pattern is not as straightforward.
Differences in wet deposition for NO_y for the motorway increase for both in- and decreased Δh, just around 1 km. Just after 1 km, a crossover point is seen for increased Δh only, resulting in relatively low wet deposition up to 100 km from the source. Here, another crossover point is seen in the case of increases as well as decreasing Δh. Beyond this point wet deposition of NO_y increases with increasing Δh and vice versa. The patterns are the result of the added contribution of the individual compounds. The relative difference in wet deposition of NO_3^-+HNO_3 (not shown) generally follows the NO_3^-+HNO_3 concentration pattern depicted in Figure A1. However, wet deposition of NO_x (not shown) does not follow the NO_x concentration pattern but rather gives an opposite sensitivity. Wet deposition of NO_x increases with increasing Δh and vice versa, with a stronger sensitivity for increased Δh. This is because washout is neglected for NO_x in OPS and a change in wet deposition is dominated by a change in rainout. With increased Δh, the contribution from rainout is increased, and vice versa. Together with the neglected washout this puts a limit to the negative sensitivity.
The NO_y wet deposition near the source is dominated by the contribution of NO_x. The non-symmetrical impact of Δh on wet NO_x deposition combined with the pattern of wet deposition of NO_3^-+HNO_3 yields the non-linear result near the source, seen in Figure A3 for the motorway. At greater distances, the impact of NO_3^-+HNO_3 becomes more apparent.

1.1.4 Total deposition – impact due to plume rise Δh

For the livestock farm the relative differences in total NH_x deposition (sum of the dry and wet deposition) due to changes in Δh resemble those of the dry deposition nearby the source (Figure A4) . Here, the contribution from the sensitivity to dry deposition is dominant. Further away, wet deposition becomes more relevant. Since the sign of the variation is the same for both processes, a similar impact is seen. In general, larger total deposition is seen with increased Δh and vice versa. The sensitivity to Δh remains small: up to ~ ±1.5% between 1 and 10 km and less at greater distances.
For the industrial stack, the relative differences of total deposition of NO_y is more irregular than for dry deposition, but it is not as 'chaotic' as for the wet deposition. Differences in total deposition resemble the ones dry deposition most, especially in the first few kilometres. Largest sensitivities to Δh are found at distances less than about 50 km, with peaks varying between maximum –17% and 19%. Between 50 and ~70 km the sensitivity decreases and becomes negligible beyond 70 km.

Up to 50 km lower total deposition is found with increased Δh compared to the reference run, and vice versa. This is likely caused by higher plumes resulting in lower concentrations at the surface and hence less dry deposition, and vice versa. This switches sign after about 70 km, where more plumes are touching the ground. Concentrations are then higher because less material has deposited over the trajectory when Δh increases. As a result, compared with the reference run, more dry deposition is obtained. The interruptions in the decreased total depositions are partly due to entrainment of pollutants initially emitted above the boundary layer. The entrainment is caused by a growing boundary layer, and may suddenly lead to higher concentrations and hence higher dry deposition at a certain distance. Also, the more variable sensitivity to wet deposition influences these patterns (see 1.1.3).
The relative differences in the case of the motorway are also dominated by the changes in dry deposition nearby the source. The contribution of wet deposition increases with distance. As such, the crossover point moves a bit towards a location between the crossover points for dry and wet deposition. The impact is largest near the source, with decreased total deposition of NO_y for increased Δh (down to –6%) and increased total deposition for decreased Δh (~ 4%). The crossover point is located just before 100 km; variations are about 1% at 400 km.

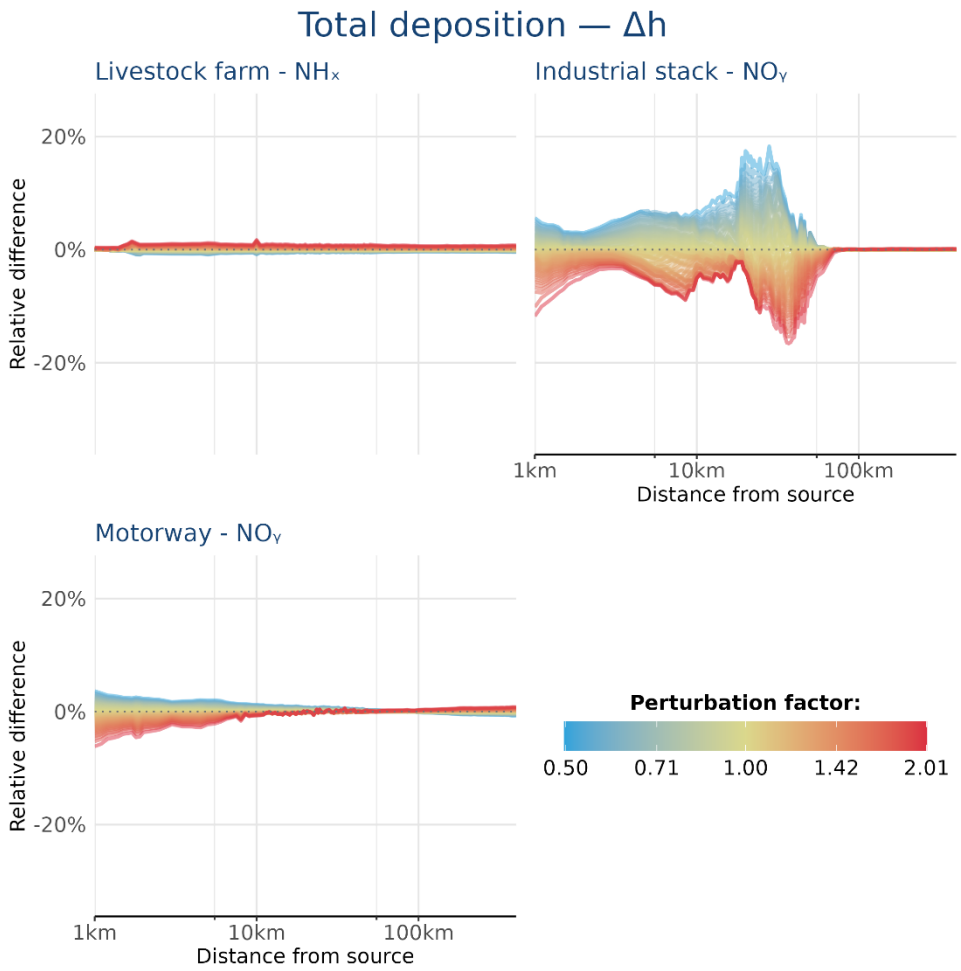

Figure A4 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying the plume rise (livestock farm and industrial stack) or emission height (motorway) Δh. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.2 Vertical dispersion length σ_z

A change in vertical dispersion length, σ_z, has several effects:

- Increasing σ_z gives deeper plumes. This results in reflection of the plumes at the surface and top of the boundary layer at shorter distances, compared to the reference run. The reverse is true for decreasing σ_z.
- The plume depth affects dilution of the plume and hence impacts the concentration directly. Depending on the receptor location this can result in higher or lower concentrations:
  - Increased σ_z and hence, a deeper plume, can lead to higher concentrations if for the reference run the receptor was outside the plume while it is inside the plume for increased σ_z. However, if the receptor was already near or at the plume axis, a deeper plume will reduce concentration because it implies more dilution.
  - Reduced σ_z and hence a shallower plume can lead to lower concentrations if the receptor height at a given distance is no longer reached, or is located more towards the edge of the

plume. Higher concentrations will be found if the receptor was already near or at the center of the plume, since reduced σ_z implies less dilution.

- σ_z, combined with z_i, also impacts the so-called phase of the plume which OPS distinguishes: not yet well-mixed (Phase 2) versus well-mixed (Phase 3), respectively. The distance at which Phase 2 transitions to Phase 3 decreases with increasing σ_z. This affects source depletion because the deposition velocity is larger in Phase 2 than in Phase 3.
- The distribution between washout (below cloud) and rainout (in cloud) directly depends on σ_z. The relative contribution from rainout increases faster with distance when σ_z is increased, and vice versa. This impacts the wet deposition, since wet deposition by rainout is larger than by washout.
- The total loss due to wet deposition also depends on the travel time, which is inversely proportional to the wind speed at the height of the plume axis averaged over the trajectory. Since wind speed in the atmospheric boundary layer usually increases with height, the travel speed also increases with increasing σ_z as this parameter affects the plume height after reflection.

1.2.1 *Concentration – impact of σ_z*

Impacts of σ_z on concentrations are shown in Figure A5. For the livestock farm the effect of σ_z on both NH_3 and NH_4^+ concentrations is rather straightforward. Close to the source increased σ_z results in lower concentrations compared to the reference run. This is due to the more diluted plume when receptors are already close to the initial plume height, as is the case for this low source with limited plume rise. On the one hand this causes source depletion due to dry deposition to start at a shorter distance. On the other hand, deposition becomes less due to the lower concentrations. Since more mass remains in the atmosphere, this leads to higher concentrations after a few kilometres. The reverse is true for decreased σ_z.

Relative reductions of concentrations near the source reach about 40% for the maximum increase of σ_z applied here; maximum decreases of σ_z result in maximum relative increases of concentration of about 85% near the source. Relative changes are slightly larger for NH_4^+, probably because there is hardly any secondary material yet near the source. The sensitivity switches sign after about 10 to 30 km and maximum differences gradually increase to about 25% (increased σ_z) and about –30% (decreased σ_z) at 400 km, where differences are now slightly larger for NH_3.

For the industrial stack the sensitivity to σ_z for concentrations of both NO_x and NO_3^- + HNO_3 shows a different pattern than for the livestock farm case. Differences range between about –35% (around 2 km) for increased σ_z and about 50% (around 3 km) for decreased σ_z. After ~50 km the sensitivity to σ_z becomes negligible. At 1 km from the source a decrease in concentration is obtained with increased σ_z, while there is hardly any impact with decreased σ_z. In the latter case concentrations increase at slightly greater distances. Since increased σ_z implies a deeper plume, the plume reaches the receptor height closer to the source (< 1 km) than in the reference run. This leads to increased concentrations with increased σ_z and vice versa at distances near the

source. At greater distance the dilution effect becomes more important and a peak in the sensitivity is obtained, with lower concentrations for increased σ_z and vice versa. This peak is located closer to the source for increased σ_z than for decreased σ_z because it takes longer for the plume to reach the receptor height in the latter case.
At distances between 10 to 50 km the sensitivity changes sign again. Here, concentrations are higher than in the reference run with increased σ_z and lower with decreased σ_z. Maximum differences are about 10% for NO_x and 20% for NO_3^-+HNO_3. This can be explained by the deposition over the trajectory. If the dilution effect results in lower concentrations, there will be less dry deposition over the trajectory, and vice versa. Additionally, the contribution of the differing plume phases to deposition changes. Because the dry deposition is parametrised differently for the various phases of the plume, an increased distance over which Phase 2 (not yet fully mixed) is active, does not balance the decreased contribution by Phase 3. Because the deposition velocity is larger in Phase 2, the impact of changes due to this phase is larger than the one due to Phase 3. The distance over which Phase 2 is active generally becomes smaller with increased σ_z and larger for Phase 3. Hence, source depletion due to dry deposition reduces with increased σ_z, resulting in higher concentrations at a given distance further away from the source. The reverse applies to decreased σ_z.
Increased σ_z also leads to an increased scavenging, and vice versa. However, these changes hardly affect concentration due to compensation of the changed scavenging by effects on travel time. So the main effect of source depletion on concentration is caused by differences in dry deposition. Effects from different plume phases appear to cancel out after about 50 kilometres, beyond which as the sensitivity to σ_z becomes negligible.
For the motorway, variations for NO_x are largest near the source. Relative differences range between about –40% for increased σ_z and just over 100% for decreased σ_z. The crossover point is found just before 100 km. Beyond that distance, the relative differences remain between around –15% and 7%. For the relative difference in concentration NO_3^- + HNO_3 the impact is comparable near the source and slightly smaller after ~200 km. Maximum effects range between –40% and 110% with increased and decreased σ_z respectively. They occur at a distance of about 5 km. Beyond that distance, there is a decrease towards a crossover point around 150 to 200 km, and a reversed impact of only a few percent up to 400 km.
For the motorway, all emissions occur entirely within the boundary layer. Hence, the pattern of changes shows some similarity with the one from the livestock farm. The crossover points are found at greater distances due to the less efficient dry deposition of NO_y compared to NH_x. The relative differences in concentration are mainly determined by dilution effects, resulting in lower concentrations at increased σ_z and vice versa. However, the reversed impacts at distances beyond the crossover point remain relatively small. Decreases and increases of the relative difference are concentrated more around the crossover point than in the case of the livestock farm. Thus, the difference pattern looks more like a "plateau" well before and after the crossover point. This is due to the contributions from the many source segments at differing distances. Contributions from source segments at greater distance compensate reductions from their neighboring segments.

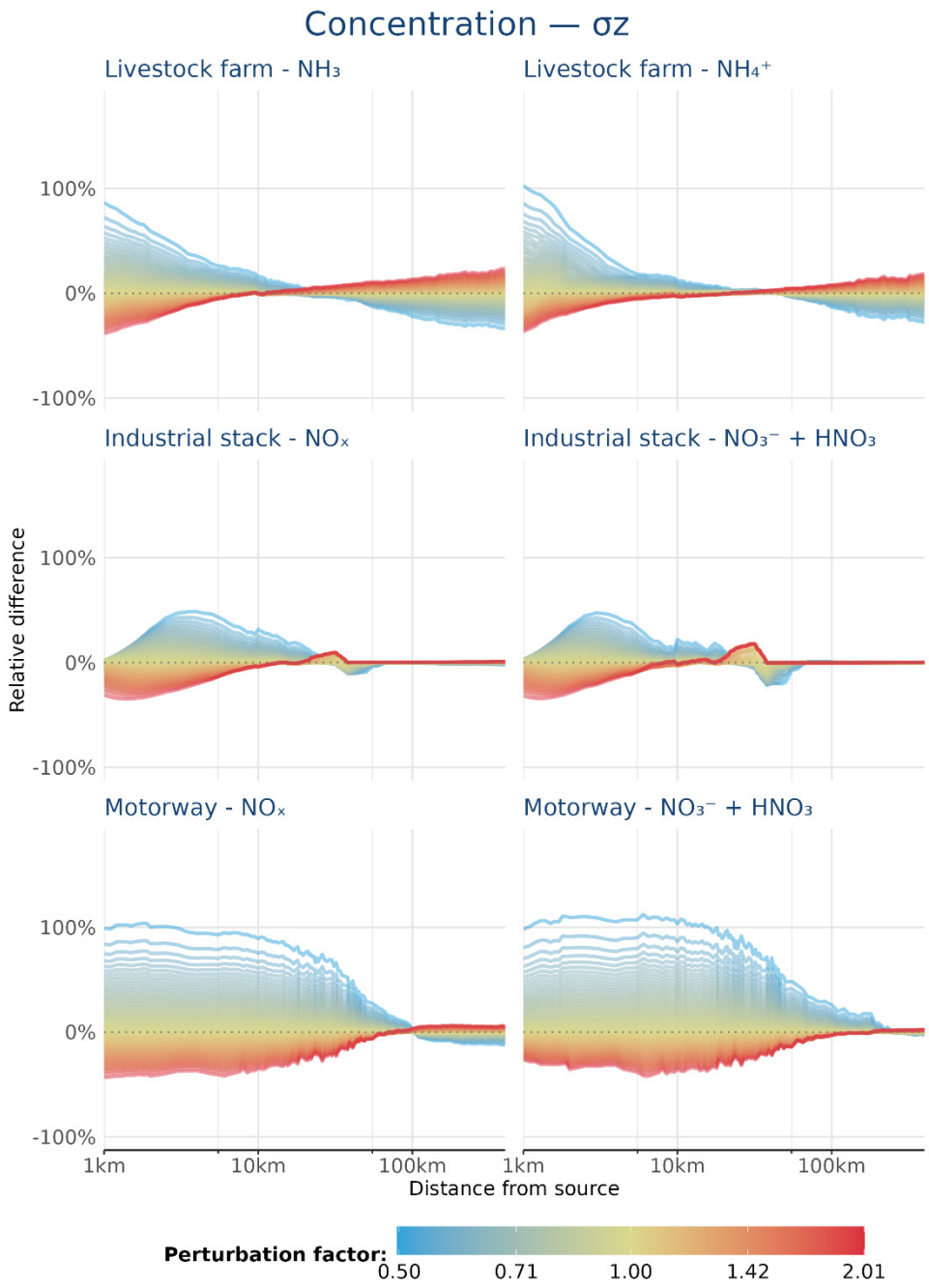

Figure A5 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying vertical dispersion length σ_z. Top row: livestock farm NH_3 and NH_4^+; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.2.2 *Dry deposition – impact of σ_z*

Dry deposition follows the pattern of the concentration (Figure A6). Wider plumes with increased σ_z touch the ground closer to the source, so that dry deposition can start at a shorter distance, compared to the reference run. Apparently, the effect of reduced concentration due to dilution dominates, since reduced dry deposition is found nearby the source. In the case of the livestock farm, after a few kilometer, dry deposition is higher than in the reference run. Because of the reduced dry deposition over the trajectory up to that point concentrations become higher. Such a crossover point is found at greater distances from the industrial stack and motorway. Dry deposition is less effective for the oxidised compounds, compared to the reduced compounds. For decreased σ_z the opposite is found. For the motorway the impact of multiple source segments at various distances from a specific receptor point also plays a role.

Near the source, the relative differences in NH_x deposition vary between –40% and 85% for the livestock farm. Relative differences in NO_y deposition vary between –50% and almost 110% in case of the motorway, and between –40% and 70% between 1 and 5 km for the industrial stack. Up to the crossover point, increased σ_z causes lower dry deposition, and vice versa. Beyond the crossover point differences signs reverse and at about 400 km, relative differences range between about 25% and –35% for the livestock farm, but are only a few percent for NO_y from the motorway and negligible for the industrial stack.

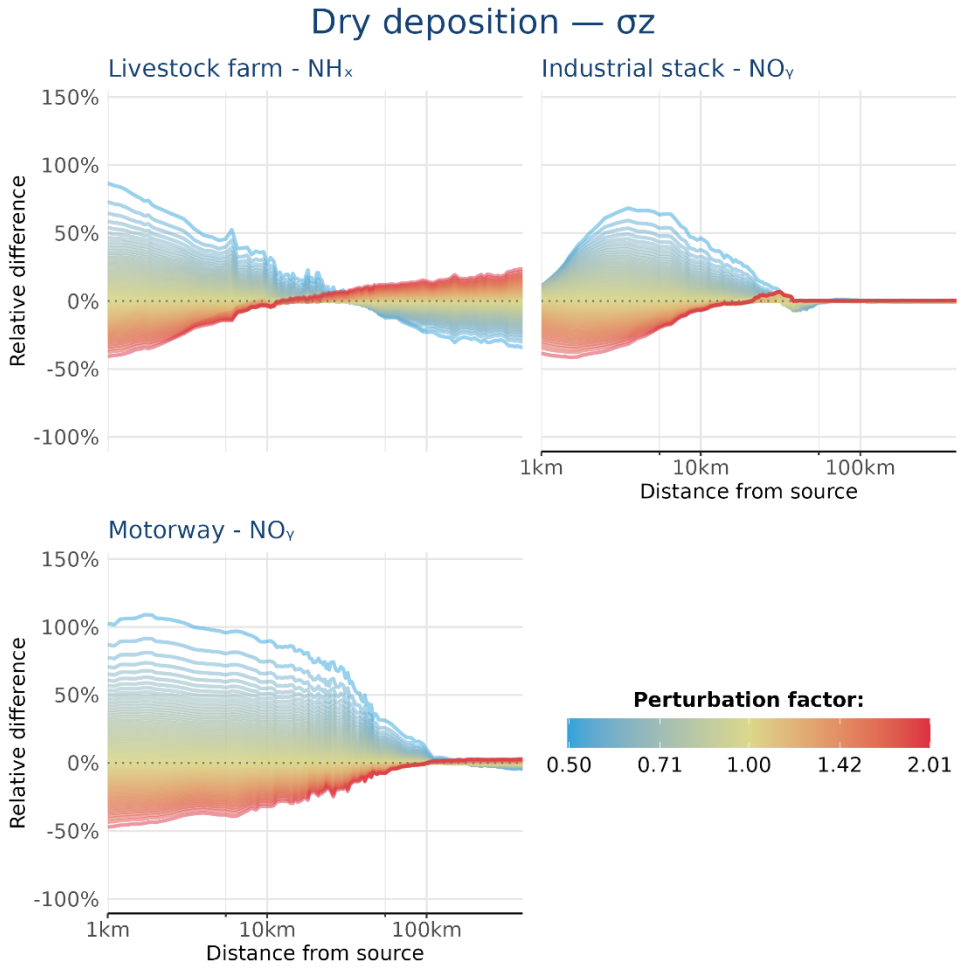

Figure A6 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying vertical dispersion length σ_z. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.2.3 *Wet deposition – impact of σ_z*

The sensitivity of wet deposition to σ_z behaves differently for NH_x and NO_y, respectively (Figure A7). For NH_x from the livestock farm the impact is smallest around 1 km, around 7% to –10% for increased and decreased σ_z respectively. Then, differences increase up to about 5 to 20 km, to values between almost 35% and –45%. There is a more steady variation at greater distances, between around 20% and –35%. For the industrial stack the impact is highest, up to 80%, just after 1 km for increased σ_z, and up to –40% at a slightly greater distance for decreased σ_z. Differences then decrease with distance to a few percent at 400 km. For the motorway again a plateau is seen with highest impact between 50% and –45% in the first 10 km. Relative differences then decrease to values between about 7% and –13% at 400 km.
In general, for all source types wet deposition increases with increasing σ_z, and vice versa. This is likely related to the increased scavenging rate for increased σ_z, and vice versa as explained at the start of Section 1.2. Also, wet deposition takes place over the entire column. Thus, the

sensitivity pattern does not necessarily follow the one of concentrations at the receptor points (cf. Section 1.2.1).

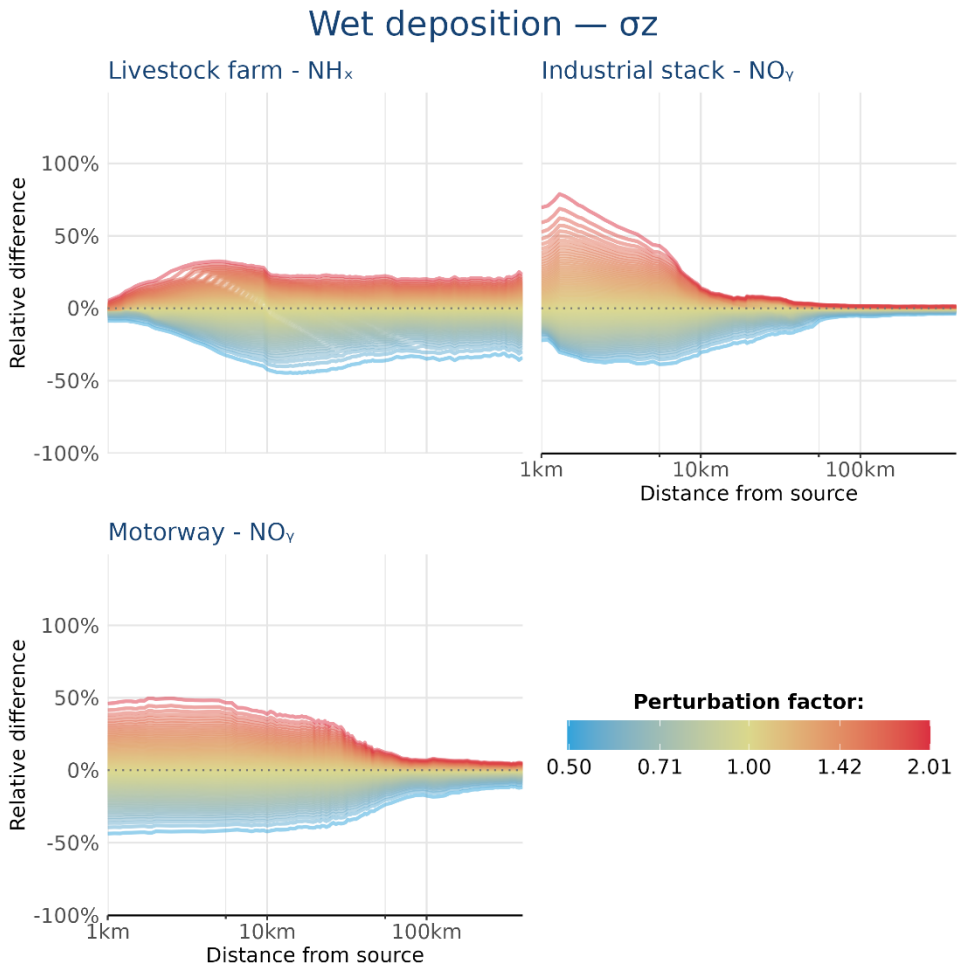

Figure A7 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying vertical dispersion length σ_z. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.2.4 *Total deposition – impact of σ_z*

The sensitivity of total deposition to σ_z (Figure A8) is dominated by the sensitivity of dry deposition, both for NH_x or NO_y and especially near the source. With increasing distance the sensitivity contribution from wet deposition increases. This is especially clear for the low sources, the livestock farm and motorway, where the crossover point is now located at a shorter distance from the source than for sensitivity of dry deposition to σ_z. For the industrial stack the contribution from wet deposition can be seen at distances beyond 50 km in particular. Here, whereas differences in dry deposition are negligible, small differences in total deposition are visible. Furthermore, there are somewhat dampened peaks.

For the livestock farm the combined patterns from wet and dry deposition give the strongest relative difference at 1 km with changes in total deposition between –40% and 80% for increased and decreased σ_z,

respectively. Just before 10 km the sensitivity switches sign and a more or less leveled sensitivity to σ_z of about 25% to −35% is found for increased and decreased σ_z, respectively.
For the industrial stack the relative difference peaks around 2 km for increased σ_z, at about −40% and at a slightly greater distance at 60% for decreased σ_z. Relative differences decrease and change sign between around 25 km, with a small peak of just over 7%, followed by a relative difference of a few percent or less beyond 50 km.
For the motorway the highest impact is also obtained near the source. Relative differences between −45% for increased σ_z and 100% for decreased σ_z are seen. They are about 5% to −10%, respectively beyond the crossover point which is located just before 100 km.

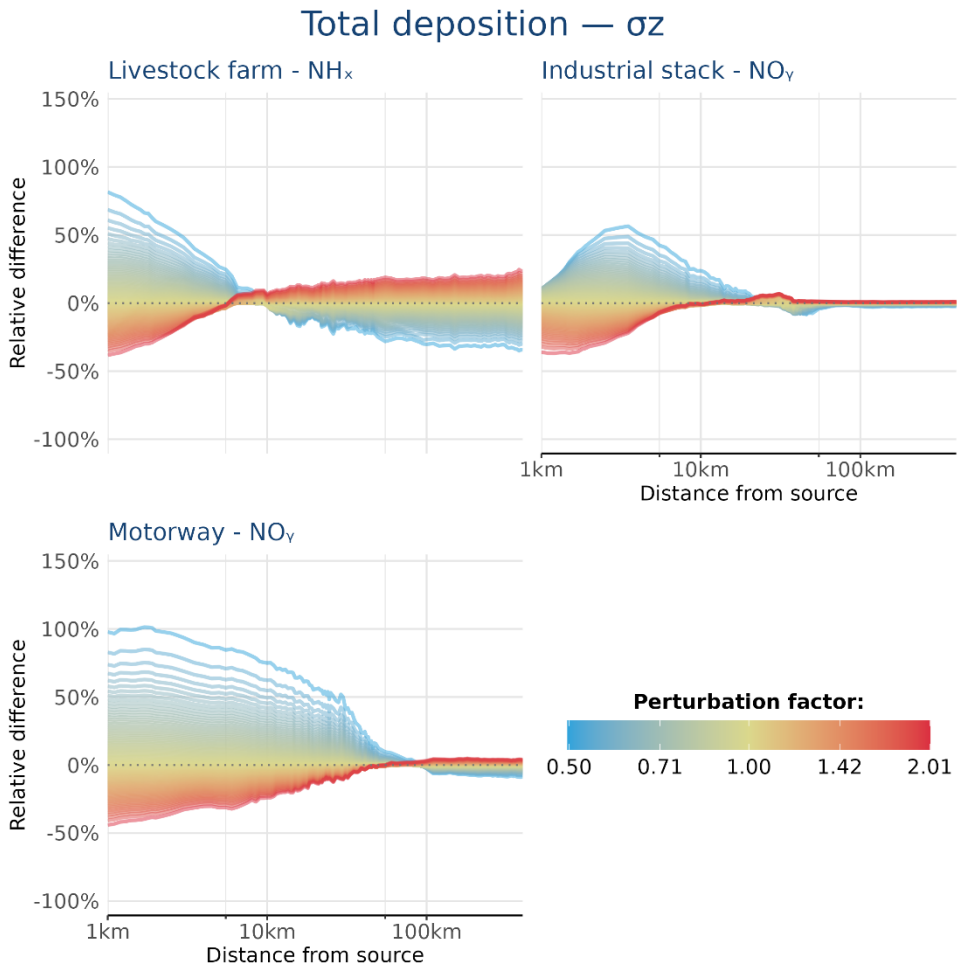

Figure A8 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying vertical dispersion length σ_z. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.3 Mixing layer height z_i

A change in mixing layer height, z_i, has several effects:

- The volume over which pollutants are mixed is affected. With increased z_i pollutants will generally be more diluted than with a decreased z_i. We will refer to this as "the dilution effect".

- Upon emission, the plume height (including plume rise), may exceed z_i. No, or only part, of the pollution is emitted into the mixing layer in that case. The fraction of pollutant assumed to be immediately incorporated into the mixing layer is based on the difference between the plume height and z_i. At greater distances, part of the pollutant mass originally emitted above the mixing layer may be entrained into the mixing layer again, due to boundary layer growth. In either case, if the emission occurs above z_i, OPS sets the emission height of the fraction incorporated in the mixing layer to z_i.
- For plumes emitted below z_i, the distance at which reflection at the top of the boundary layer height occurs is larger (increased z_i) or smaller (decreased z_i). Therefore it may also take longer or shorter, respectively, before the plume becomes well-mixed. The plume height and hence plume speed are influenced as well. In general, increased z_i results in higher plume heights and speeds after reflection at the top of the mixing layer.
- In OPS, the ratio of emission height including plume rise to z_i determines how vertical dispersion length and wind speed at plume height are computed. A distinction is made between the surface layer (lower part of the mixing layer) and the convective or neutral boundary layer above the surface layer, in which different parameterisations are used. In some cases an interpolation between these layers is applied. This results in, sometimes strongly, non-linear effects of z_i on σ_z, rendering it extremely difficult to exactly pinpoint the cause for the various sensitivities z_i.
- Source depletion due to dry deposition in both the not yet well-mixed phase and the well-mixed phase of the plume shows non-linear impacts similar to the aforementioned ones.
- The relative contribution from washout (below cloud) and rainout (within cloud) depends on z_i as well as σ_z. The contribution from rainout increases faster with distance with *decreasing* z_i, and the other way around. This in its turn leads to higher loss rates for decreased z_i and lower ones for increased z_i. Travelling speed of the plume and hence travel time are also affected by z_i. At greater distances a crossover point may be reached for wet deposition at the receptor due to the impact of source depletion on the atmospheric concentration.

1.3.1 Concentration – impact of z_i

Impacts of z_i on concentrations are shown in Figure A9. For the livestock farm, when increasing z_i, lower NH_3 concentrations are found compared to the reference run over the first 200 km or so. This is due to the dilution of the emissions over the larger mixing layer. Concentrations increase with decreasing z_i due to the smaller mixing volume. This effect is somewhat smaller at 1 km, when the plume is likely not yet fully mixed, and increases towards a maximum at about 10 to 20 km. Here, differences range between about –40% (increased z_i) and 80% (decreased z_i). At greater distances, a crossover point is found for NH_3 concentration. This is probably caused by the interaction between concentration and deposition. Because a smaller concentration leads to less deposition over the trajectory, more mass remains in the atmosphere at greater distance with increased z_i, and vice versa. The

crossover is found at shorter distances for the decreased z_i than for increased z_i. Beyond the crossover point, sensitivities of NH_3 concentration increase again. Differences are between about 10% to –25% after 400 km.
The relative differences in NH_4^+ concentration look somewhat similar to those for NH_3. However, since dry deposition of NH_4^+ is less efficient than for NH_3, no crossover point is obtained within 400 km. The relative sensitivity is a bit larger than for NH_3 concentration. Around 10 km impacts range between –50% for increased z_i and 125% for decreased z_i. They decrease to about –25% and 15% at 400 km.
Results for the industrial stack are not as easily explained due to the aforementioned non-linear effects. The sensitivity to z_i is much more variable than for both lower sources. Differences between about –50% and 35% (NO_x) and –60% and 160% (NO_3^- + HNO_3) for increased z_i are found, and between –25% and 90% (NO_x) and –45% and 105% (NO_3^- + HNO_3) for decreased z_i. Some features are discussed next.
For very stable conditions component model results (not shown) indicate that the plume height initially exceeds z_i. Increased z_i results in fumigation (entrainment into the mixing layer) at relatively short distance in comparison with the reference run, but decreased z_i results in fumigation at longer distance, up to ~100 km.
Near the source, at 1 km, the impact of the increased z_i is small. Probably, the dilution effect is counteracted by the additional mass that is emitted inside the mixing layer with increased z_i. With decreased z_i slightly lower concentrations are found compared to the reference run, presumably because more mass is now emitted above z_i.
After a few kilometres the effects switch sign, possibly because of the dilution effect: less dilution with reduced z_i and more dilution with increased z_i. However, at a slightly greater distance, between around 3 km and a few tens of km, both with increased and decreased z_i, NO_x concentrations become higher than the reference run. For the increased z_i -runs this might be because more mass is fumigated into the mixing layer. Furthermore, there may be more or less source depletion due to dry deposition in Phase 2 of the plume (not yet well mixed), with either increased or decreased z_i (non-linear effects). Component model results studied in eastward direction (not shown) indicate that for all runs the mass previously emitted above the z_i has been entrained into the mixing layer after 100 km. We expect that in most simulations the plume has then reached the well mixed phase, so that increased z_i leads to lower concentration and vice versa. Since source depletion due to dry deposition in Phase 3 and due to wet deposition was mostly less for increased z_i we presume that the sensitivity obtained after this distance is largely explained by the dilution effect: increased z_i leads to more dilution and hence to lower concentrations, and vice versa. The magnitude of this effect reduces with distance due to altered source depletion.
For the motorway relative differences have the same sign as for NH_3 from the livestock farm. No crossover point is reached within 400 km. The impact on NO_x concentration peaks after about 100 km and ranges between about –45% and just over 60% at increased and decreased z_i, respectively. For NO_3^-+HNO_3 no clear maxima are obtained and variations range between –15% and –40% for increased z_i and between 35% and 75% for decreased z_i, depending on distance.

Figure A9 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying the mixing layer height, z_i. Top row: livestock farm NH_3 and NH_4; middle row: industrial stack NO_x and NO_3^-+ HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.3.2 *Dry deposition – impact of z_i*

Impacts of z_i on dry deposition are shown in Figure A10. For the livestock farm source the dry deposition of NH_3 and NH_4^+ is expected to reflect the individual concentration patterns shown in the previous Section. This is because z_i has no impact on the dry deposition process as such. However, the influence of NH_3 on the NH_x deposition appears to be dominant over that of NH_4. This is due to the more efficient dry

deposition of NH_3 than of NH_4. As such, a peak in sensitivity is again obtained at ~10 – 20 km, with differences ranging between about –40% and 70% for increased and decreased z_i, respectively. This sensitivity then gradually decreases and switches sign after 200 – 300 km. The crossover point is located a bit further away compared to the one for NH_3 concentration, probably because the dry deposition of NH_4^+ does not switch sign within 400 km since it is less efficient. At 400 km sensitivities are slightly smaller than the ones for NH_3 concentration.
For the industrial stack, relative differences in dry deposition of NO_y also tend to follow the ones of the concentrations. Over the 400 km studied here, an increased z_i results in a relative difference in dry deposition of NO_y between about –50% and 15%. This is between about –5% and 90% for decreased z_i. A notable difference with the sensitivity pattern for concentration is, that the switches in sign for the peaks between ~5 km and several tens of km are less profound. This seems to be rather odd, since z_i has no impact on the dry deposition process *per se* and the influence of source depletion over the trajectory is expected to be minor *at the receptor*. This requires further investigation.
For the motorway the relative differences in dry deposition of NO_y near the source again resemble the variations seen for NO_x concentration. Here, not much secondary material has formed and most dry deposition of NO_y is due to NO_x. For greater distances dry deposition of both primary and secondary compounds are important. This results in a relative difference of NO_y dry deposition between –10% with increased z_i, and 20% with decreased z_i near the source, and –40% and 70%, respectively around 100 km, decreasing to around –40% and 40%, respectively, at 400 km.

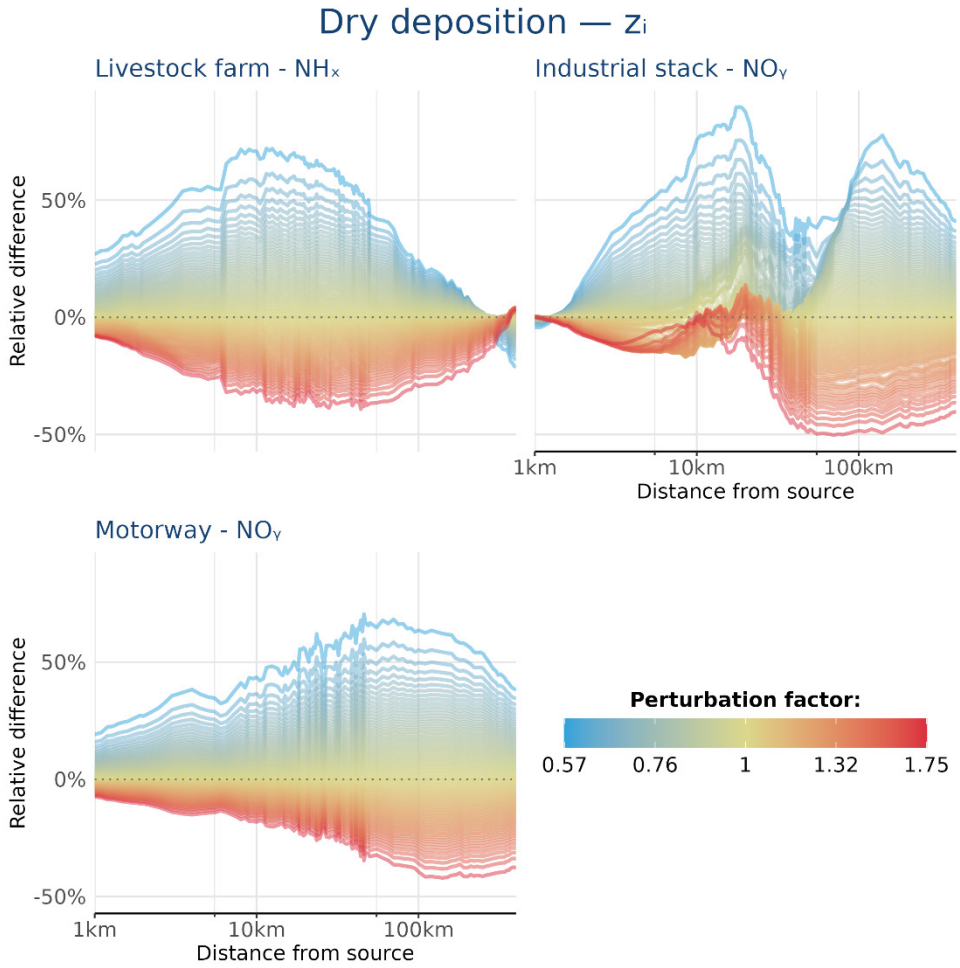

Figure A10 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying z_i. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.3.3 *Wet deposition – impact of z_i*

Impacts of z_i on wet deposition are shown in Figure A11. Since wet deposition occurs over a column it is less related to the actual concentrations at the receptor level shown in Section 1.3.1 It was found from the component model results (not shown) that for several stability classes the wet deposition was less efficient when z_i was increased. This can be recognised in the figure for the livestock farm. Up to 50 to 100 km from the source, less wet deposition is found with increased z_i and vice versa. The largest differences are found at a few kilometres up to around 10 km from the source. They range between –35% for increased z_i and 50% for decreased z_i. The effect changes sign after ~80 – 150 km. This is probably caused by increased deposition over the trajectory when z_i decreases, so that less material remains in the atmosphere, and vice versa. The sensitivity then gradually increases again. At 400 km, differences up to about 40% are obtained, which have become negative for decreasing z_i and positive for increasing z_i.
For the industrial case, in general, less wet deposition is found with increased z_i and vice versa, over almost the entire distance studied here. The sensitivity tends to decrease with distance and changes sign

just around 400 km. The latter is due to the aforementioned source depletion effect. As such on average the variations range between about −70% and 90% near the source, decrease with distance up to the crossover point and amount from a few percent to −15% at 400 km. A somewhat stronger variation is found up to several tens of km. This could be related to variations in the amount of mass inside the mixing layer.
For the motorway case also a general decrease in relative difference of wet deposition of NO_y is seen when increasing z_i, and vice versa. This is due to the impacted wet deposition loss rates. Relative differences are highest, between −45% and 85% with in- and decreased z_i, respectively in the first 10 km. Variations are relatively small, which is likely due to the impact of many source segments, at varying distance from a specific receptor point. Beyond ~10 km, the sensitivity reduces until the crossover point just around 400 km. Differences of several percent to −15% are found at 400 km.

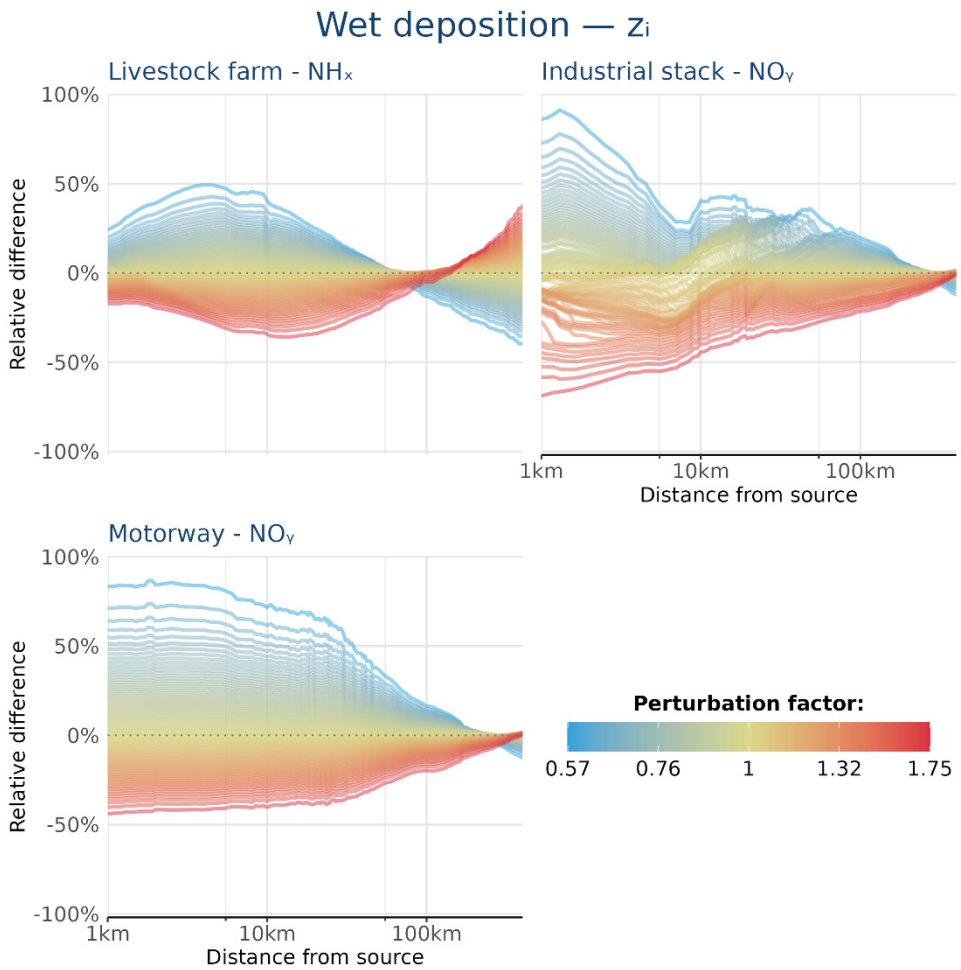

Figure A11 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying z_i. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.3.4 Total deposition – impact of z_i

Near the sources studied here the relative differences in total deposition due to changes in z_i (Figure A12) is influenced most by dry deposition, for NH_x as well as NO_y. The impact of wet deposition increases with distance. For NH_x from the livestock farm the latter is evident from, for example, the location of the crossover point. This is located between the one for dry deposition and wet deposition, respectively. In the case of the industrial stack and the motorway, the relative sensitivity of total deposition at large distance is clearly less than the one of dry deposition.

For the livestock farm the sensitivity peaks at a short distance from the source, with generally lower total deposition for increased z_i, and vice versa. Relative differences amount to values between about –35% and 60%. Due to the dilution of concentration with increased z_i, there is less dry deposition. In addition, the wet deposition decreases. This implies that less material is lost due to source depletion. This leads to a crossover point for total deposition of NH_x, in this case at a distance of about 150 km. Reversed effects apply to decreased z_i. Beyond the crossover point the sensitivity increases again, leading to relative differences at 400 km between 25% and –35% for increased and decreased z_i, respectively.

For the industrial stack the sensitivity to an altered z_i is more variable over distance than for the livestock farm case. Differences range between about –40% for increased z_i and about 75% for decreased z_i. Strongly non-linear effects can be recognised between a few kilometres from the source, up to about 50 – 80 km. Increased z_i predominantly leads to lower total deposition compared to the reference run, but at some distances higher total deposition is also found. Such a feature was also clearly visible in the dry deposition differences in particular. After about 50 – 100 km the differences become more consistent, with lower total deposition for increased z_i, and vice versa. This is likely due to the dominant impact of dilution.

For the motorway case the relative differences in NO_y total deposition predominantly show a decrease when z_i increases, and vice versa. This is the dilution effect. The impact is smallest close to the source where differences remain between about –10% and 25%. The sensitivity then peaks between roughly 50 – 100 km, with differences ranging from about –35% to 60%. Beyond 100 km the sensitivity decreases again with distance and differences between –15% and 10% are obtained at 400 km. Contrary to the livestock farm, the crossover point is not reached within 400 km, likely due to the less efficient dry deposition of the oxidised compounds.

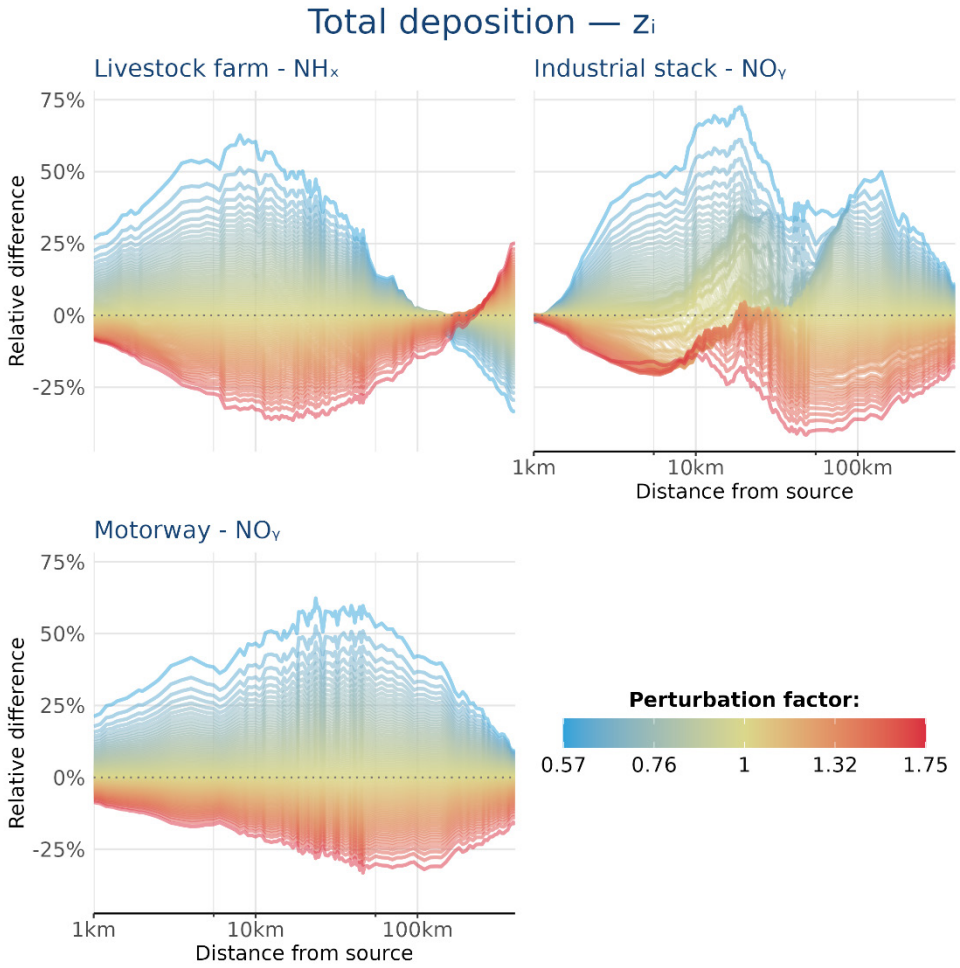

Figure A12 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying z_i. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.4 Chemical conversion rate k_{chem}

A change in chemical conversion rate, k_{chem}, has the following main impacts:

- Increasing k_{chem} reduces the atmospheric concentration and hence deposition of the primary emitted material and vice versa. In the present - Lagrangian - framework, the effect of k_{chem} on the concentration of primary material can to first order be described by an exponential decay. Relative differences between runs can also be described by an exponential function, with the difference in chemical conversion rate being the time constant.
- With higher k_{chem}, more secondary material is formed and vice versa. This is complementary to the decay of primary material. First-order effects of k_{chem} on formation and concentration of secondary compounds therefore also follow an exponential function, complementary to the one of primary compounds.
- Effects on deposition, both dry and wet, follow those on concentration, but may be strongly modulated by differing deposition efficiencies of primary versus secondary compounds.

1.4.1 Concentration – impact of k_{chem}

Effects of k_{chem} on concentration (Figure A13) are virtually absent for the primary material near the source since not much time has passed yet. They clearly increase with distance from the source, closely following an exponential curve, which is expected from theory. Thus, the sensitivity increases with distance. At 400 km differences between about –70% and 70% (NH_3, livestock farm) and –65% and 60% (NO_x, both industrial stack and motorway) are obtained. The relative difference in NO_x concentration for the motorway is slightly larger already near the source due to the impact of multiple source segments with varying source-receptor distance.

For the secondary material, an increase in k_{chem} gives higher concentrations and vice versa. Contrary to the primary material, the effect of conversion rate is highest at the source and decreases with distance, like may be expected from the exponential decay of primary material (see above). The relative difference in concentration of the secondary material near the source is about 120% to 130% for increased k_{chem} and –60% for decreased k_{chem}. This applies to all source types studied here. At 400 km differences vary between about –45% and 50% (NH_4^+, livestock farm) and –45% and 30% (NO_3^- + HNO_3, both industrial stack and motorway).

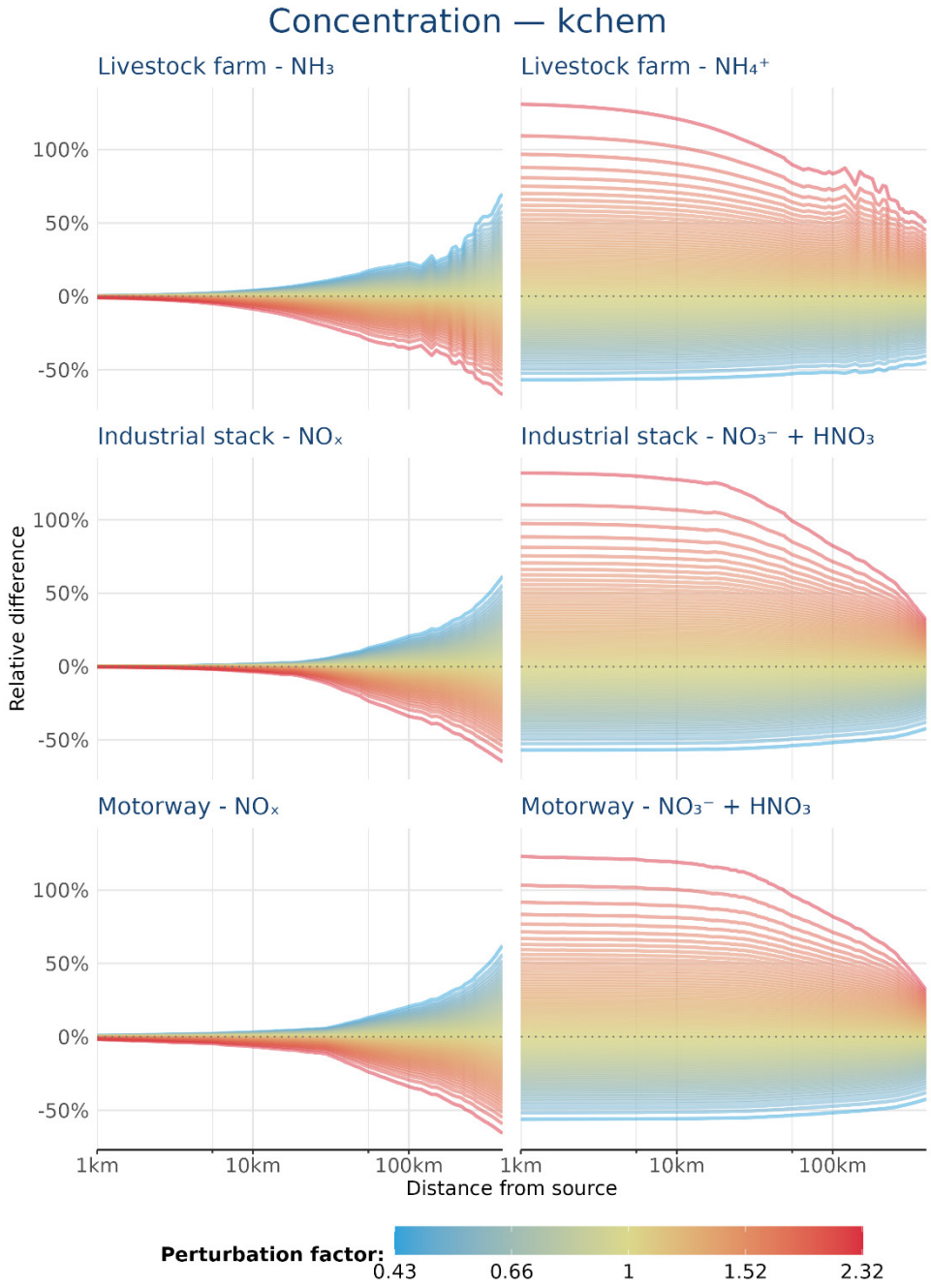

Figure A13 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying k_{chem}. Top row: livestock farm NH_3 and NH_4; middle row: industrial stack NO_x and NO_3^- + HNO_3; bottom row: motorway NO_x and NO_3^- + HNO_3.

1.4.2 *Dry deposition – impact of k_{chem}*

Impacts of k_{chem} on dry deposition are shown in Figure A14. For primary as well as secondary material, dry deposition is strongly linked to atmospheric concentration. The sensitivity of dry deposition roughly follows the one of their concentrations. For the primary compounds the impact on dry deposition decreases with increasing k_{chem}, and vice versa.

For the secondary compounds the impact on dry deposition increases with increasing k_{chem} and vice versa.
Since the effects of primary and secondary material have opposite signs, the impact on the dry deposition of NH_x and NO_y depends on the relative efficiency of the dry deposition of the various compounds. Strong fluctuations in sensitivity are obtained, due to the contribution from secondary compounds. The fluctuations are most pronounced for NO_y.
For the livestock farm, the sensitivity of NH_x deposition roughly follows the one of concentration of the primary compound, NH_3. However, compared with concentrations, the sensitivities of deposition are somewhat reduced. This is the case especially at longer distance from the source, where the impact levels off due to the increased NH_4^+ concentration and hence increased dry deposition of NH_4^+. However, in general the effect of NH_4^+ seems limited, due to the much more efficient dry deposition of NH_3 compared to NH_4^+. The relative difference in dry deposition of NH_x due to perturbed k_{chem} reaches just over ±40% at 400 km.
By contrast, for the industrial stack and motorway, the sensitivity of the NO_y dry deposition also increases with distance, but the sign of the difference is consistent with the one of the secondary material. Increased k_{chem} gives increased NO_y dry deposition, and vice versa, reaching a maximum relative difference of +30% around 300 km for increased k_{chem} and –25% just before 400 km for decreased k_{chem}. This can be explained by either or both of the following:

- the dry deposition of NO_3^- + HNO_3 is more efficient than the dry deposition of NO_x.
- deposition is expressed in NO_y equivalents. Because the molar mass of NO_3^- and HNO_3 is higher than of NO and NO_2, dry deposition of NO_3^-+HNO_3 contributes more to the computed combined deposition.

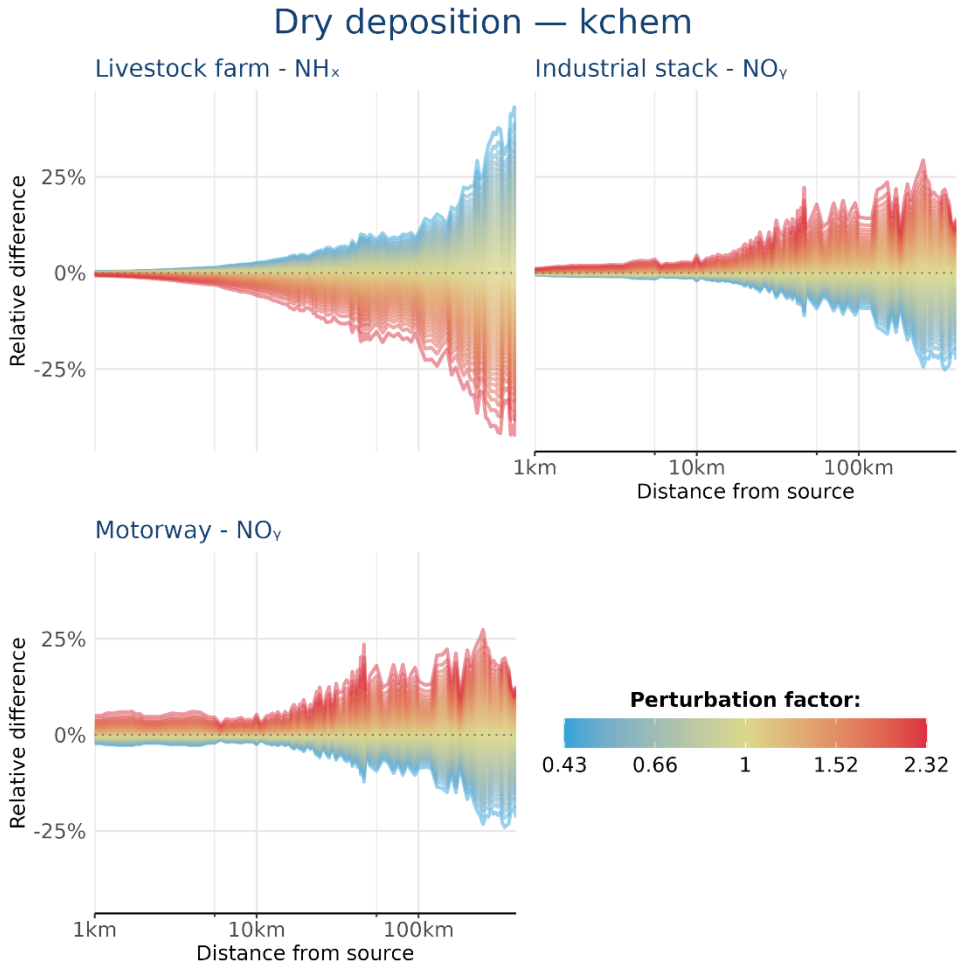

Figure A14 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying k_{chem}. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.4.3 *Wet deposition – impact of k_{chem}*

Impacts of k_{chem} on wet deposition are shown in Figure A15. Wet deposition is also strongly linked to concentration, or mass in the atmospheric column. Hence, for all source types, the relative difference in wet deposition of the primary and secondary material follows the one of their concentration. The impact of k_{chem} is smallest near the source for the primary material, which is most abundant near the source. In spite of this, we see different impacts for NH_x and NO_y, respectively.

Wet deposition is quite effective for both NH_3 and NH_4^+, but is more effective for the latter compound. Furthermore, the slight difference in mass between NH_3 and NH_4^+ may cause a slightly increased contribution from NH_4^+ to the NH_x wet deposition. For the livestock farm case, this causes a rather strong effect on the sensitivity of wet deposition of NH_x to k_{chem}.

The individual contributions from NH_3 and NH_4^+ tend to compensate each other due to the opposing sign of the differences. This results in a smaller range of relative differences than for concentration at greater distances. On the other hand, the sign of the sensitivity follows the one of the NH_4^+ concentration, which is opposite to the one of NH_3

concentration. This is indicative of a strong impact NH_4^+. This also becomes apparent as the local maximum in the sensitivity at about 10 km. Beyond this distance, a slight decrease up to around 50 km is obtained, which is followed by a large increase. At 400 km relative differences range between about –15% and 20%. Differences are positive for larger k_{chem} and negative for smaller k_{chem}. Some of the variation over distance may be caused by the changing relative contribution from washout and rainout to wet deposition, and the difference in efficiency these processes between NH_3 and NH_4^+.

Sensitivity of NO_y wet deposition is mainly driven by HNO_3+NO_3^-. This is explained by the low solubility of NO_x, which renders NO_x removal by wet deposition an inefficient process in comparison with HNO_3+NO_3^-. Again, the higher molar mass of the secondary material also causes a larger contribution from HNO_3+NO_3^-. The latter effect explains that an increase in wet deposition of NO_y is seen in the first few kilometres. Even though there is not yet much secondary material available, a removal of primary material with lower mass is replaced by secondary material with higher mass. For greater distance the more efficient wet deposition of the secondary material becomes more important. Again the variation in the first tens of kilometres may be attributed to the variation in efficiency in wet deposition and the related relative contribution of rainout and washout. The impact is stronger than for the livestock farm, because of the stronger difference in efficiency between wet deposition of NO_x and NO_3^-+HNO_3 in comparison with NH_3 versus NH_4^+. The relative difference is also more constant over the 400 km with maximum differences up to 90% for increased k_{chem} and –40% for decreased k_{chem}.
For the motorway, the sensitivity resembles the one for the industrial stack. The variation in the first 100 km shows less variation, due to the impact of multiple source segments with varying source-receptor distance. Relative differences in wet deposition amount to maxima of about 90% for increased k_{chem} and –40% for decreased k_{chem}, respectively.

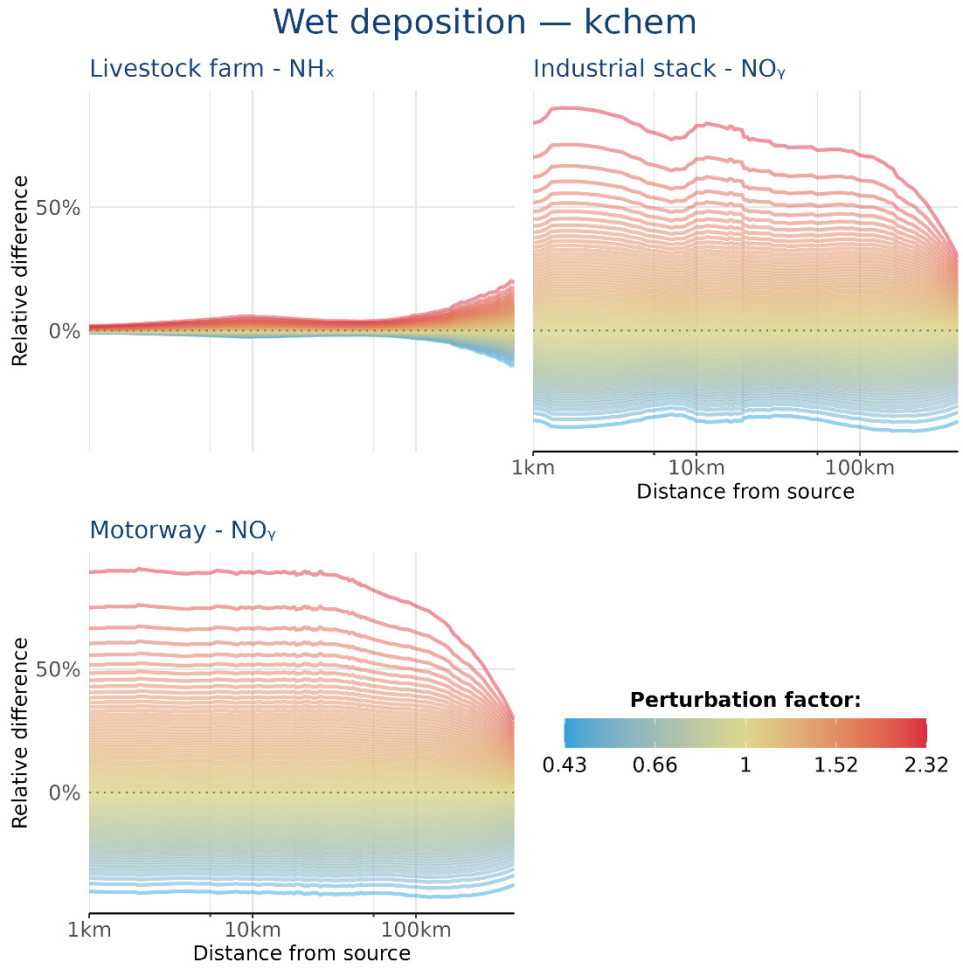

Figure A15 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying k_{chem}. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.4.4 *Total deposition – impact of k_{chem}*

The impact of k_{chem} on the total deposition (Figure A16) of NH_x from the livestock farm is the result of the different processes described above, but not directly as straightforward. In general, the total deposition of NH_3 and NH_4^+ follows the concentration of NH_3 and NH_4^+, respectively. However, the slight variations in the relative differences of the individual deposition components (dry and wet, NH_3 and NH_4^+) in the various spoke directions result in "bumps" and "bends" in the average differences. In general, nearby the source dry deposition is most important. It can be seen that the wet deposition starts playing a more important role at greater distances, since the impact of the dry deposition is strongly reduced. As such, the relative differences in total NH_x deposition are only a few percent or less, though with minor peaks amounting to values between about plus and minus 8%, between 50 and 300 km.

The impact of an altered chemical conversion on the total deposition of NO_y for the industrial stack and motorway is more straightforward than for NH_x from the livestock farm. Near the source the effect of the dry deposition of NO_y is dominant over the wet deposition. The large

sensitivity of wet deposition at this distance is nearly invisible in the total deposition. With distance the impact of the wet deposition of NO_y becomes more important. The sensitivity of dry deposition is amplified by the one of wet deposition as they are of similar sign. In general, an increased total deposition of NO_y is obtained with increased k_{chem}, and a decreased total deposition of NO_y with decreased k_{chem}. This is explained by the higher efficiency of deposition for the secondary material in combination with the higher molar mass of the secondary material. Differences are smallest near the source, up to a −2% and 4% for the industrial stack and −4% to 8% for the motorway. They increase up to −30% at 400 km with decreased k_{chem}. With increased k_{chem} maximum differences are about 45% with at ~200 km, decreasing slightly at greater distances to just over 20% at 400 km.

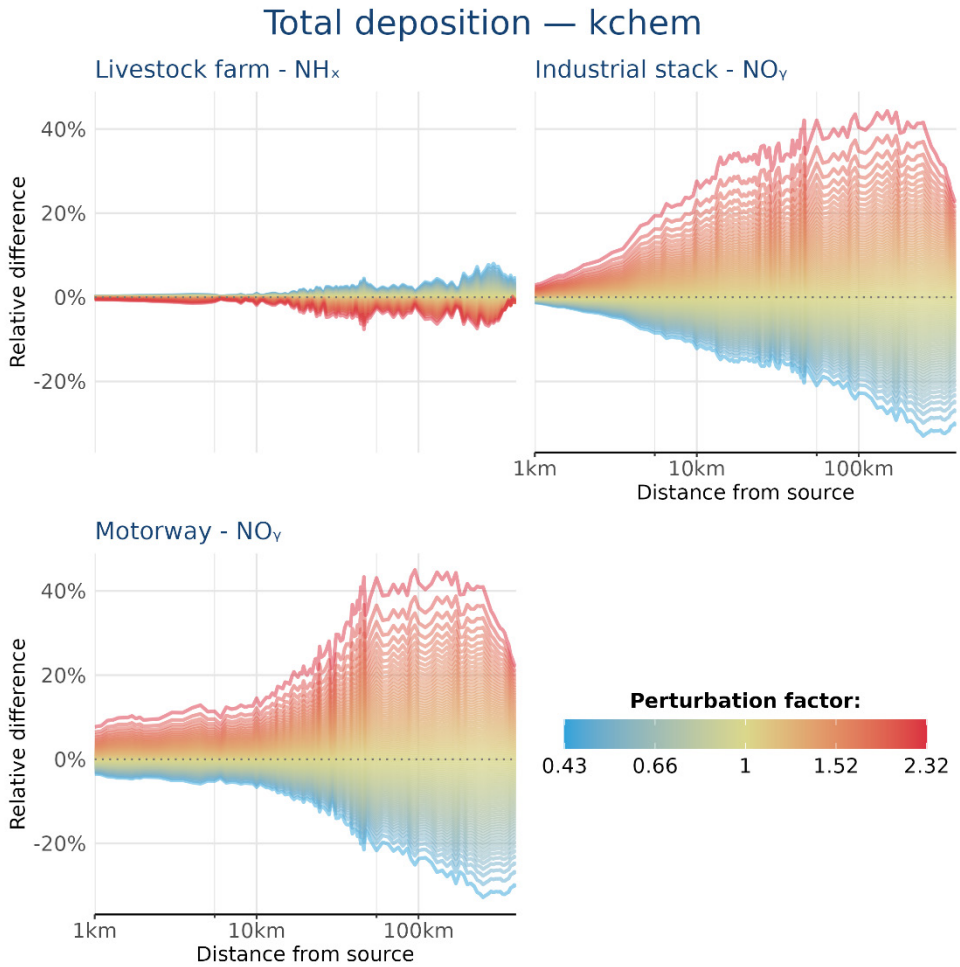

Figure A16 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying k_{chem}. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.5 Dry deposition velocity – primary material $v_{d,pri}$

The dry deposition velocity of the primary material ($v_{d,pri}$) was adjusted for both the trajectory (source depletion) and at the receptor (local effect), using the same perturbation factor for both. This has the following general impacts:

- Increasing $v_{d,pri}$ results in a decrease of concentration, since more material will be removed from the atmosphere due to source depletion over the trajectory. The reverse will be true for a decrease in $v_{d,pri}$. This effect increases with distance since there has been more time for source depletion to take place.
- Initially, close to the source, effects of source depletion are very small or even nearly absent. Because dry deposition is basically computed as the product of deposition velocity and concentration, a given relative change in $v_{d,pri}$ leads to an almost equally large relative effect on dry deposition at the receptor. Hence, a strong contribution of this parameter to the uncertainty in dry deposition close to the source is anticipated. However, because the effect of source depletion on concentration increases with increasing distance, the initial effect of $v_{d,pri}$ on the dry deposition gradually decreases with distance. It can be shown that -theoretically- this feedback loop may ultimately lead to a crossover point, where the effect of $v_{d,pri}$ on the dry deposition is reversed. This occurs at a distance from the source that depends on the magnitude of $v_{d,pri}$. Around the crossover point, the sensitivity and hence the contribution of $v_{d,pri}$ to the uncertainty is minimal. Many complicating factors may obscure the effect, rendering the exact location of the crossover point rather unpredictable in practice.
- Since wet deposition is a strong function of the mass in the atmospheric column, the sensitivity of wet deposition roughly follows the one of concentration.
- The sensitivity pattern of total deposition to $v_{d,pri}$ largely resembles the one of dry deposition. However, the contribution of wet deposition causes the crossover point to shift somewhat towards the source since it enhances source depletion over the trajectory.
- Concentration of the secondary material is impacted as well, because less (increased $v_{d,pri}$) or more (decreased $v_{d,pri}$) primary material can be converted to secondary material. Since $v_{d,sec}$ is not perturbed in this part of the study, impacts on the dry and wet deposition of the secondary material will mirror the impact of $v_{d,pri}$ on the concentration of secondary compounds.

1.5.1 Concentration – impact of $v_{d,pri}$

Impacts of $v_{d,pri}$ on concentrations are shown in Figure A17. Increased $v_{d,pri}$ leads to decreased concentration of both primary and secondary material compared to the reference run, and vice versa. In general, the impact increases with distance. The effect on the concentration of primary material is caused by increased and decreased source depletion, respectively. This is the first step in the aforementioned compensation mechanism, which is due to the feedback between concentration and dry deposition.

At close proximity to the source, the time during which mass can be removed from a parcel of air is limited. For example, at an average wind speed of 5 m/s an air parcel needs a bit more than three minutes to travel a distance of 1 km. In such a short period of time only a limited amount of mass can be removed from the air by dry deposition. Nevertheless, the dry deposition of NH_3 is sufficiently efficient to cause a clearly distinguishable effect at 1 km from the source already.

The concentration changes of primary compounds imply, respectively, less and more conversion to secondary material. Since the dry deposition of NH_3 is more efficient than the dry deposition of NO_x, the impact is larger for the livestock farm simulations than for the industrial stack and motorway.
For NH_3 from the livestock farm the relative difference in concentration ranges between around –30% to –35% for increased $v_{d,pri}$ and 12.5% to 15% for decreased $v_{d,pri}$, at short distances up to 10 km from the source. Maximum relative differences increase to values between about –85% and 150% at 400 km. For NH_4^+ concentration, the relative differences are quite similar to the ones of NH_3 near the source. However, they are smaller at greater distance, between around –70% and 70%, which may be due to the fact that $v_{d,sec}$ is not perturbed.
The effect of $v_{d,pri}$ on the NO_x concentrations in the case of the industrial stack also increases with distance. However, the relative differences are negligible near the source, and only between about –35% (increased $v_{d,pri}$) and 12% (decreased $v_{d,pri}$) at 400 km, which is clearly less than in the case of NH_3 from the livestock farm. This difference is due to the fact that NH_3 deposits much more readily than NO_x. Relative differences in the concentration of NO_3^-+HNO_3 are minor in the first tens of kilometres as well, with very slight deviations from zero between 15 and 40 km. This may be related to a small effect of fumigation in the case of this tall stack. These variations are small, up to a few percent only. After 40 km the sensitivity increases more steadily resulting in differences between about –22% for increased $v_{d,pri}$ and 8% for decreased $v_{d,pri}$.
For the motorway the relative differences for NO_x concentration close the source range between about –10% and 2% for increased and decreased $v_{d,pri}$, respectively. The sensitivity increases with distance to yield differences between –5% and 17% at distances between 100 and 400 km. The effect of multiple source segments with varying distances to the receptor can also be distinguished. In comparison with NO_x from the industrial stack, the differences are larger close to the source center and their increase is less steep at greater distances. For the concentration of NO_3^-+HNO_3 changes range between –15% and 6% near the source, and –35% and 15% at 100 km. A small decline in impact is seen at greater distances.

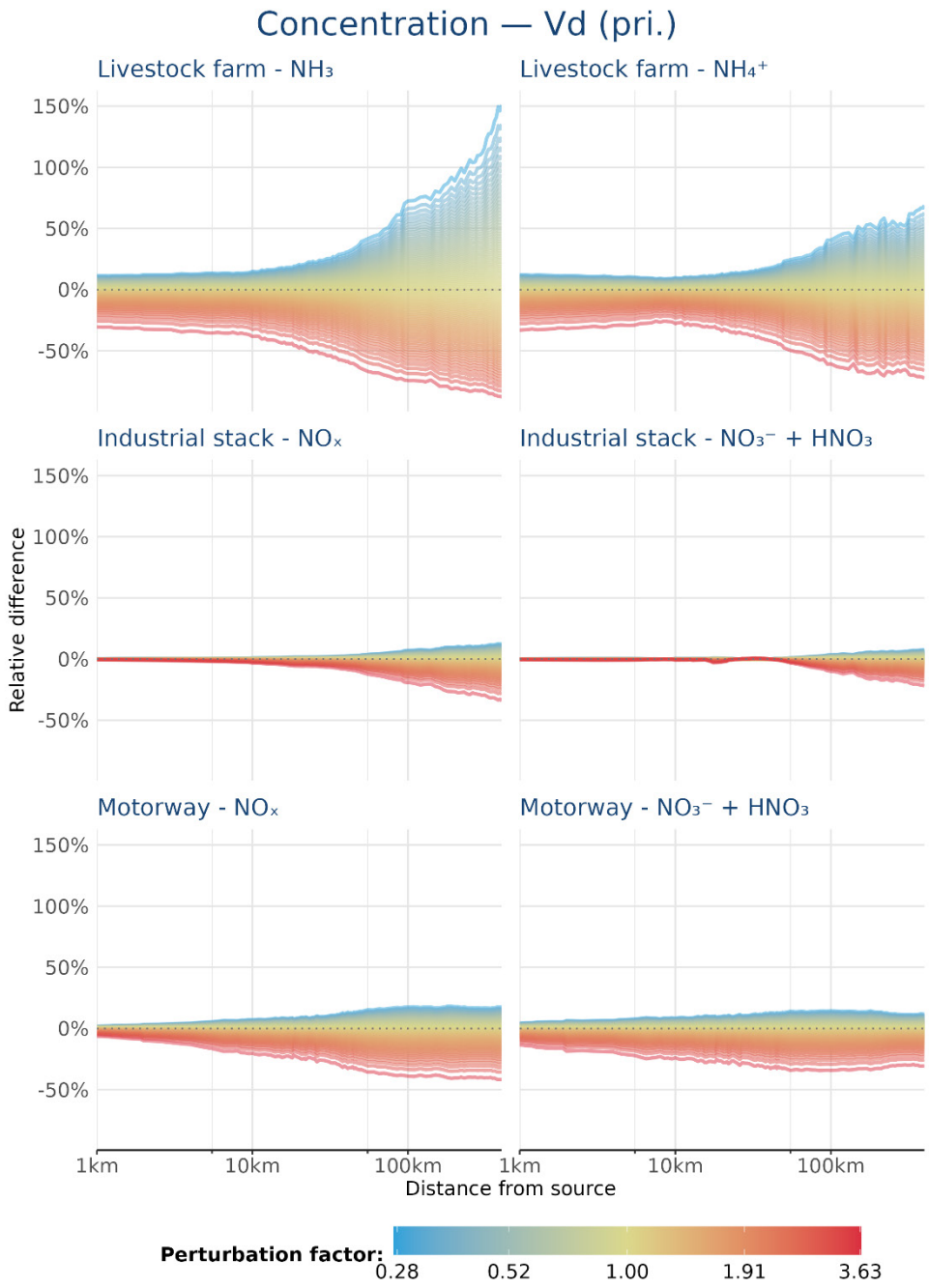

Figure A17 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying $v_{d,pri}$ both for the trajectory and at the receptor. Top row: livestock farm NH_3 and NH_4^+; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.5.2 *Dry deposition – impact of $v_{d,pri}$*

Impacts of $v_{d,pri}$ on dry deposition are shown in Figure A18. It was shown in the previous Section that the sensitivity of concentration to $v_{d,pri}$ is small near the source. This implies that the effect of a perturbation of $v_{d,pri}$ on dry deposition of the primary material at the receptor is nearly proportional to the change in $v_{d,pri}$, in particular for

species of which $v_{d,pri}$ is small. For NH_3, with high $v_{d,pri}$, some influence of changed concentrations on the dry deposition can already be distinguished at 1 km from the source. In this case the sensitivity to $v_{d,pri}$ is clearly smaller than for NO_x.
The effect of $v_{d,pri}$ on concentration of the primary material increases with increasing distance from the source (see previous Section). The increasing effect on the concentration may ultimately outweigh the effect of the changed $v_{d,pri}$. This results in the aforementioned cross-over point. Up to that point the sensitivity of dry deposition to $v_{d,pri}$ decreases to become near-zero. At the crossover point the sensitivity changes sign and increases again at greater distances.
The impact on dry deposition of the secondary material is somewhat different since $v_{d,sec}$ has not been perturbed. An increased $v_{d,pri}$ reduces the amount of primary material in the atmosphere. Thus, there is less conversion to secondary material, which also reduces its dry deposition. Obviously, the opposite is true for decreased $v_{d,pri}$. This impact is smallest near the source and increases with distance. This general behavior is seen for all source types.
Figure A18 depicts the combined effect, i.e., effects of $v_{d,pri}$ on dry deposition of NH_x and NO_y, respectively. For the livestock farm the impact on the dry deposition of NH_3 is dominant over that of NH_4^+. Near the source dry deposition of NH_x changes in accordance with the perturbation of $v_{d,pri}$, though it is modulated somewhat by the concentration changes of NH_3 in particular. This makes sense in two ways: 1) the dry deposition of NH_3 is more efficient than that of NH_4^+; 2) not much NH_4^+ has formed yet near the source. At greater distances, the impact is slightly reduced by the dry deposition of NH_4^+, of which the changes have an opposite sign compared to dry deposition of NH_3. This is the case where the crossover point for NH_3 has been passed. For NH_x dry deposition, this combination of impacts results in a relative difference near the source from around 140% for increased $v_{d,pri}$ and –70% for decreased $v_{d,pri}$. At some distance from the source a crossover point is reached. This occurs at shorter distances for increased $v_{d,pri}$ than for decreased $v_{d,pri}$. Beyond the crossover point the sensitivity increases such, that at 400 km the impact varies between about –65% for increased $v_{d,pri}$ (crossover point just before 100 km) and a few percent for decreased $v_{d,pri}$ (crossover point not yet reached for the strongest decrease in $v_{d,pri}$).
For the industrial stack and the motorway the impact of $v_{d,pri}$ on the dry deposition of NO_x dominates the impact on the dry deposition NO_y. Near the source the feedback from concentration is much smaller than for NH_3. At greater distances the dry deposition of NO_3^-+HNO_3 becomes more important. The less efficient dry deposition velocity of NO_x compared to that of NH_3 explains that the crossover point of NO_y dry deposition is not reached within 400 km. Therefore, the impact of $v_{d,pri}$ does not switch sign within the distance range studied here, neither for the industrial stack, nor for the motorway. In the case of the industrial stack, the relative difference of the NO_y dry deposition compared to the reference run varies between about 260% (increased $v_{d,pri}$) and –75% (decreased $v_{d,pri}$) near the source and about 15% and –10% at 400 km. The impact for the motorway is slightly smaller, with relative differences between 230% and –70% near the source, and between 5% and –8% at 400 km.

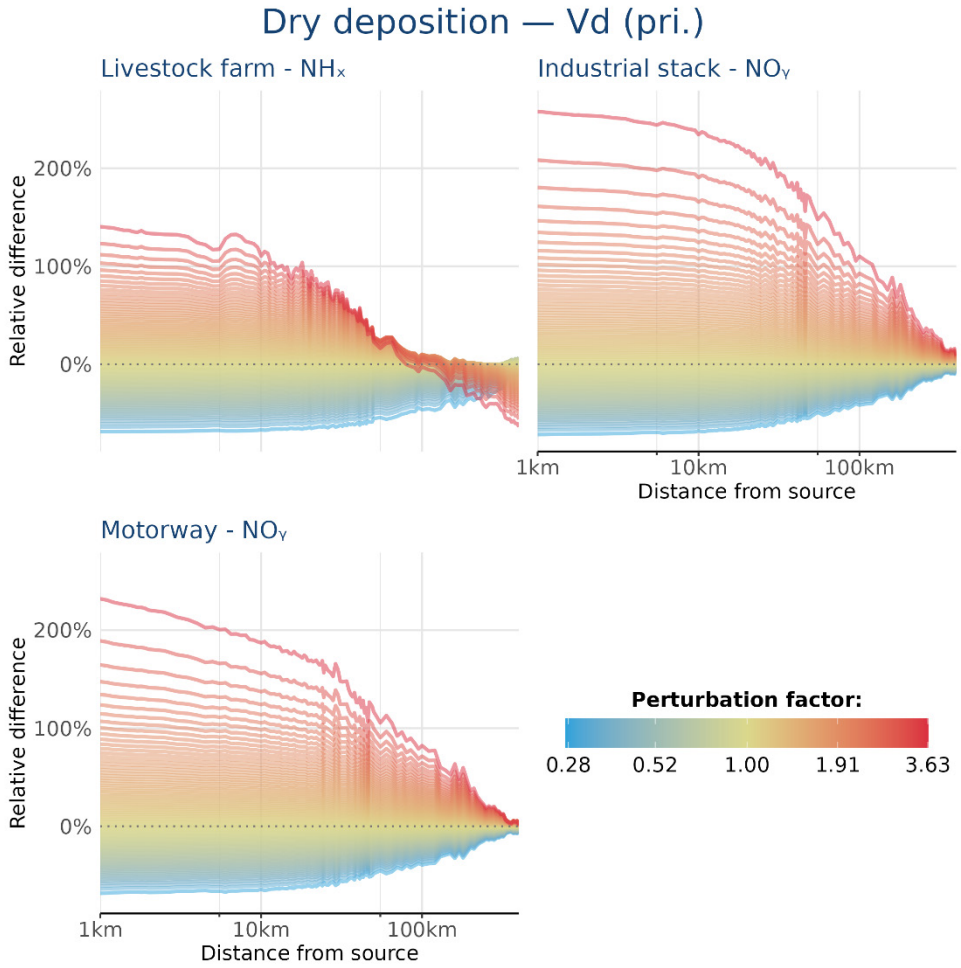

Figure A18 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying $v_{d,pri}$ both for the trajectory and at the receptor. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.5.3 *Wet deposition – impact of $v_{d,pri}$*

The combined effect of wet deposition of primary and secondary material (NH_x and NO_y, respectively) is depicted in Figure A19. Decreased wet deposition is obtained with increased $v_{d,pri}$ and vice versa. For the livestock farm variations near the source range between about –30% and 12% for increased and decreased $v_{d,pri}$, respectively, and between about –75% and 90% at 400 km. The sensitivity is much smaller for the industrial stack, for which wet deposition is nearly insensitive near the source. The sensitivity increases with distance, to yield differences between about –23% and 9% at 400 km. The motorway shows the combined impact of source segments with varying distances to the receptor, which gives variations between about –10% and 4% near the source, up to –30% and just over 10% at 400 km. Since the wet deposition process per se is not affected by $v_{d,pri}$, the wet deposition more or less follows the variation in the atmospheric concentration, which is this case represents the mass in the atmospheric column rather than the concentration at a specific height. With increased $v_{d,pri}$ more material is removed from the atmosphere, such that there is

less wet deposition of the primary material. Since less material can be converted to the secondary material, the same is true for wet deposition of the secondary material. The opposite occurs for decreasing $v_{d,pri}$. Note that for the industrial stack for wet deposition of secondary material the variations of concentration differences in the first tens of kilometres did not translate into significant differences in wet deposition (not shown).

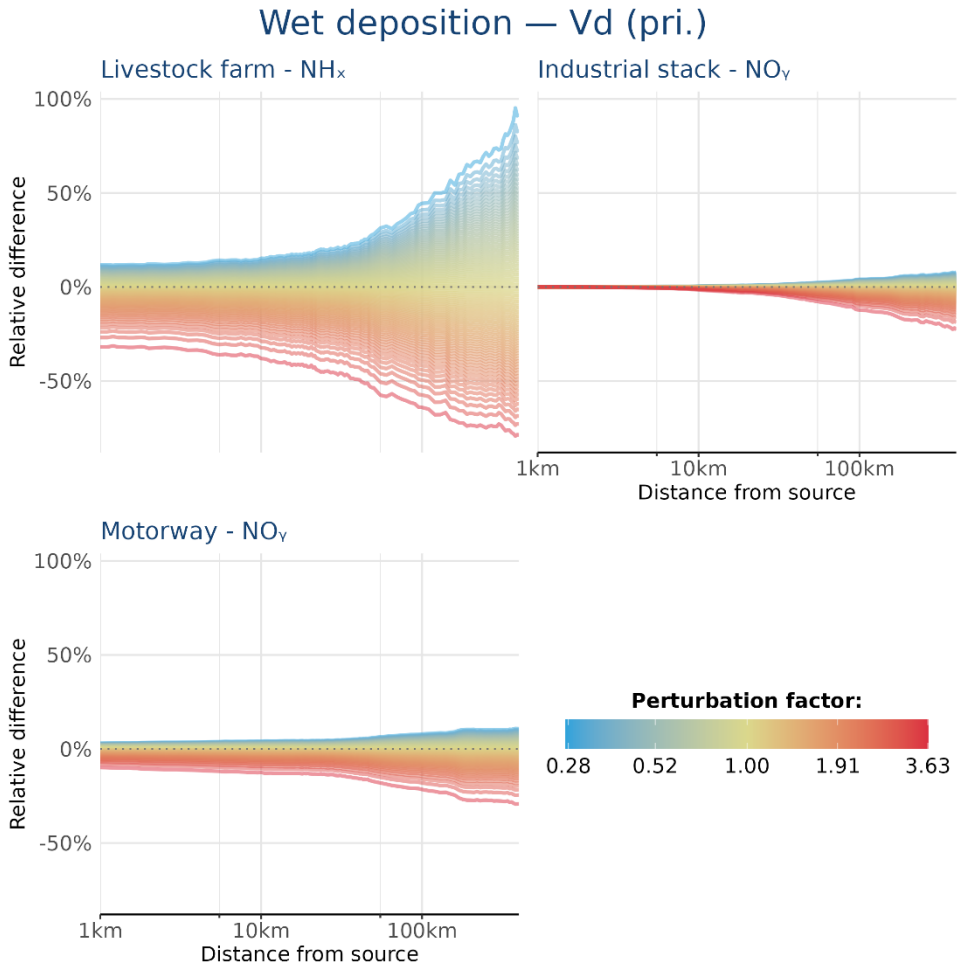

Figure A19 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying $v_{d,pri}$ both for the trajectory and at the receptor. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.5.4 *Total deposition – impact of $v_{d,pri}$*

Differences in total deposition of NH_x and NO_y, respectively, are shown in Figure A20. For the livestock farm the present perturbations of $v_{d,pri}$ result in relative differences between 135% and –65% near the source, with increased and decreased $v_{d,pri}$. A switch in sign indicates a crossover point somewhere around 50 km. Relative differences then grow to values between –75% and 65% at 400 km.
For the industrial stack the relative differences near the source range between around 255% and –70% near the source. There is hardly any variation at 400 km, which indicates a crossover point around that distance, here just before 400 km for increased $v_{d,pri}$. For the motorway,

variation near the source is between about 225% and –70%, with a switch in sign just before 400 km and negative differences of around 15% for increased $v_{d,pri}$ and positive differences of only a few percent at 400 km.

Nearby the source the impact of changing $v_{d,pri}$ on total deposition closely follows the one on dry deposition. This indicates that here, dry deposition of primary material dominates the impact on total deposition. At some point, the compensation mechanism will have caused sufficient effects on source depletion to strongly reduce changes in total deposition. With distance the impact of the wet deposition gains importance. Recall that wet deposition always showed a decrease with increased $v_{d,pri}$, and the other way around. This causes a more rapid change of total deposition than of dry deposition towards the crossover point, that shifts to shorter distances from the source compared to distances of the crossover points for dry deposition. Because of this, the change of sign of the relative difference for NO_y deposition can now also occur within the distance domain studied here: at 400 km for the industrial stack and just under 400 km for the motorway.

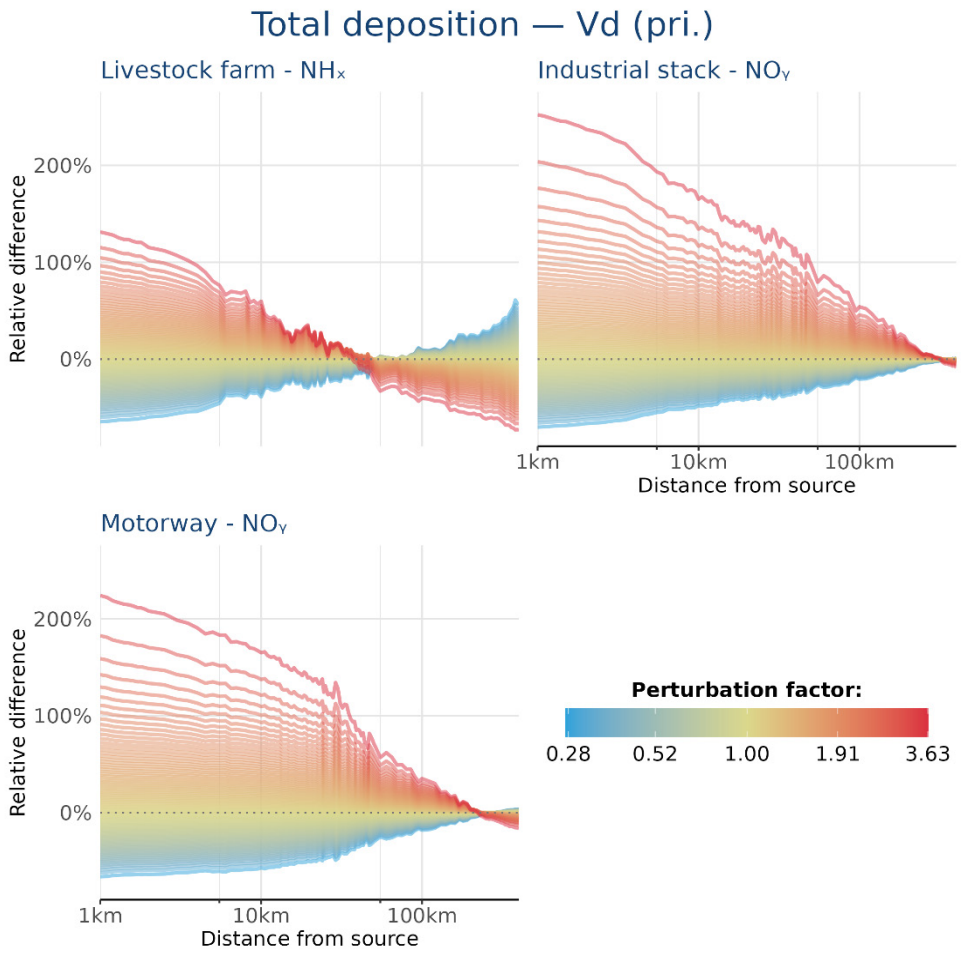

Figure A20 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying $v_{d,pri}$ both for the trajectory and at the receptor. Top left: livestock farm NH_x; top right: industrial NO_y; bottom: motorway NO_y.

1.6 Dry deposition velocity – secondary material $v_{d,sec}$

The dry deposition velocity of the secondary material ($v_{d,sec}$) was perturbed for the trajectory as well as at the receptor. The concentration and deposition of the primary material remain unaffected, so that the relative difference is always 0%. This is because we assume irreversible chemical conversion. Hence, we focus on secondary compounds only. Secondary material is affected as follows:

- $v_{d,sec}$ affects the concentration of secondary material, since it affects the amount of material removed from the atmosphere by source depletion over the trajectory.
- Initially, near the source, a change in $v_{d,sec}$ at the receptor leads to an almost equally large effect on dry deposition because effects of source depletion are small over short distances. Source depletion is more effective over longer distances, but the compensation effect due to the interaction between concentration and $v_{d,sec}$ applies to secondary material also (Hoogerbrugge et al., 2023). The effect may result in a crossover point, where the effect of $v_{d,sec}$ on the dry deposition of secondary material is reversed. This occurs at a distance from the source that depends on the magnitude of $v_{d,sec}$. Many complicating factors may obscure the effect, rendering the exact location of the crossover point rather unpredictable in practice.
- Since wet deposition is a strong function of the mass in the atmospheric column, the sensitivity of wet deposition to $v_{d,sec}$ roughly follows the one of concentration.

1.6.1 Concentration – impact of $v_{d,sec}$

Increasing $v_{d,sec}$ decreases concentrations of the secondary material due to the impact on source depletion (Figure A21). The opposite is true for decreasing $v_{d,sec}$. The impact is negligible near the source for the livestock farm and the industrial stack and increases with distance. For the motorway the impact is slightly higher near the source (between –7% and 2% for increased and decreased $v_{d,sec}$ respectively) due to the combined effect of multiple source segments with varying distances to the receptor. At 400 km relative difference have increased and vary between about –30% and 15% for increased and decreased $v_{d,sec}$, respectively, for NH_4^+ in the case of the livestock farm. For NO_3^-+HNO_3 from the industrial stack, differences are larger at 400 km and range between about –45% and 25%. In the case of the motorway, at this distance differences up to about –45% and 27% are obtained for the concentration of NO_3^-+HNO_3.

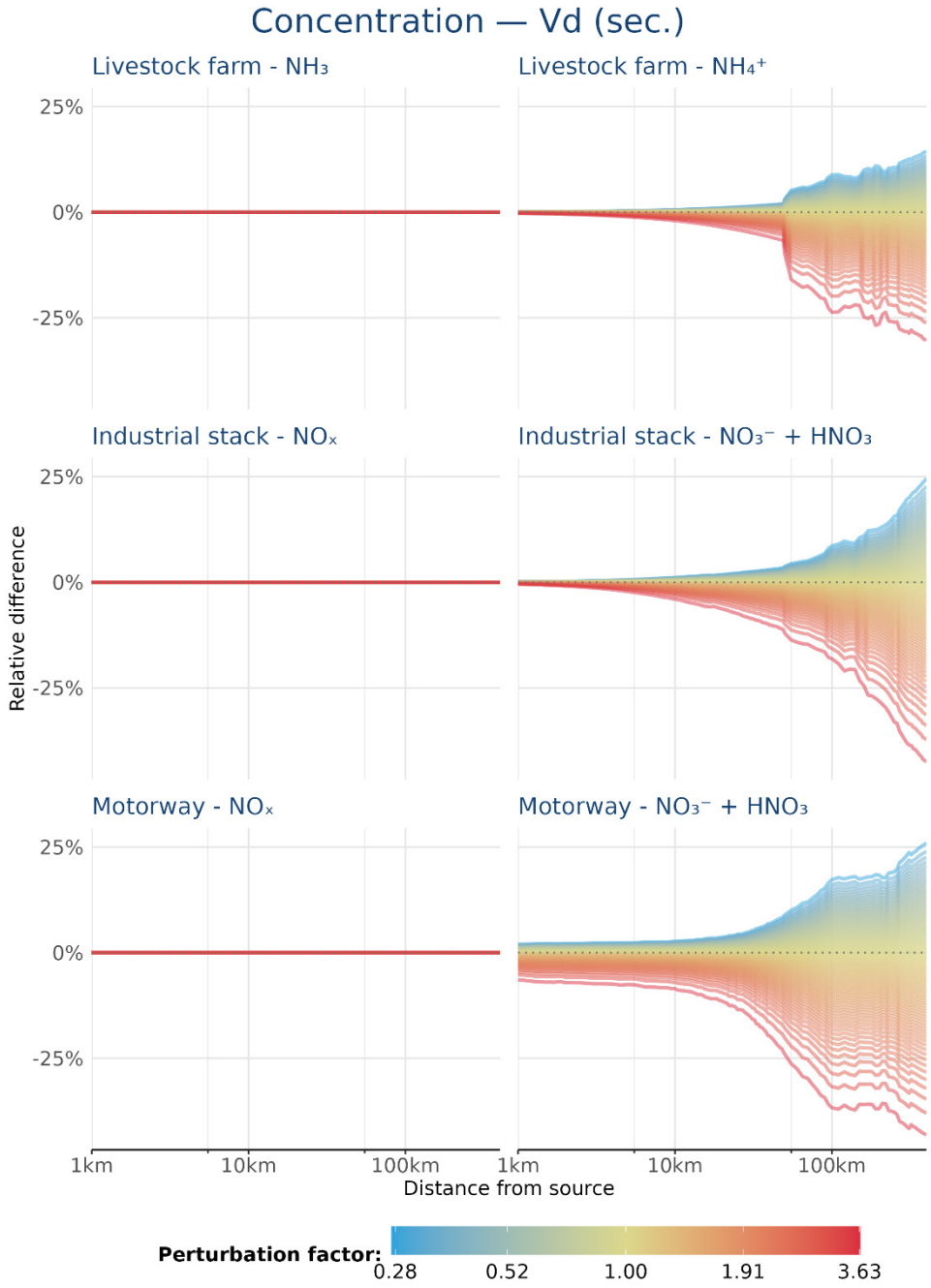

Figure A21 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying $v_{d,sec}$ both for the trajectory and at the receptor. Top row: livestock farm NH_3 and NH_4^+, middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^- +HNO_3.

1.6.2 *Dry deposition – impact of $v_{d,sec}$*

Impacts of $v_{d,sec}$ on dry deposition are shown in Figure A22. Since perturbation of $v_{d,sec}$ has no impact on the dry deposition of primary compounds, all changes in the combined dry deposition, that is, of NH_x and NO_y, respectively, must be due to changes in the secondary compounds. Hence, close the source the relative differences are small.

This is to be expected because at short distance the combined dry deposition will be strongly dominated by the dry deposition of the primary compounds, due to their abundance relative to secondary compounds. Note that the *relative* changes in dry deposition of secondary compounds per se may be large near the source (cf. Section 1.4).
In the case of the livestock farm the relative differences in NH_x increase with distance up to about 30% (increased $v_{d,sec}$) and just over –15% (decreased $v_{d,sec}$) at 400 km. At longer distances there is enough NH_4^+ available to significantly affect the NH_x dry deposition that also includes the unchanged NH_3 deposition, even if the latter is quite efficient. The increased $v_{d,sec}$ systematically results in increased dry deposition. This indicates that the impact on the dry deposition due to altered $v_{d,sec}$ dominates over the reduced concentration due to more effective source depletion. The changes increase with distance, without a noticeable effect of the feedback between NH_4^+ concentration and dry deposition. This is likely due to very low $v_{d,sec}$ of this compound.
By contrast, $v_{d,sec}$ of NO_3^-+HNO_3 is much higher. Results are comparable for the industrial stack and the motorway. Here, the response of NO_y dry deposition to $v_{d,sec}$ increases with distance, like for NH_x dry deposition, but an opposite sign is obtained. Increased $v_{d,sec}$ leads to decreased dry deposition of NO_y, reaching a relative difference of –33% at 400 km. Decreased $v_{d,sec}$ leads to increased dry deposition of NO_y, with a relative difference of up to 20% at 400 km. As such, it can be concluded that for NO_3^-+HNO_3 the source depletion effect on the concentration is quite strong, due to the efficient deposition. With the more efficient dry deposition of NO_3^-+HNO_3 compared to that of NH_4^+, increasing $v_{d,sec}$ leads to lower concentrations of NO_3^-+HNO_3 and vice versa. Hence the compensation effect becomes effective. Because the unchanged $v_{d,pri}$ of NO_x is low, this effect of $v_{d,sec}$ is strong enough to dominate the changes in NO_y dry deposition.

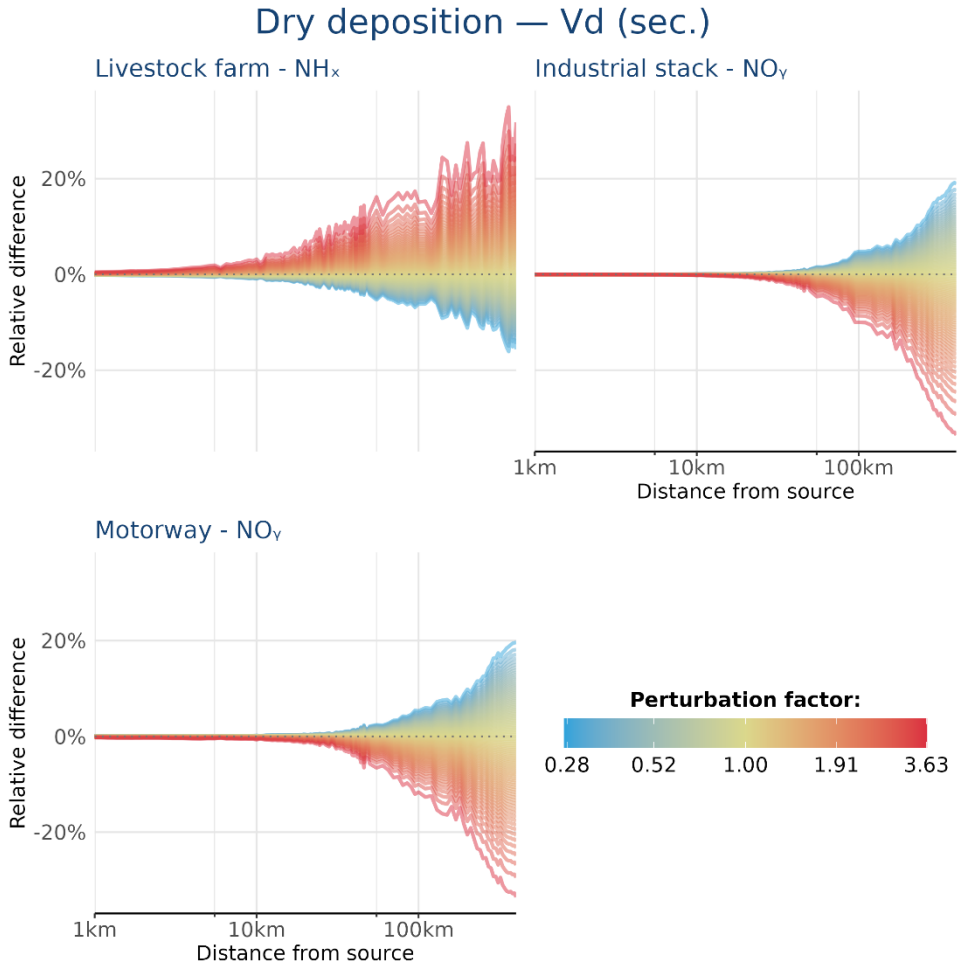

Figure A22 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying $v_{d,sec}$ both for the trajectory and at the receptor. Top left: livestock farm NH_x; top right: industrial NO_y; bottom: motorway NO_y.

1.6.3 *Wet deposition – impact of $v_{d,sec}$*

Impacts of $v_{d,sec}$ on wet deposition are shown in Figure A23. The relative difference in wet deposition of the secondary material due to altered $v_{d,sec}$ follows, in general, the relative difference in the concentration of the secondary material. This effect, related to the secondary compounds, is sufficiently large to have an impact of the wet deposition of NH_x and NO_y, respectively. Since with increased $v_{d,sec}$ the concentration of secondary material decreases, a decreased wet deposition is found. The opposite is true for decreased $v_{d,sec}$. This is seen in the wet deposition of both NH_x and NO_y.

Also for wet deposition the impact of $v_{d,sec}$ is small near the source. For the livestock farm and industrial stack relative changes are almost negligible, but the differences amount to up to a few percent for the motorway. The relative impact increases with distance, up to between about –15% and 6%, for increased $v_{d,sec}$ and decreased $v_{d,sec}$, in the case of the livestock farm at 400 km. For the industrial stack and motorway relative changes range between about –35% and 18% at that distance.

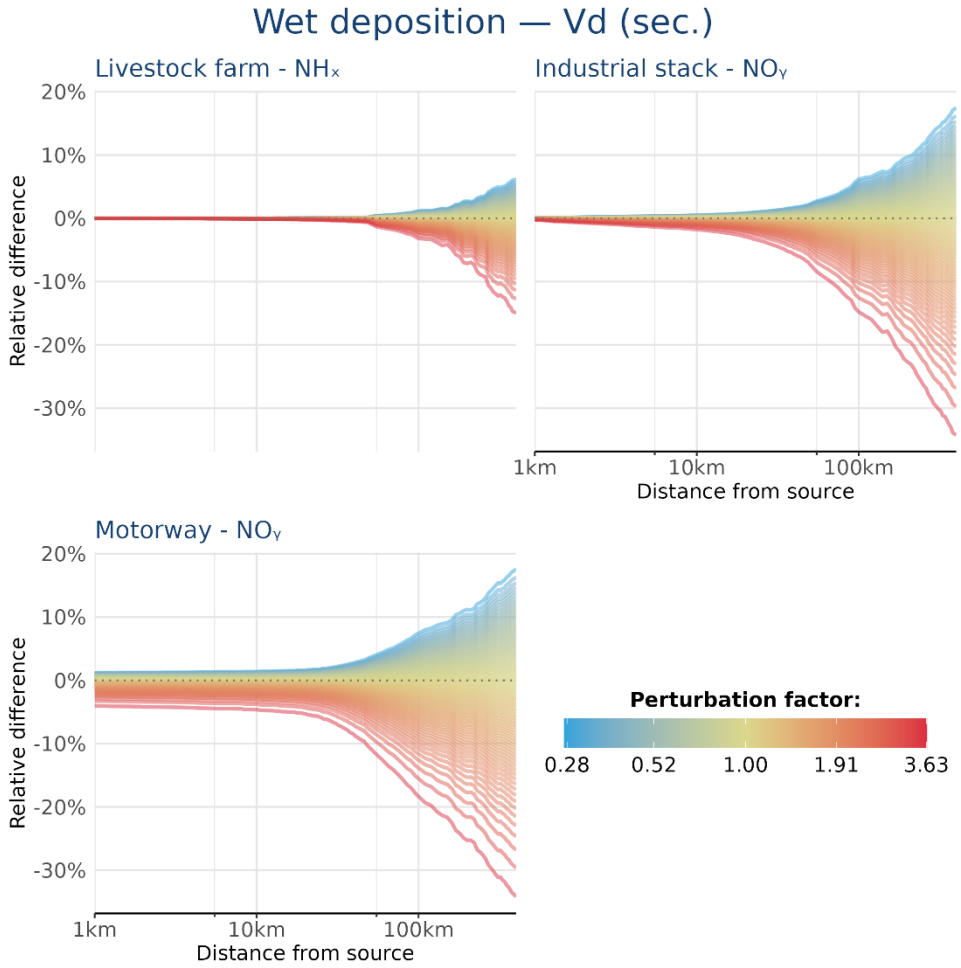

Figure A23 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying $v_{d,sec}$ both for the trajectory and at the receptor. Top left: livestock farm NH_x; top right: industrial NO_y; bottom: motorway NO_y.

1.6.4 *Total deposition – impact of $v_{d,sec}$*

The impact of changes in $v_{d,sec}$ on the total deposition (Figure A24) of NH_x does not show a straightforward variation with distance from the source. Though in general, again, the impact near the source is small, a consistently increasing effect is not obtained here, but rather a noisy pattern with strong differences among the various receptor distances. The effect of dry deposition and wet deposition counteract each other. The increase found for dry deposition with increased $v_{d,sec}$ can become a decrease where wet deposition becomes more dominant. This results in a shift in pattern with sometimes higher and sometimes lower total deposition with increased $v_{d,sec}$. The same is true for decreased $v_{d,sec}$. However, on average the relative impact is minor, being only tens of a percent near the source up to a few percent, up to ~8% for increased $v_{d,sec}$ and ~ −3% for decreased $v_{d,sec}$, over the full 400 km.
Contrary to the livestock farm, the more straightforward patterns for dry and wet deposition of NO_y, which showed the same sensitivity pattern, renders the result for total deposition of NO_y for the industrial stack and motorway case rather straightforward. With increased $v_{d,sec}$ decreased

total NO_y deposition is modelled, and vice versa. This suggests that the source depletion effect is most dominant. The impact is negligible near the source and increases with distance. The relative differences at 400 km are between −35% and 18% with increased and decreased $v_{d,sec}$, respectively, for both the industrial stack and the motorway.

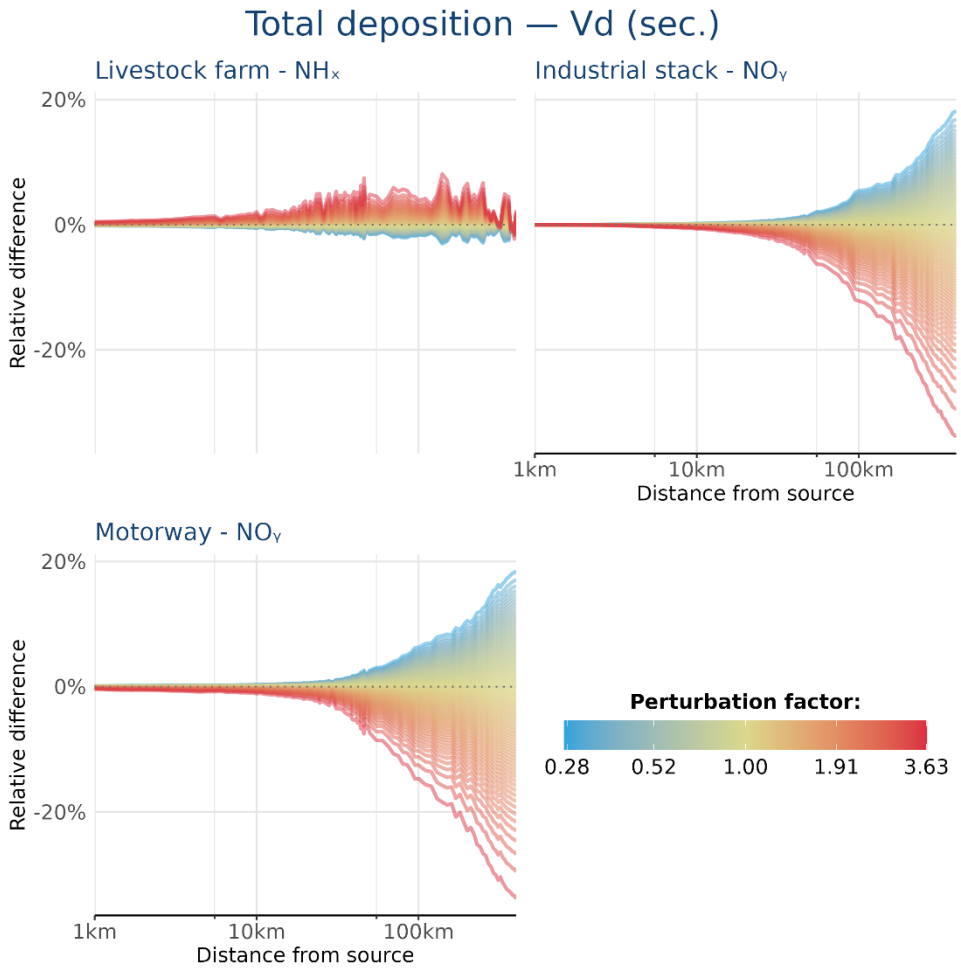

Figure A24 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying $v_{d,sec}$ both for the trajectory and at the receptor. Top left: livestock farm NH_x, top right: industrial stack NO_y, bottom: motorway NO_y.

1.7 Washout scavenging rate – primary material $\Lambda_{w,pri}$

Washout describes removal of pollutants by falling precipitation below the cloud's baseline. This can especially be important near pollutant sources, where there has been no interaction of the pollutants with clouds yet. For the sensitivity analysis, the scavenging rate or scavenging coefficient of the washout process of wet deposition of the primary material ($\Lambda_{w,pri}$) was perturbed. This has the following impacts:

- A change in $\Lambda_{w,pri}$ affects the concentration of primary material since it affects the amount of material that will be removed from the atmosphere due to source depletion over the trajectory.
- Dry deposition will roughly follow the changed concentration pattern.

- Initially, a given relative change in $\Lambda_{w,pri}$ leads to an approximately equally large effect on wet deposition. However, the wet deposition also affects concentration and this reduces the initial effect of $\Lambda_{w,pri}$ on the wet deposition.
- Concentration of the secondary material is impacted because less (increased $\Lambda_{w,pri}$) or more (decreased $\Lambda_{w,pri}$) primary material can be converted to the secondary material. The scavenging rate of the secondary compounds is not adjusted in the runs described in this Section.

Note that in OPS the rainout process of NO_x is assumed to be of minor importance because primary emitted NO_x species have a low water solubility. Hence, the below-cloud scavenging is set to 0 for NO_x in OPS. As a result, there is no variation in the simulations of the industrial stack and motorway when varying $\Lambda_{w,pri}$. Thus, the relative difference is always 0%. Therefore, only the impact for the results on the livestock farm will be discussed.

1.7.1 Concentration – impact of $\Lambda_{w,pri}$

Increasing $\Lambda_{w,pri}$ results in a decrease of concentration since more material will be removed from the atmosphere due to source depletion (Figure A25). The opposite is true for decreased $\Lambda_{w,pri}$. Though initially it was expected that this would only have an effect near the source, where the emitted ammonia has not yet been in contact with the clouds, it is seen that the signal progresses to greater distances as well. This may be due to the impact on source depletion, which has an effect over greater distances as well. Furthermore, for some meteorological classes in OPS there can still be some washout at greater distances. However, note that the impact on NH_3 concentration is small, with maximum relative differences of 0.95% only, and often even less. The wet deposition of ammonium is not impacted directly, since $\Lambda_{w,sec}$ does not change. As such, the pattern for NH_4^+ follows the one of the NH_3 concentration, but with smaller sensitivity. Maximum differences of about 0.3% are attained over the 400 km distance on average.

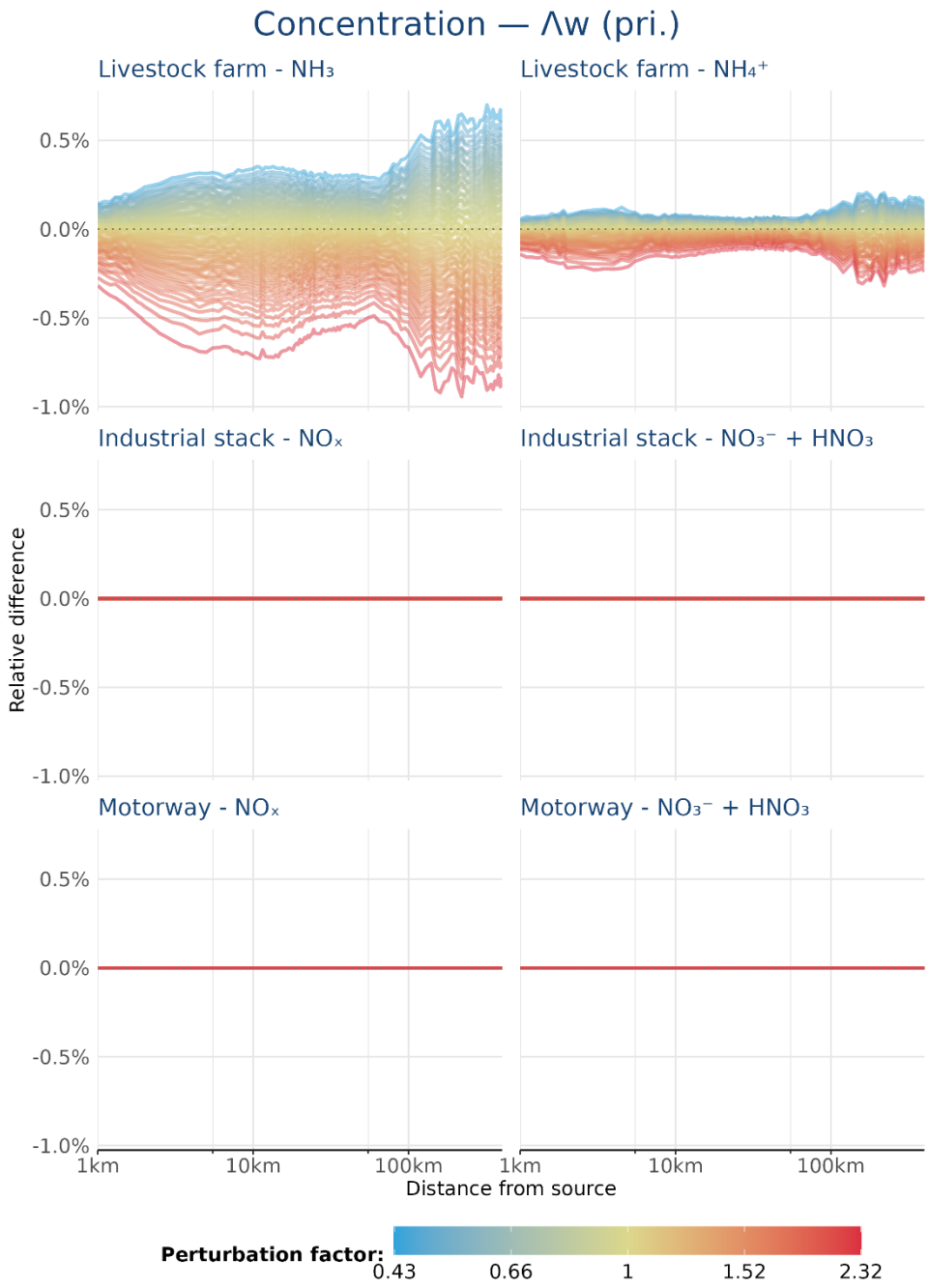

Figure A25 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying $\Lambda_{w,pri}$. Top row: livestock farm NH_3 and NH_4; middle row: industrial stack NO_x and NO_3^-+ HNO_3; bottom row: motorway NO_x and NO_3^-+ HNO_3.

1.7.2 *Dry deposition – impact of $\Lambda_{w,pri}$*

Impacts of $\Lambda_{w,pri}$ on dry deposition are shown in Figure A26. Since the dry deposition process is not impacted when altering $\Lambda_{w,pri}$, the relative difference in dry deposition of NH_x approximately follows the combined impact on the concentrations of NH_3 and NH_4^+ regarding the direction. A larger $\Lambda_{w,pri}$ results in a smaller dry deposition of NH_x and vice versa.

Again, the impact is small, though larger than for the individual concentrations of the primary and secondary compounds. The maximum difference amounts to about −1.7% for increasing $\Lambda_{w,pri}$ and ~0.85% for decreasing $\Lambda_{w,pri}$.
Regarding the variation over distance, the pattern is somewhat more 'spikey'. This is probably caused by the varying land use along the various trajectories which becomes more noticeable for these small relative differences.

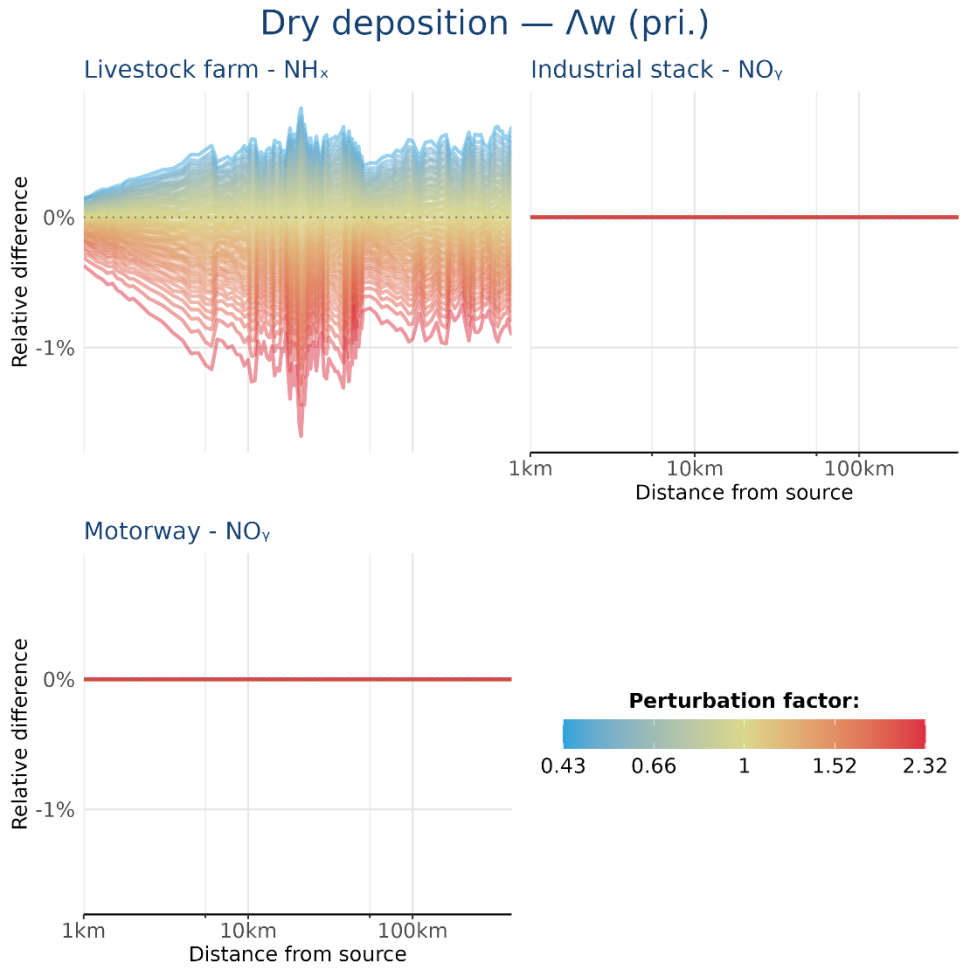

Figure A26 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying $\Lambda_{w,pri}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.7.3 *Wet deposition – impact of $\Lambda_{w,pri}$*

Increased $\Lambda_{w,pri}$ leads to an increased wet deposition, and vice versa (Figure A27). Near the source, a given relative change in $\Lambda_{w,pri}$ results in an approximately equally large relative impact on wet deposition of NH_x. At small distances the impact of $\Lambda_{w,pri}$ on the local wet deposition is much larger than the impact of $\Lambda_{w,pri}$ on the concentration via source depletion. Also, the washout process is much more important than

further away, where rainout becomes more important. For these reasons, the impact of $\Lambda_{w,pri}$ on the wet deposition of NH_x is highest near the source, between almost 100% and –45% for increased and decreased $\Lambda_{w,pri}$, respectively. The sensitivity then reduces quickly, and becomes negligible beyond 100 km. Because the wet deposition of NH_4^+ is left unchanged, the impact of varying $\Lambda_{w,pri}$ on wet deposition of NH_4^+ approximately follows the atmospheric concentration of this compound. However, the effect of $\Lambda_{w,pri}$ on NH_4^+ concentration remains within ±1% over the 400 km (see Figure A25) and the impact on wet deposition of NH_4^+ is thus small as well. Hence, the relative change in wet deposition of NH_x is dominated by the relative change in wet deposition of NH_3, since NH_3 is the most abundant material near the source.

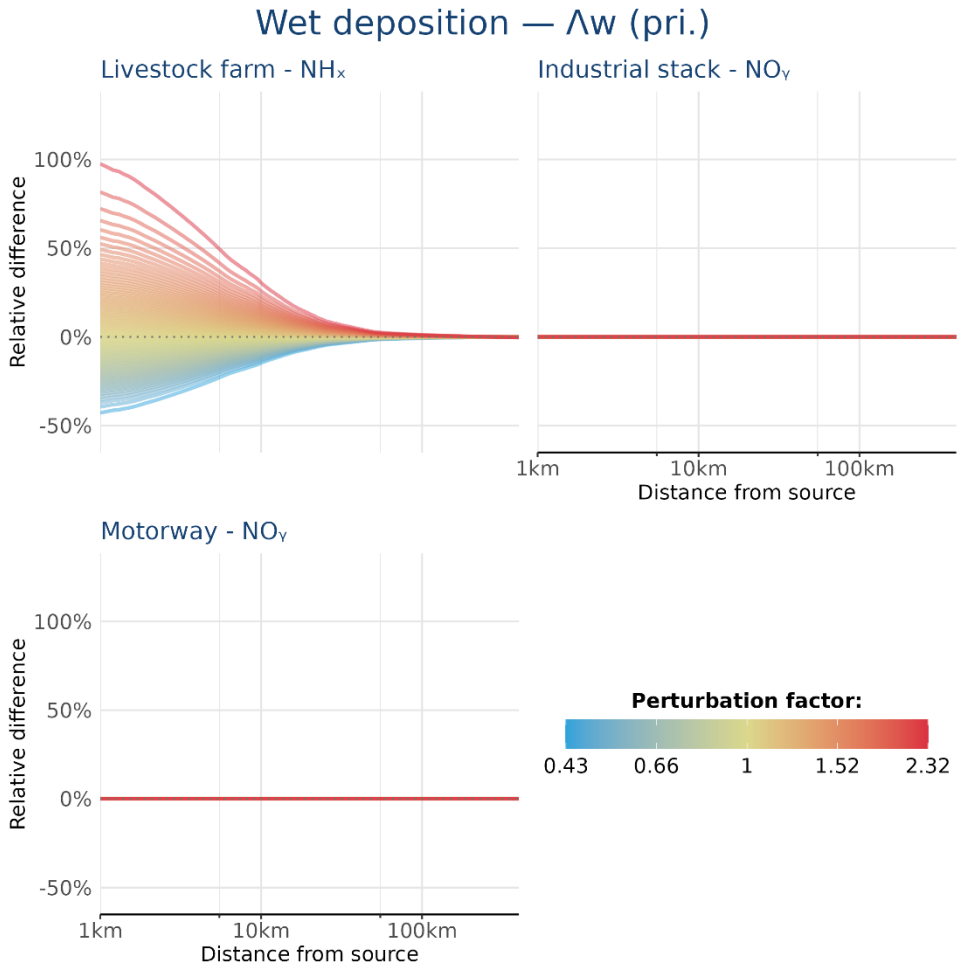

Figure A27 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying $\Lambda_{w,pri}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.7.4 *Total deposition – impact of $\Lambda_{w,pri}$*

Impacts of $\Lambda_{w,pri}$ on total deposition are shown in Figure A28. The total deposition of NH_x is the combination of its dry and wet deposition. Though the sensitivity of dry deposition to $\Lambda_{w,pri}$ was found to be small,

it still plays an important role in the sensitivity of total deposition. It is found that it clearly reduces the impact of wet deposition near the source. The relative contribution of wet deposition changes to differences in total deposition does gain importance with distance. As such, the impact of $\Lambda_{w,pri}$ on the total deposition peaks around 5 – 10 km, instead of directly at the source. The maximum effect amounts to about 17% for increased $\Lambda_{w,pri}$ and −8% for decreased $\Lambda_{w,pri}$. The impact then decreases and switches sign at ~150 km from the source. This is indicative of the compensation mechanism due to the interaction between concentration and deposition. The small effect on wet deposition with distance is counteracted by the small effect of opposite sign on the dry deposition, via the impact of source depletion on concentration. Between the crossover point and 400 km the relative impact slightly increases again but remains less than ±1%.

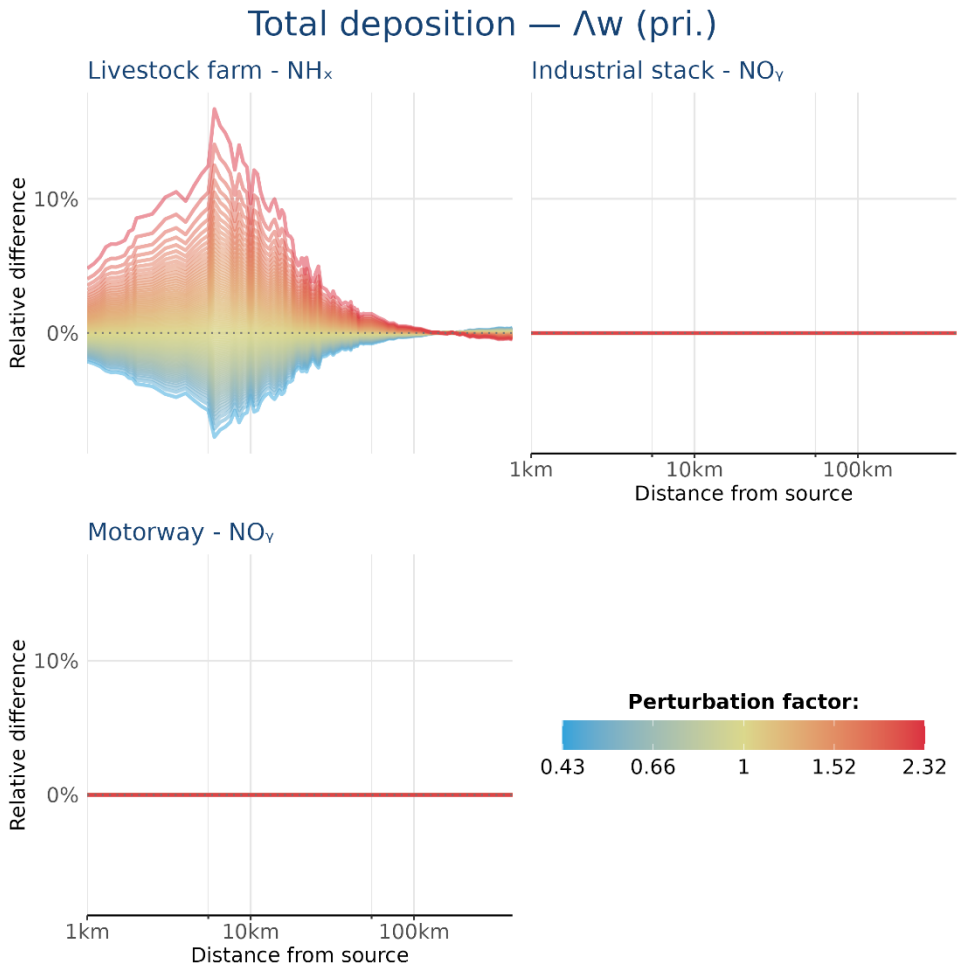

Figure A28 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying $\Lambda_{w,pri}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.8 Washout scavenging rate – secondary material $\Lambda_{w,sec}$

Perturbation of the scavenging rate of the washout process of the secondary material ($\Lambda_{w,sec}$) only has impact on the secondary material:

- A change of $\Lambda_{w,sec}$ affects the amount of material that will be removed from the atmosphere due to source depletion over the trajectory and hence the concentration.
- Dry deposition will roughly follow the changed concentration pattern of the secondary material.
- Initial impacts of $\Lambda_{w,sec}$ on the wet deposition of secondary material are expected to be comparatively large. However, the wet deposition also affects concentration and this reduces the initial effect of $\Lambda_{w,sec}$.

The concentration and deposition of the primary material are not affected. As such, relative differences in the concentration of the primary material are not discussed, since relative difference is always 0%.

1.8.1 Concentration – impact of $\Lambda_{w,sec}$

A decrease in $\Lambda_{w,sec}$ results in an increase of the concentration of secondary compounds since less material is removed from the atmosphere due to source depletion (Figure A29). The reverse applies to an increase of $\Lambda_{w,sec}$. For all source types the sensitivity of the concentration to $\Lambda_{w,sec}$ is small. Relative differences are maximally about plus or minus 0.5% for NH_4^+ in the livestock farm case, only half of that for NO_3^-+HNO_3 in the industrial stack case, and less than about 1.5% for NO_3^-+HNO_3 in the motorway case.

For the livestock farm the impact is steady, between about –0.15% and 0.07% for the first 50 km and then increases with distance. For the high source of the industrial stack the impact shows a minimum at about 50 km. An explanation for this may be that for very stable conditions, part of the emission initially occurs above the mixing layer, and is fumigated into this layer at greater distance. When this occurs, a small change in NO_3^-+HNO_3 has a smaller relative impact. For the motorway the impact on the relative difference in the concentration of NO_3^-+HNO_3 increases steadily with increasing distance, up to about 200 km, and then decreases again. Since the motorway source contains many source segments, the concentration at a given distance will be affected by source segments at various distances from the receptor. This may explain the difference in variation over distance between the motorway and the livestock farm case, which are both low sources.

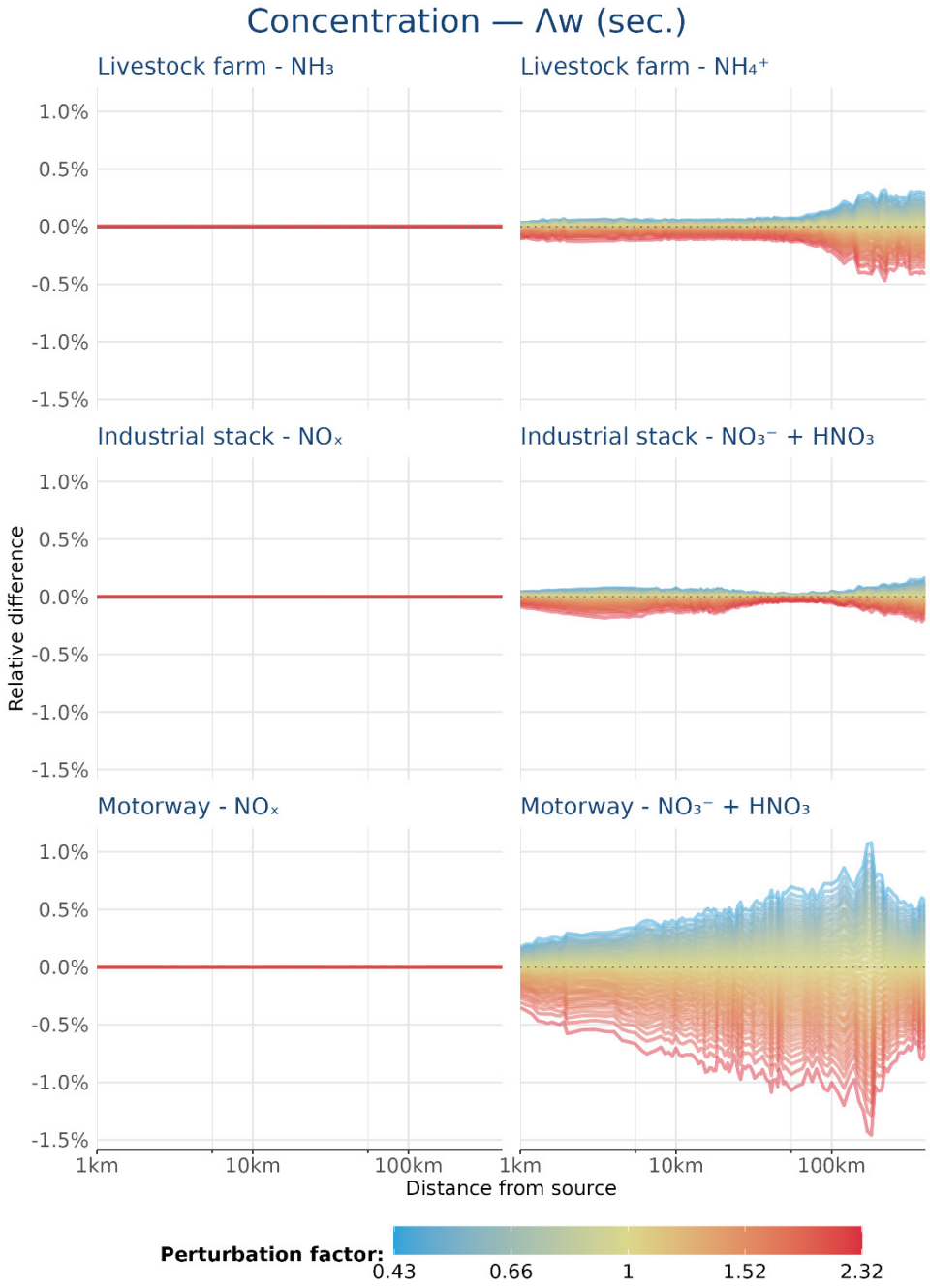

Figure A29 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying $\Lambda_{w,sec}$. Top row: livestock farm NH_3 and NH_4; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.8.2 *Dry deposition – impact of $\Lambda_{w,sec}$*

Since the dry deposition process *per se* is not impacted by $\Lambda_{w,sec}$, the relative difference in dry deposition of the secondary material with respect to the reference run (Figure A30) follows the impact on its concentration. A decrease in dry deposition is seen with increased $\Lambda_{w,sec}$, and vice versa. This is due to the impact on the source depletion, which affects concentration. The dry deposition of the NH_x and NO_y dry

deposition, respectively, combines the effect on dry deposition of the primary and secondary material. It therefore behaves slightly different than the concentrations of the secondary material only. Since the primary material is more abundant and not as much secondary material is formed yet on the smaller distance, the impact of varying $\Lambda_{w,sec}$ is very small near the source and increases with distance for all source types. Again, the impact is very small, and the highest for the motorway case with variations up to –0.65% and 0.5% for increased and decreased $\Lambda_{w,sec}$ respectively at 400 km.

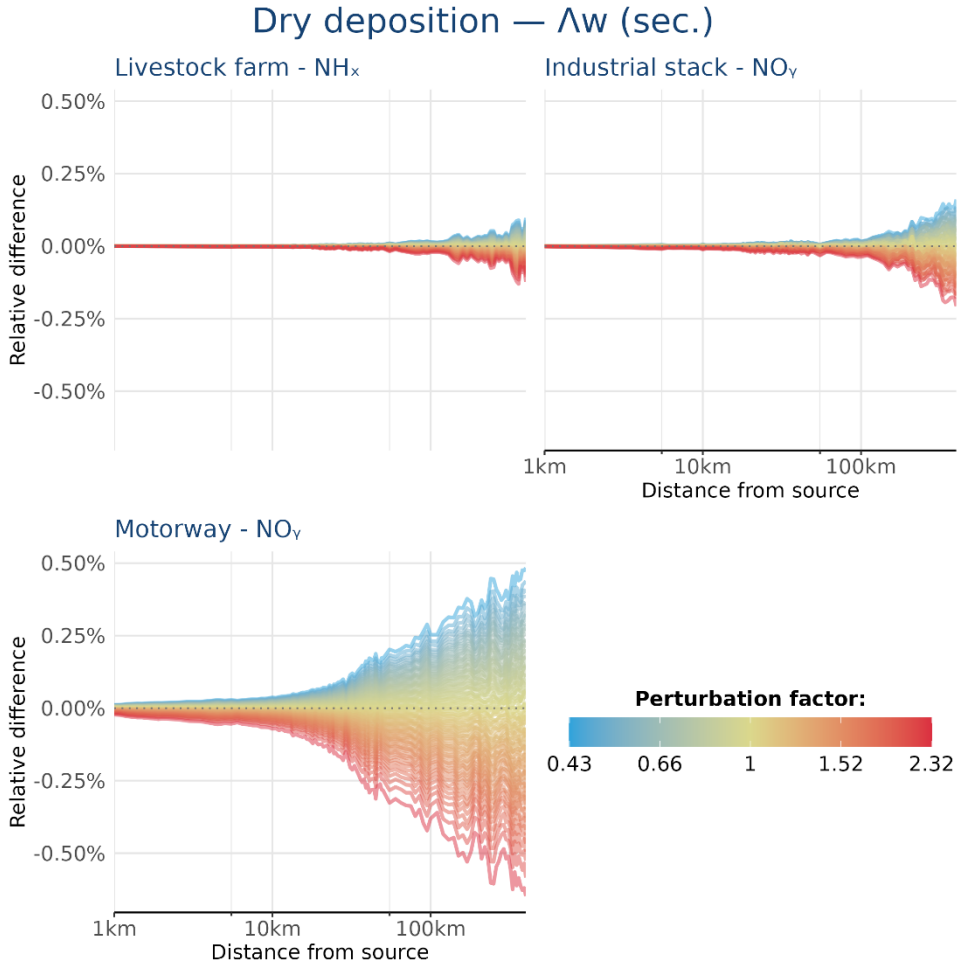

Figure A30 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying $\Lambda_{w,sec}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.8.3 *Wet deposition – impact of $\Lambda_{w,sec}$*

For wet deposition an increase of wet deposition of NH_x and NO_y at the receptor is seen for all sources when increasing $\Lambda_{w,sec}$, and vice versa (Figure A31). Clearly, the direct impact of $\Lambda_{w,sec}$ on the local deposition dominates over the impact of $\Lambda_{w,sec}$ related to source depletion. The impact is highest at the source in the case of the industrial stack. For the livestock farm and the motorway, it is highest at a few kilometres from the source, and then decreases with distance where the role of washout gradually vanishes. Note that for the impact on NH_4^+ deposition

is highest already close to the source (not shown). However, since the contribution of wet deposition of NH_3 is by far larger and is not affected by $\Lambda_{w,sec}$, the contribution from the effect of NH_4^+ is strongly reduced. For the motorway the variation in wet deposition of NO_3^-+HNO_3 is similar to the total wet deposition of NO_y, which again is likely explained by the impact of the various source segments with varying distance to the receptor. For the industrial stack, the impact on NO_3^-+HNO_3 is also largest at the source (not shown), but this is not counteracted by washout of NO_x since this is not taken into account in OPS.
The impact is smallest for the livestock case with relative differences between maximum 0.9% and −0.5% over the 400 km distance for increased and decreased $\Lambda_{w,sec}$, respectively. The impact is somewhat larger for the industrial stack, with a maximum of a 6.5% increase in relative difference with increased $\Lambda_{w,sec}$ and a 3% decrease with decreased $\Lambda_{w,sec}$ near the source. The relative impact then reduces to a few tens of a percent after 100 km. The impact is highest for the motorway case with up to 15% increase with increased $\Lambda_{w,sec}$ and up to an 8% decrease with decreased $\Lambda_{w,sec}$ around 5 km, reducing to less than ±1% at 400 km.

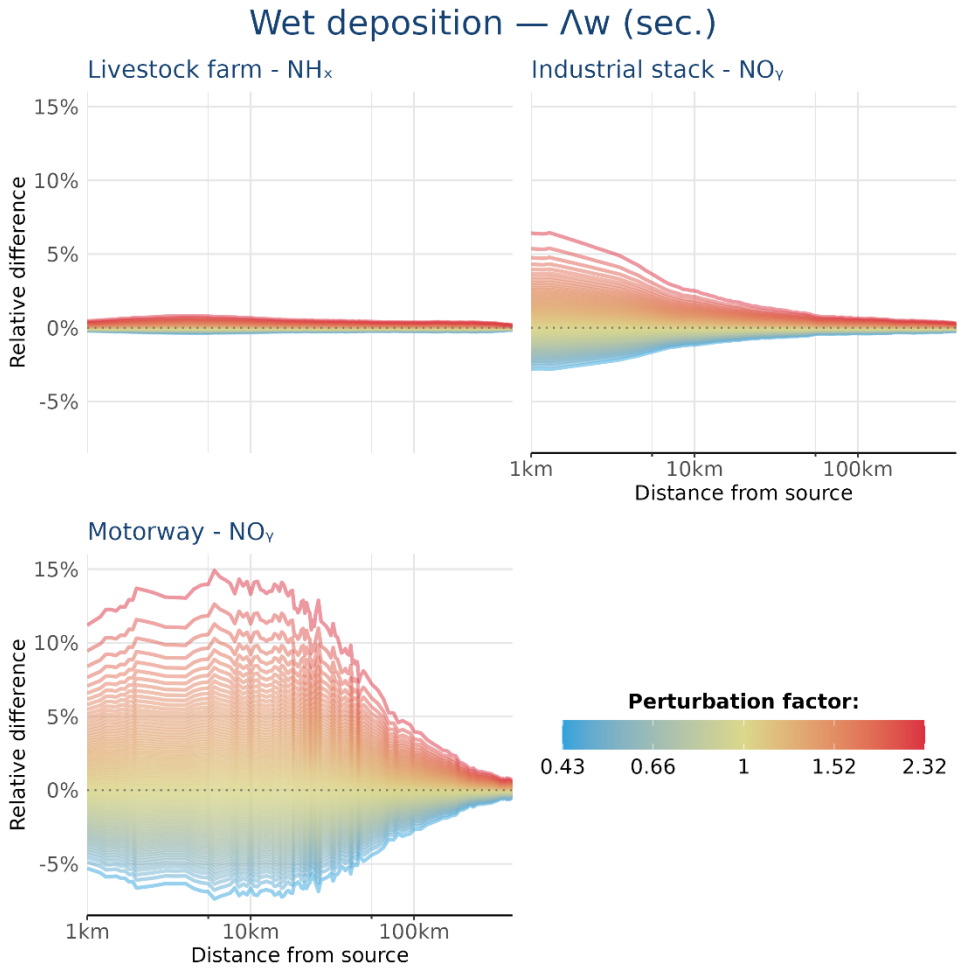

Figure A31 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying $\Lambda_{w,sec}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.8.4 Total deposition – impact of $\Lambda_{w,sec}$

The sign of the relative difference in total deposition of NH_x (livestock farm) and NO_y (industrial stack and motorway) compared to the reference run (Figure A32) corresponds to the one of the wet deposition at the receptor. An increase in total deposition is obtained with increased $\Lambda_{w,sec}$, and vice versa. Like for wet deposition, the impact is smallest for the livestock farm case, slightly larger for the industrial stack and largest for the motorway. In all cases the impact remains small, up to maxima of 0.3% for the livestock farm, 0.8% for the industrial stack, and 3% for the motorway over the 400 km.

The aforementioned results suggest a dominant role of the sensitivity of wet deposition to $\Lambda_{w,sec}$. However, dry deposition is more important near the source. This contribution is hardly affected by $\Lambda_{w,sec}$, especially in the vicinity of the source. Whereas the sensitivity of wet deposition was generally larger near the source, the contribution from dry deposition reduces the relative changes in total deposition in the first few kilometres. The sensitivity of total deposition then increases with distance up to about 5 – 50 km. At greater distances, the contribution from wet deposition grows, such that the sensitivity pattern of wet deposition is clearly recognised. Nevertheless, the reversed impact on the dry deposition does reduce the effects somewhat.

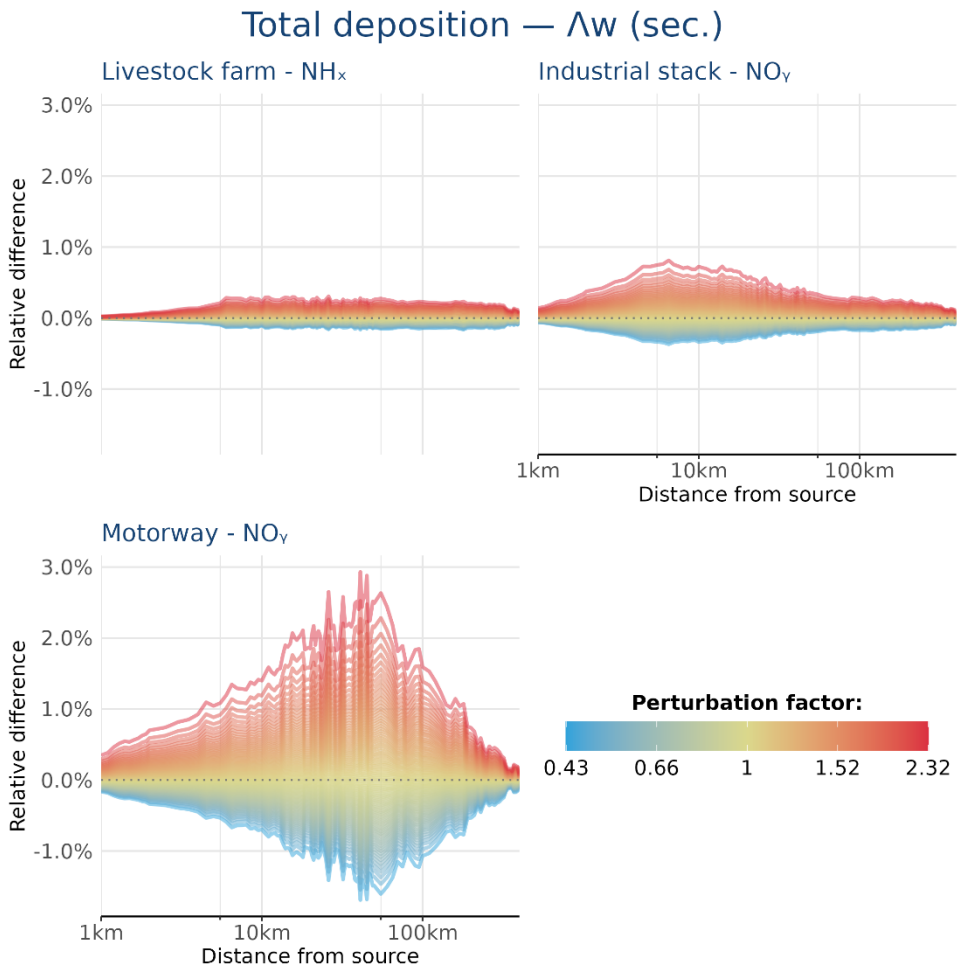

Figure A32 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying $\Lambda_{w,sec}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.9 Rainout scavenging rate – primary material $\Lambda_{r,pri}$

Rainout describes the uptake of pollutant by precipitation being formed within clouds. In OPS this is parameterised with a bulk scavenging ratio which integrates all processes within and below the cloud. In contrast with washout, it is more important at the longer distances. Varying the scavenging rate for rainout of the primary material ($\Lambda_{r,pri}$) has the following impact:

- A change in $\Lambda_{r,pri}$ affects the concentration of primary material since it affects the amount of material that will be removed from the atmosphere due to source depletion over the trajectory.
- Dry deposition will roughly follow the changed concentration.
- Initially, a given relative change in $\Lambda_{r,pri}$ leads to an approximately equally large effect on wet deposition. However, the wet deposition also affects concentration and this reduces the initial effect of $\Lambda_{r,pri}$ on the wet deposition. This induces a feedback loop that may ultimately lead to a crossover point, where the effect of $\Lambda_{r,pri}$ on the wet deposition is reversed. This occurs at a distance from the source that depends on the

magnitude of $\Lambda_{r,pri}$. In practice however, many complicating factors may obscure the effect, rendering the location of the crossover point quite uncertain.

- Concentration of the secondary material is impacted because less (increased $\Lambda_{r,pri}$) or more (decreased $\Lambda_{r,pri}$) primary material can be converted to the secondary material. The scavenging rate of the secondary compounds is not adjusted in the runs described in this Section.

1.9.1 Concentration – impact of $\Lambda_{r,pri}$

For the primary material for all source types increasing $\Lambda_{r,pri}$ causes the concentration to decrease, and vice versa (Figure A33). This is the effect of the altered source depletion due to rainout. The concentration changes of the secondary compounds follow those of the primary compounds since less (increased $\Lambda_{r,pri}$) or more (decreased $\Lambda_{r,pri}$) primary material is available for chemical conversion. The relative impact is smaller than for the primary material, probably because $\Lambda_{r,pri}$ is not changed. For example, if the primary concentration increases due to a decreased $\Lambda_{r,pri}$, higher concentrations of secondary material are obtained. However, with the same source depletion factor for the secondary material, this will give more loss of the secondary material over the trajectory than in the reference case. This reduces the relative impact on the concentration of the secondary compound.
In general, the impact of $\Lambda_{r,pri}$ on concentration increases with distance, in accordance with the growing influence of rainout on the wet deposition. The maximum impact is largest for the livestock farm case with variations between ~ –9% and just over 20% at 400 km for NH_3, and between –2.5% and 6% for NH_4^+. This is somewhat smaller for the industrial stack and motorway, for which variations range between ~ –7% and 3% for NO_x concentration, and between about –3% and 1.5% for NO_3^-+HNO_3. The difference between the livestock farm versus the industrial stack and the motorway could be explained the higher solubility in water of NH_3 than of NO_x. This results in higher concentration ratios between water and air for NH_3, indicative of a higher efficiency of the rainout process for NH_3.

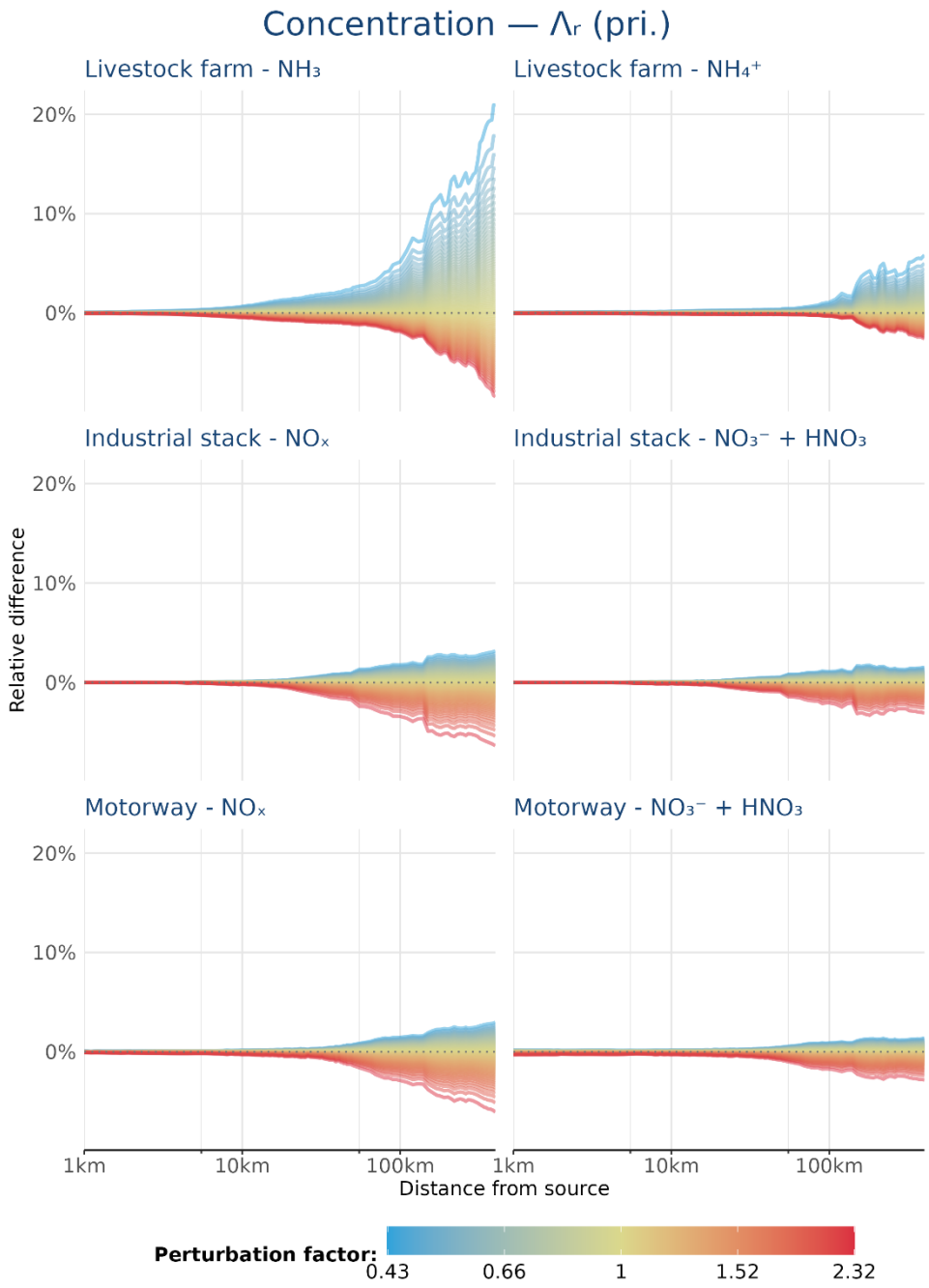

Figure A33 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying $\Lambda_{r,pri}$. Top row: livestock farm NH_3 and NH_4^+; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.9.2 *Dry deposition – impact of $\Lambda_{r,pri}$*

The dry deposition process *per se* is not affected by rainout. Therefore, the sensitivity of dry deposition to $\Lambda_{r,pri}$ (Figure A34) is indirect. As a result, the relative difference in dry deposition of the primary material follows the one in its concentration. The same is true for the secondary material. The sensitivity of dry deposition of NH_x in the case of the livestock farm and of NO_y for the industrial stack and motorway,

respectively, is negligible near the source. Relative changes increase with distance. Changes in dry deposition relative to the reference run vary between about –7% and 20% for NH_x (in- and decreased $\Lambda_{r,pri}$, respectively), and between ~ –4% and 2% for NO_y at 400 km.

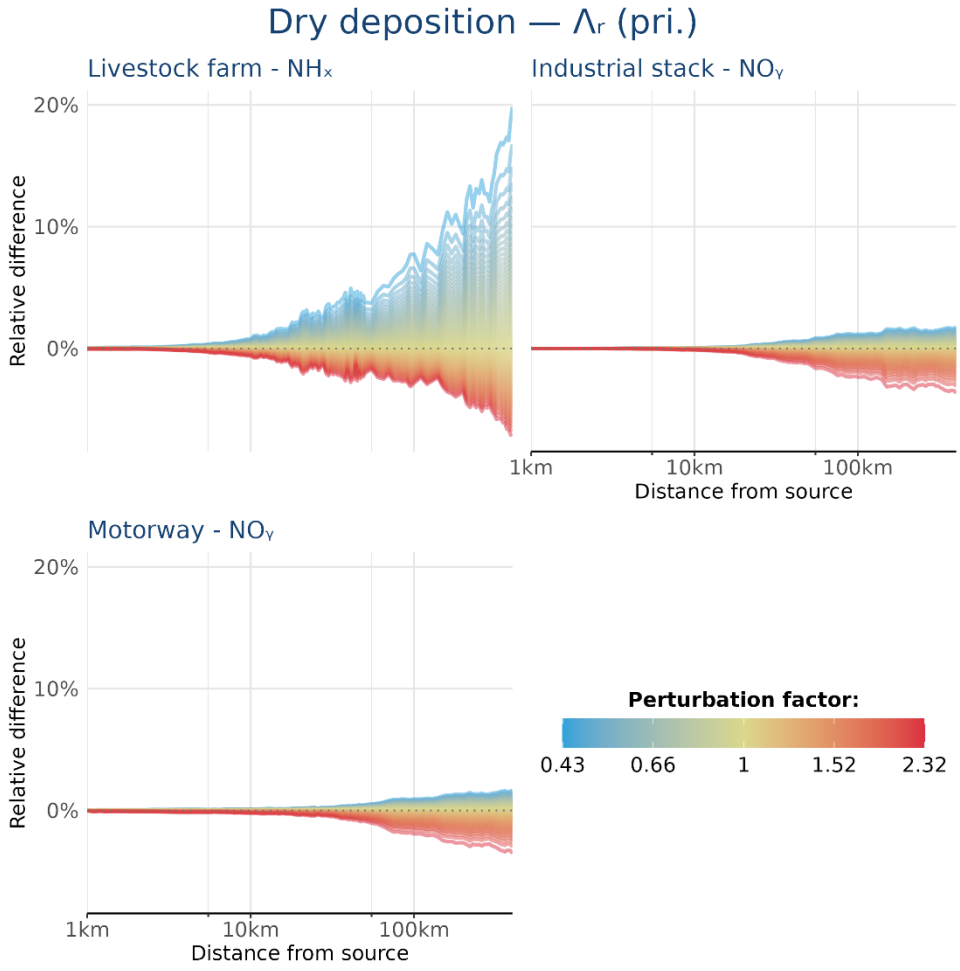

Figure A34 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying $\Lambda_{r,pri}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.9.3 *Wet deposition – impact of $\Lambda_{r,pri}$*

The sensitivity of wet deposition of NH_x to $\Lambda_{r,pri}$ (Figure A35) shows a clear maximum around 10 – 20 km, with changes between about 18% and –22% for increased and decreased $\Lambda_{r,pri}$, respectively. The sensitivity then decreases. This maximum can be explained as follows. Although the sensitivity to $\Lambda_{r,pri}$ in wet deposition is not yet influenced by source depletion and the change in rainout must be proportional to $\Lambda_{r,pri}$, the rainout only plays a minor role near the source. At greater distances, the contribution from rainout to the wet deposition increases, but the same time the source depletion increases as well. Additionally the impact of rainout of NH_4^+ becomes more important. It follows the affected concentration and is thus of opposite sign. These combined effects then induce a decrease in sensitivity again, such that a crossover

point for wet deposition of NH_x is located at ~350 km. Here, changes reverse sign, but are just a few percent at most at 400 km.
A crossover point for the wet deposition of NO_y is lacking, but the decrease towards the boundary of the distance domain studied here suggests a crossover point may be located beyond 400 km. The difference between NO_y and NH_x in this regard can be explained by the differing efficiency of the rainout process between the species involved. This is much more efficient for NH_3 than for NO_x. For the secondary species, rainout is equally efficient with identical, large scavenging rates in OPS.
For the industrial stack the sensitivity of NO_x wet deposition is largest near the source. Relative changes range between 45% and –20% for increased and decreased $\Lambda_{r,pri}$ respectively. The sensitivity to $\Lambda_{r,pri}$ is largest in this case, possibly because the rainout is more effective close the source because of the larger emission height. This promotes immediate interactions with clouds. The changes decreases after about 10 km, to a variation between about ~ 5% and –3% at 400 km. For the motorway, initial changes are somewhat smaller, between ~ 30% and –15% at 1 km distance from the source, and similar to the industrial stack at large distances. The receptors for the motorway case are impacted by source segments at various distances. Although initial emissions all originate from low levels, the contribution from more distant segments will be more enhanced by rainout than the contribution from more nearby segments. This also causes the ‘distance-effect’ to be more spread out.

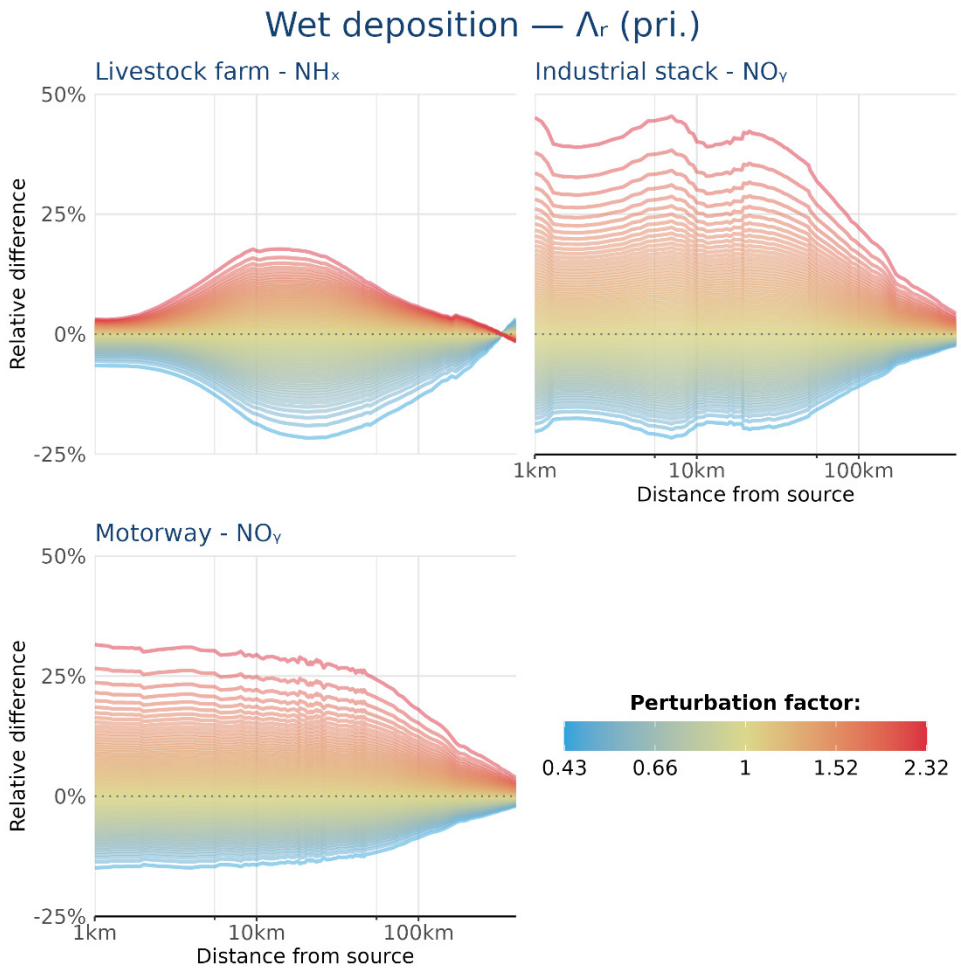

Figure A35 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying $\Lambda_{r,pri}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.9.4 *Total deposition – impact of $\Lambda_{r,pri}$*

The relative difference in total deposition, compared with the reference run when varying $\Lambda_{r,pri}$ for NH_x and NO_y shows that in general, total deposition increases when increasing $\Lambda_{r,pri}$, and vice versa (Figure A36). A notable exception is the reversal of the impact sign for the livestock farm, in which case there is a crossover point for total deposition at about 150 km. Relative differences in total deposition up to 400 km vary between 10% and −13% for the livestock farm, 17% and −8% for the industrial stack and 7% and −4% for the motorway, at increased and decreased $\Lambda_{r,pri}$, respectively.

As such, the impact on the local wet deposition is dominant over the effect of source depletion. However, contrary to the effect on wet deposition per se, there is now an increase with distance from 1 km. This indicates that the impact of dry deposition is more important than the impact of wet deposition near the source, but also that the role of wet deposition becomes stronger at greater distances. After about 20-50 km the impact of $\Lambda_{r,pri}$, on total deposition decreases again. This could indicate that source depletion becomes more important, which reduces the impact of wet deposition locally, at the receptor.

The effect of $\Lambda_{r,pri}$ on dry deposition is indirect and increases with distance. The sign of the effect is opposite to the one on wet deposition, except beyond the crossover point for wet deposition (see Section 1.9.2). Furthermore, the contribution of the deposition of the secondary species becomes relatively more important. Since the parameters of the deposition process of the secondary species are not varied, this sensitivity follows the one of concentration. This also has a sign opposite to the sensitivity of wet deposition. For NH_x this causes the aforementioned crossover point at about 150 km.

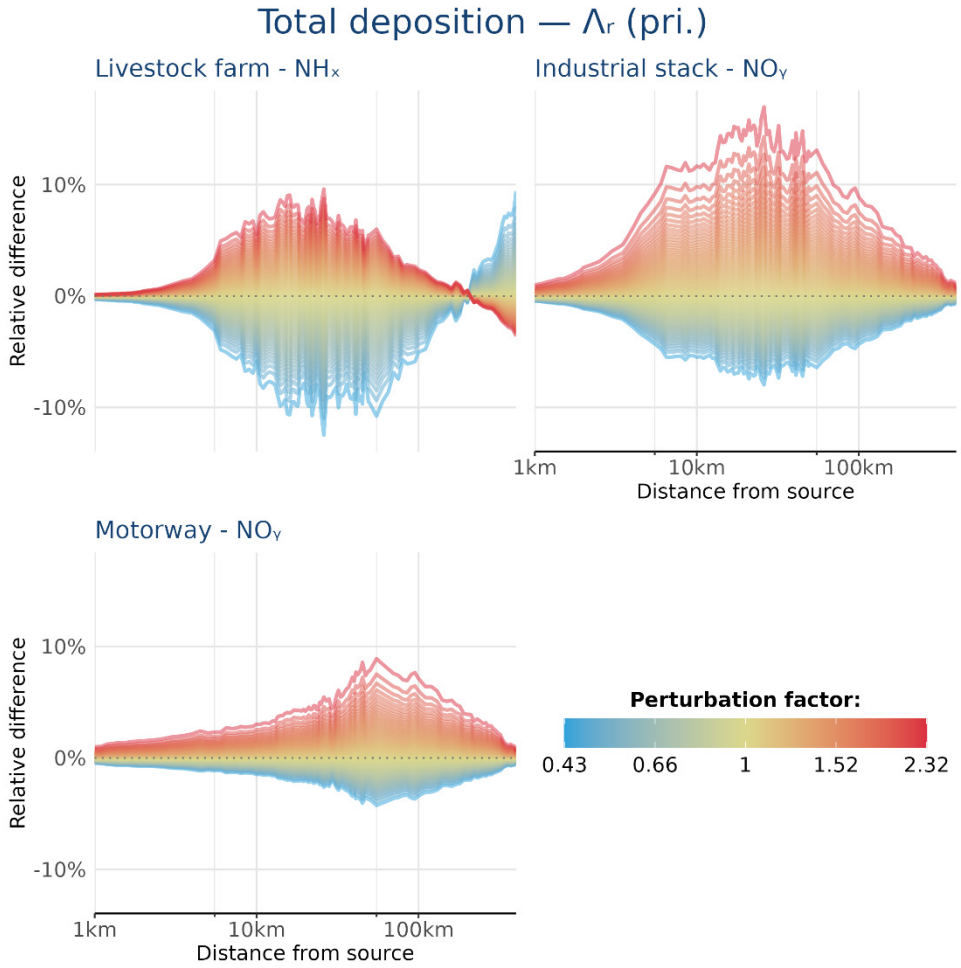

Figure A36 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying $\Lambda_{r,pri}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.10 Rainout scavenging rate – secondary material $\Lambda_{r,sec}$

A perturbation of the scavenging rate of the rainout process of the secondary material, $\Lambda_{r,sec}$, only affects the secondary material:

- A change of $\Lambda_{r,sec}$ affects the amount of material that will be removed from the atmosphere due to source depletion over the trajectory and hence the concentration.
- Dry deposition will roughly follow the changed concentration pattern of the secondary material.

- Initial impacts of $\Lambda_{r,sec}$ on the wet deposition of secondary material are expected to be comparatively large. However, the wet deposition also affects concentration and this reduces the initial effect of $\Lambda_{r,sec}$. However, this effect is expected to be small because of the small impact on concentration.
- The concentration of the primary material is not affected. Therefore, relative differences in the concentration of the primary material are not discussed (relative difference is 0%).

1.10.1 Concentration – impact of $\Lambda_{r,sec}$

Overall the sensitivity of concentration of NH_4^+ and NO_3^-+HNO_3 to $\Lambda_{r,sec}$ is very small (Figure A37). Variations due to the perturbations remain within a few tens of a percent. The largest values are obtained for the industrial stack, at a few kilometres from the source, with a maximum difference of up to 0.25% when decreasing $\Lambda_{r,sec}$ and –0.05% when increasing $\Lambda_{r,sec}$.

In general, concentrations of the secondary compounds decrease when increasing $\Lambda_{r,sec}$, and vice versa. However, at some distances there is no or hardly any effect, except for a strongly decreased $\Lambda_{r,sec}$. This likely caused by a very effective scavenging, where $\Lambda_{r,sec} \to \infty$. In that case, wet deposition due to rainout will only be determined by the number of rain events in the period considered, rather than by the amount or duration of rainfall. These meteorological parameters are not affected by $\Lambda_{r,sec}$.

For the livestock farm and industrial stack the variation of concentration differences with distance first shows an increase. This is explained by the fact that rainout becomes more important with distance and as such the impact on the source depletion is larger for greater distances. This effect is reduced when the scavenging becomes so effective, that wet deposition is entirely or predominantly determined by the number of rainfall events. After 100 km the impact increases again, mostly when reducing $\Lambda_{r,sec}$. Here, the scavenging may have become less effective again, such that altering $\Lambda_{r,sec}$ does have an effect. This may occur because $\Lambda_{r,sec}$ is not a constant, but is proportional to rainfall intensity (R_i) and inversely proportional to the height over which wet deposition takes place (h):

$$\Lambda_r = \frac{WR_i}{h}.$$

Here, W is the scavenging ratio, or ratio of a compound's -initial- concentration in rainwater and air, respectively. This ratio W is actually varied when perturbing $\Lambda_{r,sec}$. R_i and h retain their reference values, which may vary with distance. Notably h tends to increase with distance. Component model results, studied for the receptors in Easterly direction from the livestock farm and industrial stack respectively (not shown), indicate an increase in h and a decrease in R_i with distance for several meteorological classes. This suggests that $\Lambda_{r,sec}$ may decrease with distance.

The sensitivity of concentration to $\Lambda_{r,sec}$ in the motorway case is again much more constant with distance and even smaller than in the other cases, up to ~ 300 km. This is due to the distance-related effect from the many individual source segments with varying distance to the receptor.

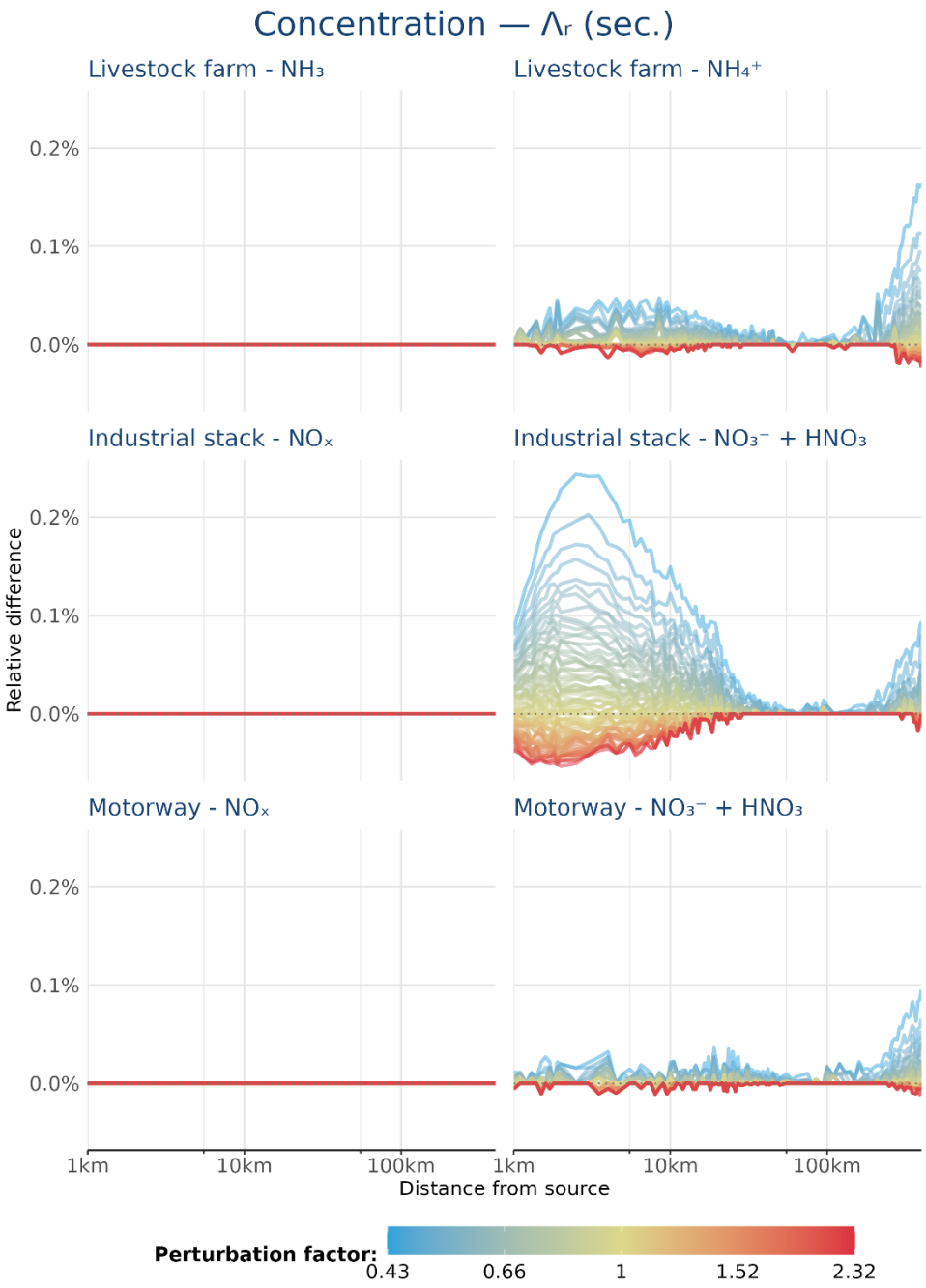

Figure A37 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying $\Lambda_{r,sec}$. Top row: livestock farm NH_3 and NH_4; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.10.2 *Dry deposition – impact due to rainout secondary material $\Lambda_{r,sec}$*

Impacts of $\Lambda_{r,sec}$ on dry deposition are shown in Figure A38. Since the dry deposition process per se is not impacted when varying $\Lambda_{r,sec}$, the relative difference in dry deposition of the secondary compounds with respect to the reference run follows the concentration differences of the secondary compounds: a decrease in dry deposition with increased $\Lambda_{r,sec}$, and vice versa, due to the impact on the source depletion. This effect is

seen in the dry deposition of NH_x and NO_y, although the patterns look slightly different than for the concentrations of the secondary material only. This is because of the contribution from the unaffected amount of primary material, which is especially close to the source more abundant than the secondary material. Hence, the sensitivity of dry deposition to $\Lambda_{r,sec}$ is very small near the source. Although it increases with distance for all source types, all variations remain within up to 0.075% only, at 400 km.

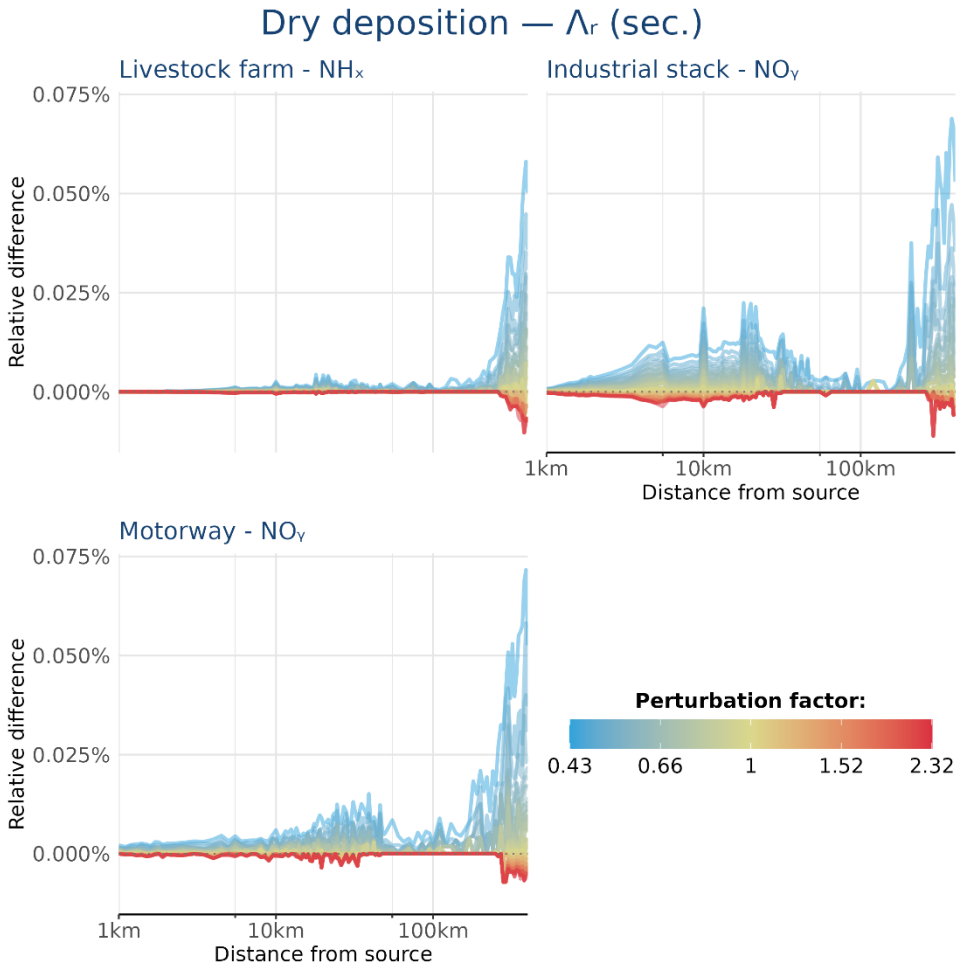

Figure A38 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying $\Lambda_{r,sec}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.10.3 *Wet deposition – impact of $\Lambda_{r,sec}$*

For all source types, an increase of $\Lambda_{r,sec}$ gives an increase in wet deposition of the secondary material at the receptor, and vice versa (Figure A39). This can be recognised in the results for wet deposition of NH_x and NO_y.

The sensitivity is smallest for the livestock case, with relative differences ranging between maximum 0.35% and –0.7% over the 400 km distance, for increased and decreased $\Lambda_{r,sec}$, respectively. The sensitivity is somewhat larger for the industrial stack, with an up to ~ 2% increase in relative difference with increased $\Lambda_{r,sec}$, and a 7% decrease with

decreased $\Lambda_{r,sec}$ near the source, and negligible after 50 km. The impact on NO_y is slightly smaller for the motorway case with an up to 1% increase with increased $\Lambda_{r,sec}$ and a 3.5% decrease with decreased $\Lambda_{r,sec}$, reducing to less than 0.2% at 400 km.
Near the source, the direct impact of $\Lambda_{r,sec}$ on the local deposition dominates over the source depletion effect. The net impact increases up to around 2 km for the industrial stack, around 5 – 10 km for the livestock farm, and up to 10 km for the motorway. This is probably the effect of the increased contribution of rainout to wet deposition with distance. The sensitivity then decreases with distance, which might be due to the larger impact of source depletion. Furthermore, the amount of scavenging at a certain distance may be so effective, that a perturbation of $\Lambda_{r,sec}$, has little to no effect, like explained in Section 1.10.1.
The scavenging ratio for both secondary compounds is higher than for the primary material. This difference is larger for NO_3^- and HNO_3 than for NH_4^+. As such, the sensitivity of total wet deposition of NH_x from the livestock farm is less than the one of NO_y from the other source types. The reduction of the sensitivity of NH_4^+ by that of NH_3 in the sensitivity of NH_x is strongest near the source. Here, the wet deposition of primary material is more important than that of the secondary material, because not much chemical conversion has occurred yet. The increase over distance in the first kilometres may be explained by the gradually increasing contribution from rainout to wet deposition. Since the scavenging of NO_x is not as effective, the sensitivity to $\Lambda_{r,sec}$ on NO_y is more profound also near the source.

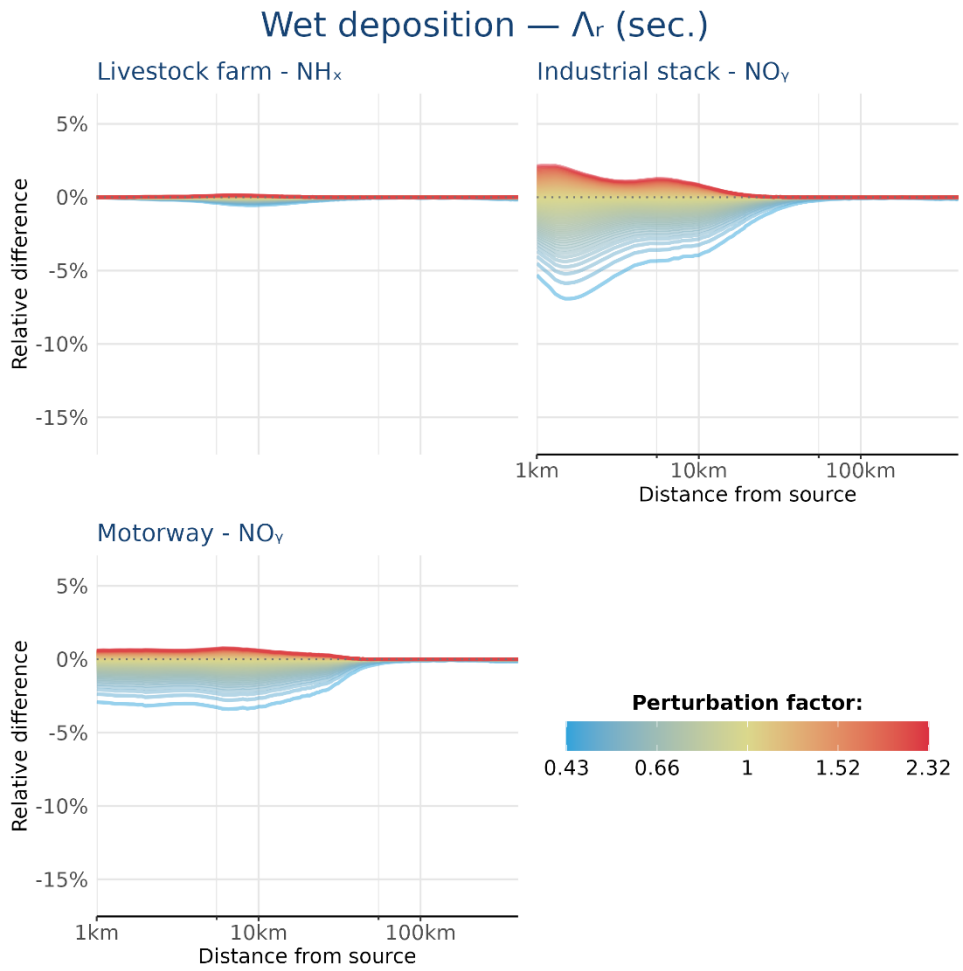

Figure A39 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying $\Lambda_{r,sec}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.10.4 *Total deposition – impact of $\Lambda_{r,sec}$*

The relative differences in total deposition of NH_x (livestock farm) and NO_y (industrial stack and motorway) compared to the reference run due to perturbation of $\Lambda_{r,sec}$ (Figure A40) have the sign of the differences in the local wet deposition at the receptor. In general, total deposition increases with increasing $\Lambda_{r,sec}$, and vice versa. This suggests wet deposition dominates the impact on total deposition. However, a clear influence of dry deposition can also be deduced as follows. Dry deposition is more important near the source. Whereas the relative impact was larger near the source for wet deposition, especially for the industrial stack, a reduced sensitivity of total deposition is obtained near the source. In fact, for the livestock farm the net impact is so small, that the relative difference near the source is almost negligible. In all cases, the sensitivity increases with distance, up to about 10 – 50 km, depending on the change in $\Lambda_{r,sec}$ and the source type. At greater distances, the relative impact of wet deposition increases, such that the variation of wet deposition with distance is followed more. Again, around 100 km a much reduced sensitivity is obtained. This is due to the very

effective rainout, such that wet deposition is predominantly influenced by the number of rain events instead of by the actual scavenging rate. The impact is smallest for the livestock farm case, slightly larger for the motorway and again somewhat larger for the industrial stack, like for wet deposition. However, the impact remains very small, with relative changes between ~0.05% and –0.25% for increased and decreased $\Lambda_{r,sec}$, respectively, for the livestock farm, between about 0.35% and –1.2% for the industrial stack, and about 0.1% and –0.45% for the motorway.

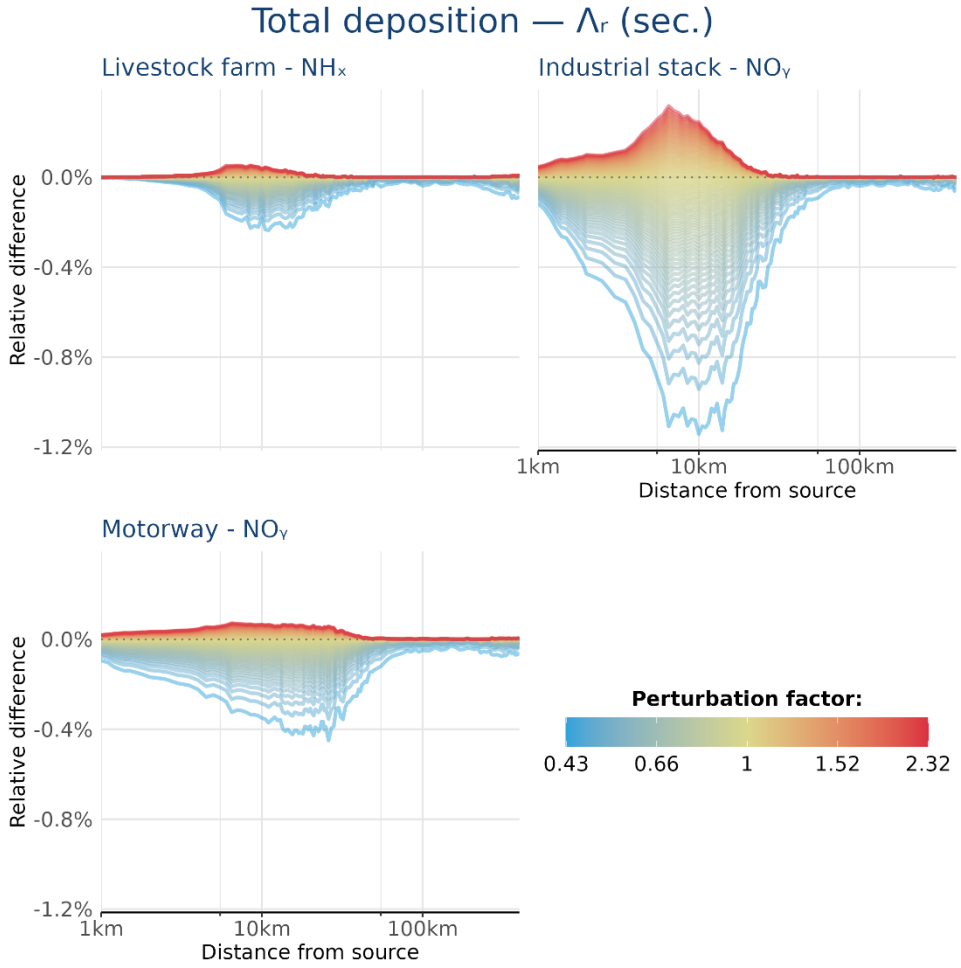

Figure A40 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying $\Lambda_{r,sec}$. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.11 Meteorological conditions, Meteo

Changes in meteorological conditions (Meteo) may affect a suite of processes at once. Important examples are:

- Wind direction determines the transport route of the plume and hence affects the meteorological conditions of the trajectories.
- A change in wind speed directly affects the transport term in the Gaussian Plume Equation. In addition, it has a strong effect on friction velocity, u_*, which is used as a scaling parameter in the description of surface-atmosphere interactions.

- As such, wind speed indirectly influences many other, u_*-related parameters, like the surface fluxes and mixing layer height, the dispersion coefficients used in the Gaussian Plume Equation and interactions among them. Interactions with other parameters may be influenced as well. For example, the interaction between plume rise, vertical dispersion and mixing layer height may induce non-linear interactions that strongly affect the sensitivity to weather conditions.
- The atmospheric resistance to dry deposition is strongly related to u_* and hence wind speed, as affected by atmospheric stability. Dry deposition of NH_3 is more sensitive to changes in wind speed than dry deposition of NO_x. This is because the surface resistance is much smaller for NH_3 than for NO_x.
- Many indirect effects of changes in wind speed will be modulated by changes in other parameters. For example, the indirect effect of wind speed on the surface fluxes will be modulated by solar radiation, temperature and humidity. In the case of the livestock farm, temperature affects the source strength, with higher temperatures leading to somewhat larger emissions.
- Because of such effects and their interactions, the classification in terms of the stability regime will be affected. For example, lower wind speed will lead to less mixing at the surface. This may result in more frequent occurrence of stable as well as convective conditions, depending on changes in, for example, solar radiation.
- Wet deposition is a complex function of rainfall characteristics, notably probability, intensity and event length. Generally, the effective scavenging rates increase with increasing rainfall probability and intensity, but it decreases with event length. However, the net effect of these characteristics depends on the physico-chemical properties of the compounds and other, Meteo-related properties, such as effective source height and therefore plume rise, mixing layer height and dispersion length. Furthermore, the relative contribution from washout and rainout affects the calculations.

Due to the many non-linear interactions related to meteorological conditions, the model sensitivity to Meteo as a function of distance becomes difficult to track and explain. In fact, this behavior is a nice illustration of the reason one has to rely on a model for the assessment of concentrations and deposition in a complex setting. For this reason, only some general observations are made in the following Sections. To facilitate interpretation of the results, average values of some key meteorological quantities are compared in Table A1.

Table A1 Average wind speed, frequency of dominant wind sector (195-225 degrees in all cases), mixing layer height (z_i), friction velocity, air temperature, and precipitation intensity, event length and probability from the seven meteorological datasets, for distance Class 1 (local, 0-50 km). The averages have been determined using the OPS pre-processor METPRO (Sauter et al., 2023), using measurements from KNMI stations in 2015.

Quantity	Reference	AWS260 De Bilt	AWS270 Leeuwarden	AWS290 Twenthe	AWS310 Vlissingen	AWS344 Rotterdam	AWS380 Maastricht
Wind speed (m/s)	3.03	2.12	2.92	2.08	4.16	2.56	2.42
Frequency wind sector 195-225°	20.3%	18.2%	17.5%	22.4%	18.1%	16.1%	23.7%
z_i (m)	176	126	180	133	299	155	157
Friction velocity (m/s)	0.205	0.147	0.212	0.156	0.339	0.182	0.180
Temperature (°C)	10.7	10.9	10.3	10.5	11.5	11.1	11.1
Precipitation							
- Intensity (mm/h)	1.25	1.20	1.27	1.19	1.51	1.31	1.19
- Event length (h)	1.98	2.95	2.89	3.28	2.57	2.80	3.64
- Probability (%)	8.16	8.54	8.05	8.67	6.59	8.11	7.29

1.11.1 Concentration – impact of Meteo

Impacts of Meteo on concentrations are shown in Figure A41. Relative differences in concentration of the primary compounds, range between –50% and +80% for the livestock farm, between –60% and 45% for the industrial stack and between –50% and 45% for the motorway. For the secondary compounds, the relative changes are even larger, up to over 200% in the livestock farm case.

For the low sources (i.e., livestock farm and motorway), the relative differences in concentration are roughly related to the wind speed. Conditions with the lower windspeeds lead to higher concentrations. Notably De Bilt and Twenthe, which both have comparatively low wind speeds, stand out, with much higher concentrations than the reference situation. Although a higher temperature leads to a somewhat enhanced source strength in the case of the livestock farm (see Section 3.2.2 in the main report), such an effect cannot be distinguished here. For the livestock farm we also see a more or less distinguished distance with a maximum deviation or sensitivity. The distance depends on the weather conditions. Beyond that distance, the sensitivity decreases and may even be inverted, typically between about 100 – 150 km from the source.

In the case of the motorway the sensitivities are fairly constant up to about 50 km from the source, beyond which the sensitivities drop and tend to reverse, with an approximately opposite pattern for Vlissingen. A broad maximum occurs because source segments of the road compensate each other's distance effects, up to several tens of kilometres from the source. Beyond that distance, this source gradually resembles a point source again.

The dominating effect of wind speed is much less obvious for the industrial stack, except for Vlissingen. This set of meteorological conditions, which has the highest wind speed by far, shows the largest negative deviations in all three cases. Apart from the direct effect of wind speed on dilution in the air, the compounds are emitted into a

mixing layer of which the height is -on average- positively correlated with wind speed (see Table A1).
Presumably, for Vlissingen the mixing layer is high enough to allow most or all of the emissions to occur within it. For the lower wind speeds, the mixing layer is also much lower and part of the pollutants may be emitted above the mixing layer. The share of the emission to which this applies is a complex function of plume rise, vertical dispersion and atmospheric stability. Part of the mass may be entrained again at greater distance from the source, also depending on the interplay between many factors influenced by Meteo. This results in a much more "erratic" pattern than in the case of the low sources.

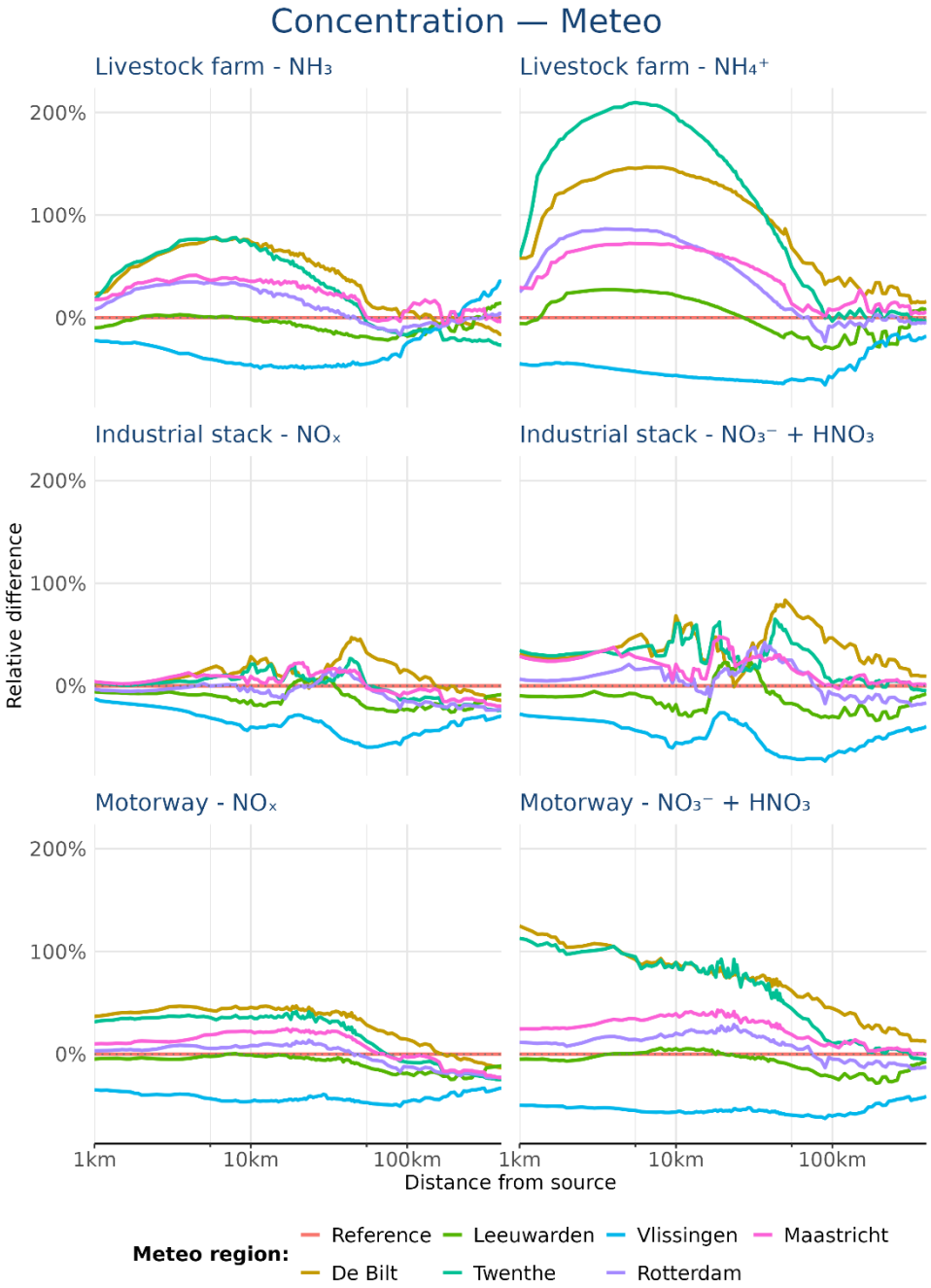

Figure A41 Relative difference (%) of concentration compared to the reference run when varying Meteo. Top row: livestock farm NH_3 and NH_4^+; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^- +HNO_3.

1.11.2 *Dry deposition – impact of Meteo*

Relative differences in dry deposition of NH_x and NO_y due changed Meteo (Figure A42) amount to a maxima of about –50% and 65% for NH_x in the livestock farm case, –50% to 45% for NO_y from the industrial stack and –40% and 40% for NO_y from the motorway. These differences are somewhat smaller than the ones for concentration. This may be explained by the compensation mechanism, that is, the feedback

between in dry deposition and concentration. Briefly, more deposition over the trajectory reduces the concentration which reduces deposition at the receptor again, and vice versa. Dry deposition is strongly related to the concentration. Hence, some similarity with the patterns in the case of concentration is expected. Indeed, the order of cases with respect to the differences is roughly the same as for concentration. However, in particular in the case of livestock farm we see a strong "erratic" behavior. This may be due to the impact of land use and surface properties and its interaction with Meteo, which may lead to variations in deposition velocity.
Land use and properties modulate the effects of the weather conditions on the deposition velocity, notably via the impact of the surface roughness on the friction velocity (see Table A1). The deposition velocity of NH_3 is much more sensitive to this modulation than that of NO_x. Hence, for the livestock farm case in particular, changed weather conditions amplify land-use related differences in deposition velocity. This explains the more "erratic" behavior of the sensitivity in that case. For NO_x, the deposition velocity is much less sensitive to the weather conditions, resulting in much smoother curves.

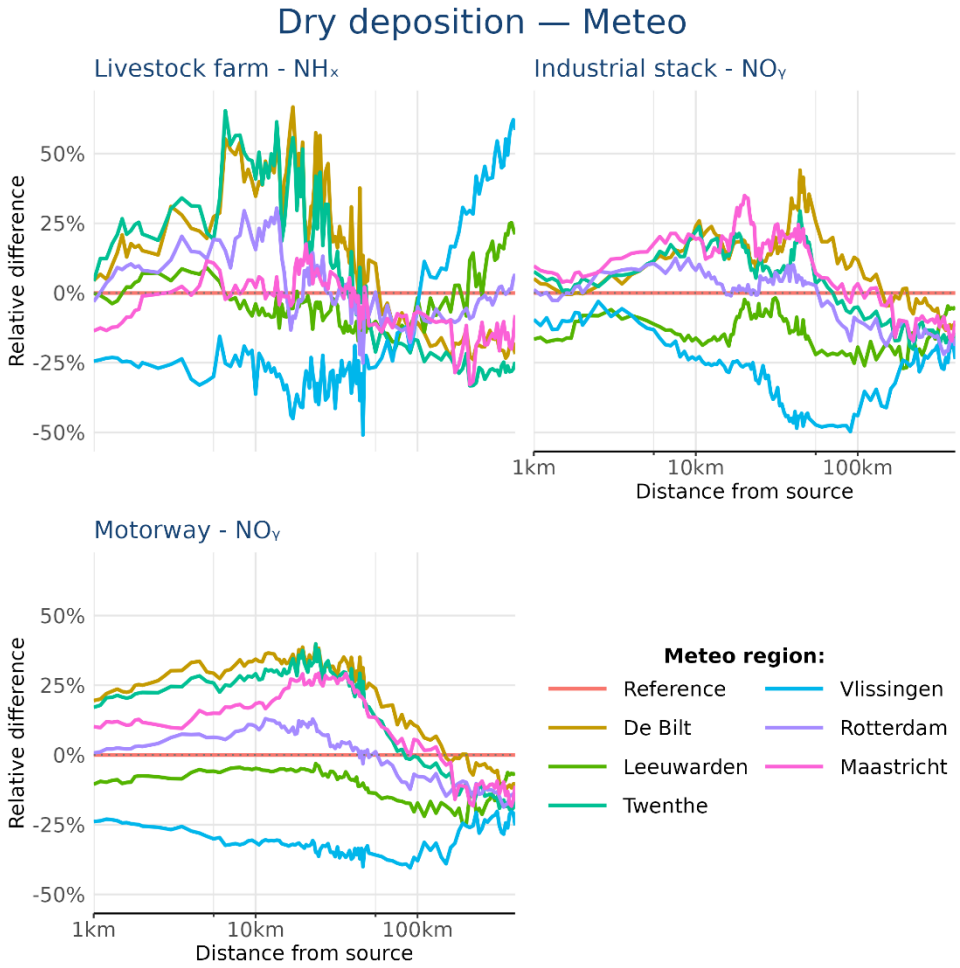

Figure A42 Relative difference (%) of dry deposition compared to the reference run when varying Meteo. Upper left: livestock farm NH_x; upper right: industrial stack NO_y; lower left: motorway NO_y.

1.11.3 Wet deposition – impact of Meteo

Impacts of Meteo on wet deposition are shown in Figure A43. Near the source, low absolute values of wet deposition, related to washout, may result in comparatively large relative differences, in particular for the industrial stack. In this case, emissions may occur above the mixing layer leading to (near) zero concentrations at lower levels and hence hardly any washout. At the same time, the rainout process needs more time to become effective. This "switch" of the wet deposition regime probably causes the steep drops in the sensitivity of wet deposition around 50 km from the livestock farm and industrial stack. This is the distance where interpolation between the local class and the class at 100 km starts. In the case of the motorway contributions from different individual source segments at different distances from specific receptor point blend again.

Wet deposition is strongly related to concentrations, albeit in a complex way. This explains that, in the case of the low sources we see some similarities with the sensitivity of concentration: low wind speeds lead higher concentration and hence, to more wet deposition. However, this first-order impact of meteo is clearly modulated by precipitation effects. This is most evident for Rotterdam. Whereas the concentration only showed moderate deviations from the reference case, up to a maximum of about 30% near the source (livestock farm case), the maximum relative effect on wet deposition amounts to over 75% (livestock farm case). This is quite similar to both sets with low wind speed, De Bilt and Twenthe, in the case of the livestock farm, and to Twenthe in the case of the motorway. Rotterdam is located in the Randstad area, which is known for enhanced rainfall (Noordhoff Atlasproducties, 2021). Indeed, this set has a comparatively high average rainfall intensity (see Table A1).

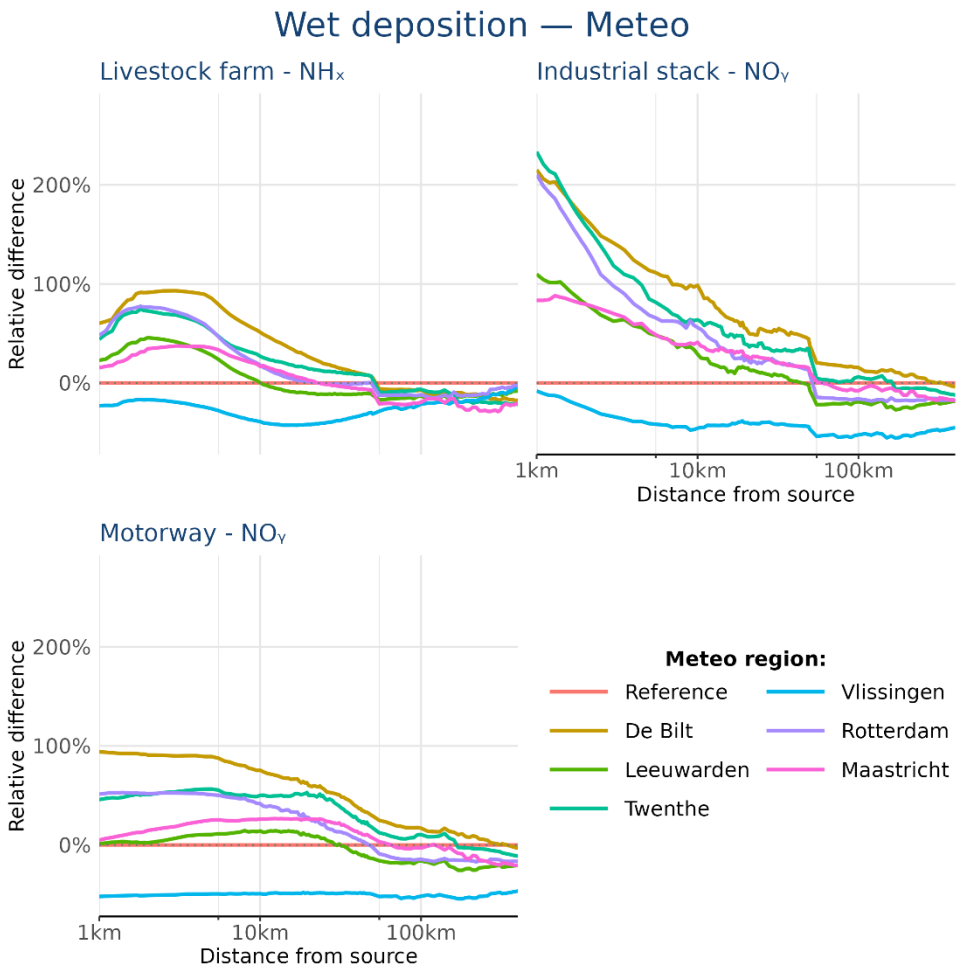

Figure A43 Relative difference (%) of wet deposition compared to the reference run, when varying Meteo. Upper left: livestock farm NH_x; upper right: industrial stack NO_y; lower left: motorway NO_y.

For the industrial stack case, there is hardly any similarity with the patterns in concentration differences. Like for the low sources, the cases with low wind speed stand out. This is unlike the differences for concentration. Relative differences near the source are over 200%, for the Meteo with low wind speed and comparatively high rainfall probability. In all cases except Vlissingen, there is the gradual decrease in the relative effect on wet deposition up to distances of 50 km. Both features may partly be due to low, but gradually increasing absolute wet deposition rates. Only in the case of Vlissingen the differences, negative in this case, increase with distance. Here, the - on average - high wind speed results in a higher mixing layer. This allows a comparatively large part of the pollutants to be emitted within this layer. Also, vertical dispersion is expected to be larger, since this quantity scales with friction velocity (see Table A1). As a result, the plume is expected to be deeper near the source. Hence, washout may be more effective near the source, in spite of the lower precipitation intensity and probability. However, the effect of lower concentration, and precipitation probability and intensity, cause gradually increasing differences for Vlissingen. Like

in the case with the low sources, the Rotterdam set shows maximum sensitivities quite similar to the ones of De Bilt and Twenthe.
Beyond 50 km from the source, rainout becomes dominant. For this process interactions are more complex and much more none-linear, with compensating effects of event length, which depends on wind speed in the Lagrangian framework of OPS. This leads to quite similar sensitivities within a limited range for most cases, between roughly –60% and +20%. Again, the set based on Vlissingen stands out. This set has been derived from observations at a coastal station in the South-West of the Netherlands. Meteorological statistics clearly deviate from the statistics of the other stations (Table A1).

1.11.4 Total deposition – impact of Meteo

In general, relative differences in total deposition due to changed Meteo (Figure A44) range between –45% and 60% for the livestock farm, between –50% to 45% for the industrial stack and between –45% and 45% for the motorway. At first sight, the relative differences in the sum of dry and wet deposition resemble the ones from dry deposition. Although relative differences in wet deposition may be much larger, dry deposition is the more important removal process in the cases studied here and hence is able to dominate the relative differences. This is most clear nearby the source. Here, relative differences in wet deposition range from several tens to hundred (low sources) and even two hundred percent (industrial stack), with quite noticeable spatial differences up to a few tens of kilometres from the source. Yet, none of these are readily recognisable in the relative differences plots for total deposition. Somewhat further away, one can detect some influence of wet deposition. Here, maximum differences may become somewhat different. Also, the relative differences between the cases as a function of distance may differ from the ones of dry deposition. Other than that, the differences are too small to detect noticeably impacts of possible non-linear interactions.

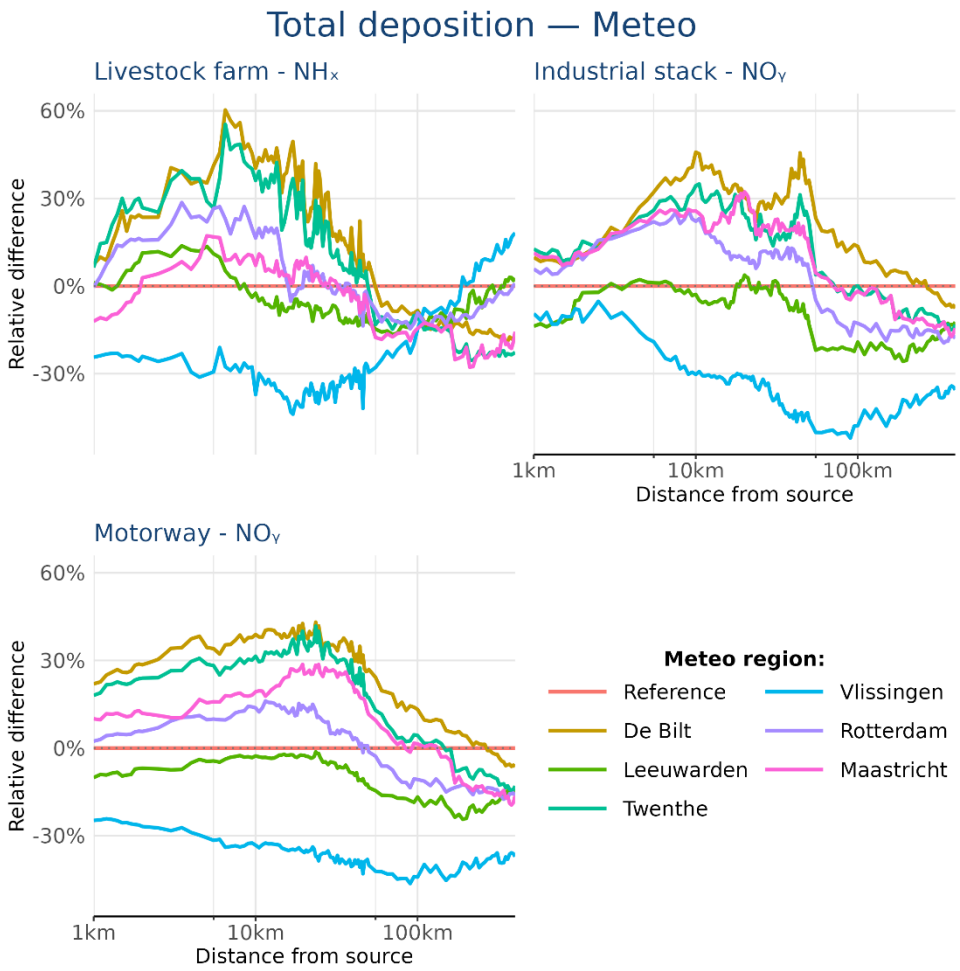

Figure A44 Relative difference (%) of total deposition compared to the reference run when varying Meteo. Upper left: livestock farm NH_x; upper right: industrial stack NO_y; lower left: motorway NO_y.

1.12 Diurnal variation *dv*

Perturbation of the diurnal variation (*dv*) has the following main impacts:

- For the industrial stack, the motorway and partly for the livestock farm, perturbation of *dv* is achieved by scaling the sinusoid of the distribution of the emission correction factors (*emf*) over the day. An increased *dv* increases the amplitude of the distribution around 100%. The opposite is true when decreasing *dv*. This implies a redistribution of the emissions over the day, while the yearly total remains unaltered.
    - For the distribution of the industrial stack, there will roughly be an increase in daytime emissions and a decrease in nighttime emissions for increased *dv* and vice versa. In the case the livestock farm part of the diurnal variation is also based on half the distribution of the industrial stack.
    - For the motorway, there will be an increase in emissions during daytime hours (with peaks for the morning and end

of the workday traffic) and decrease during nighttime hours for increased *dv*, and vice versa.

- The diurnal variation in the 12 two-hourly blocks used to describe *dv* is linked to the frequency of occurrence of a specific meteorology (stability and wind direction) and distance class. *In general* stability classes with higher frequency during daytime hours, like the unstable classes, will have a higher *emf* with increased *dv* and vice versa. In the component model results for the livestock farm and industrial stack for receptors in easterly direction (not shown), a decrease in *emf* was found for increased *dv* for the other meteorological classes, representing neutral and stable conditions, respectively. For other spokes, it could be different and/or non-linear (depending on the distribution of the hours over the stability classes which differ over the preset trajectory distances as well). The impact also varies over the trajectory. For some distances a stronger deviation from the reference run is obtained.
- The perturbation of the diurnal variation of the twelve 2-hourly blocks is both lower- and upper bounded, to prevent a flip in the "sinusoid" and negative emissions, respectively (see Section 3.2.2 of the main report). Note that the default "sinusoid" differs among the various source types. This implies that the bounds on these variations will differ. Therefore, in the case of a specific large perturbation, its relative effect on the *dv* change may depend on the source type. This also introduces asymmetrical effects in the sensitivity to *dv* (see also Figure A49 in Section 2.8)
- For the livestock farm an additional emission correction occurs. This part is based on a linear relationship between air temperature and emission strength from livestock farms. When *dv* is increased, the slope of the temperature correction increases, and vice versa. This results in an increased *emf*, and hence higher emission strength, when air temperatures are above 10 °C. A lower emission strength occurs at air temperatures below 10 °C. Because the air temperature varies over the trajectory and among the stability and wind direction classes in OPS, this results in highly non-linear effects with sometimes an increase in *emf* and sometimes a decrease in *emf* over the 400 km.
- The temperature-related perturbation in the case of the livestock farm may affect the yearly total of the emissions, with higher emissions when the yearly average air temperature is higher than 10 °C, and vice versa.
- Because the efficiency of deposition is related to stability, wind direction and distance class, the amount of deposition is affected by the redistribution of emissions over time. Therefore the impact on dry and wet deposition at the receptor, averaged in time and over the receptor spokes is not straightforward, and does not necessarily follow the impact on concentrations. The same is true for the source depletion over the trajectory. Though the source depletion factor in itself is unaltered, the source strength with which the source depletion factor is multiplied varies in a specific class compared to the reference run. This also makes that the impact of a perturbed *dv* on the amount

deposited over the trajectory is not straightforward, which will impact the sensitivity of concentration to *dv* as well. Note that the impact on the source depletion may not be the same as the impact on the dry deposition at the receptor because the variation of the diurnal variation is not identical over the trajectory and at the receptor.

1.12.1 *Concentration – impact of dv*

Impacts of *dv* on concentrations are shown in Figure A45. For the livestock farm an increase in *dv* initially leads to lower concentrations and vice versa from 1 km up to just over 100 km (NH_3) and 300 km (NH_4^+), after which the sensitivity changes sign. The opposite is seen when decreasing *dv*. The relative difference in concentration compared to the reference run around 10 km is about –12% (increased *dv*) and 7% (decreased *dv*) for NH_3, and about –17% and 8% for NH_4^+. At 400 km this is about 8% and –4% for NH_3 and 3% and –1% for NH_4^+. In general for increased *dv*, emissions in the unstable classes - when there is more dilution of the plume - have increased, while emissions in the more stable classes - when the plume is less diluted - were decreased. As a result, lower concentrations are expected in general. A weakening of the diurnal variation (i.e., decreased *dv*) likely leads to, *in general*, higher emissions for stable classes, which gives more 'dense' plumes. Also, emissions are lower in the unstable classes with more diluted plumes. Thus, higher concentrations are expected for decreased *dv*.
The impact of the temperature correction in the case of the livestock farm is more difficult to pinpoint, because of the strong non-linear interactions with the stability classes as well as with distance over the trajectory. Since the temperature correction also impacts the yearly total of the emission (see explanation at the start of Section 1.12), it could be that this effect is more pronounced at greater distances which would then explain the increased concentration with increased *dv*. However, the varying impact of the redistribution of the diurnal variation with distance may also account for this. The varying impact over the distance and meteorological classes which may also vary over the different receptor directions, make that a straightforward explanation for the impact of *dv* is extremely difficult.
For the industrial stack, over most of the trajectory, concentrations decrease with increasing *dv* and vice versa, presumably due to a interactions with atmospheric stability, like in the case of the livestock farm. Only between a distance of 1 and ~ 2 km, the impact is opposite to the one for the livestock farm. This may be due to the influence of the perturbation of the temperature correction on dv, which is absent in the case of the industrial stack. Furthermore, the diurnal variation of the industrial stack is twice the amplitude of the one is applied to the livestock farm. However, as mentioned earlier, the impact of varying *dv* is not straightforward. In general, the impact is somewhat larger than for the livestock farm, with the highest impact just before 50 km: about –25% and 12% for in- and decreased *dv* for NO_x and about –32% and 16% for NO_3^-+HNO_3.
For the motorway, the sensitivity to *dv* with distance has a shape that is more or less similar to the one of the livestock farm. However, in contrast with the livestock farm case, impact of an increased *dv* is considerably smaller than for a decreased *dv*. Differences are highest around 10 km and range between about –9% and over 40% (in- and

decreased *dv*) for NO_x, and between −11% and 55% for NO_3^-+HNO_3. The impact switches sign after about 150 to 200 km, but remains small, up to about 1% for increased *dv* and −7% for decreased *dv* at 400 km.

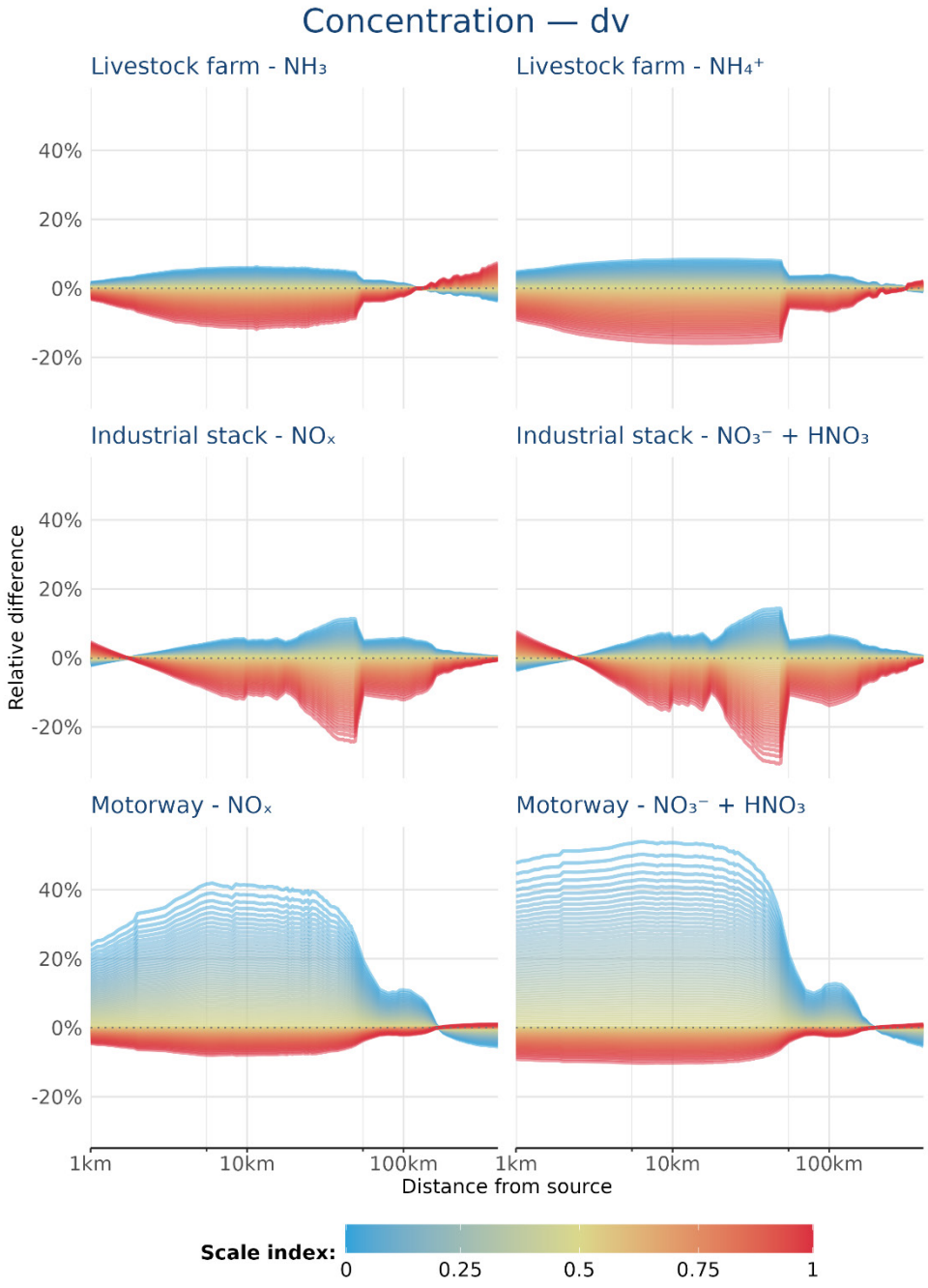

Figure A45 Relative difference (%) in concentration compared to the reference run (average over the various directions) when varying dv. Top row: livestock farm NH_3 and NH_4; middle row: industrial stack NO_x and NO_3^-+HNO_3; bottom row: motorway NO_x and NO_3^-+HNO_3.

1.12.2 *Dry deposition – impact of dv*

The impact of *dv* perturbations strongly varies over the different wind sectors (12 directions) and over the stability and distances classes.

Furthermore, the impact over the trajectory may differ from the local impact at a receptor point. Due to the many non-linearities, it impossible to explain every variation in the average relative difference pattern over distance. Therefore only the general shape and deviations from the reference run will be addressed here. Impacts of *dv* on dry deposition are shown in Figure A46.
For the livestock farm the relative difference in dry deposition of NH_x resembles the variation of the NH_3 concentration. The highest impact is seen in the first 50 km with a maximum variation between almost –10% for increased *dv*, and almost 6% for decreased *dv*. The differences change sign just after 100 km, reaching just over 5% (increased *dv*) and –2.5% (decreased *dv*).
For the industrial stack the relative difference in dry deposition of NO_y does not resemble the variation of the NO_x concentration, nor the NO_3^- $+HNO_3$ concentration. Now, the impact is highest around 1 km with up to over 18% differences for increased *dv* and –6% for decreased *dv*. The impact decreases and changes sign around 20 km. Another small peak in impact is obtained just before 50 km (between –14% and 7% for in- and decreased *dv*, respectively) after which the impact decreases to less than a percent at 400 km.
For the motorway the relative difference in dry deposition of NO_y does resemble the concentration variations to some extent, though the variation at shorter distances is somewhat reduced. The peak differences are found around 10 km, amounting to about –5% and 24% for in- and decreased *dv*, respectively. The impact changes sign around 150 km, and is about 1% and –6%, respectively at 400 km.

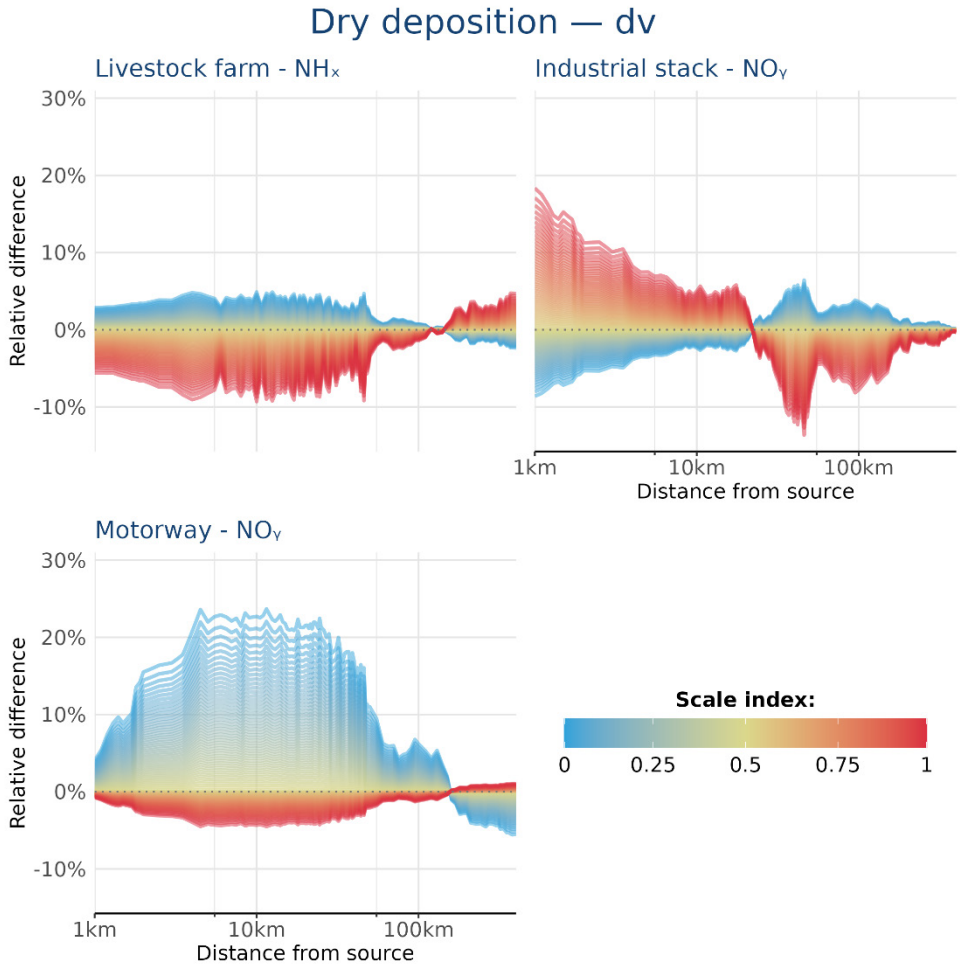

Figure A46 Relative difference (%) in dry deposition compared to the reference run (average over the various directions) when varying dv. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.12.3 *Wet deposition – impact of dv*

For the wet deposition the relative difference compared to the reference run due to perturbed *dv* (Figure A47) also differs from the one of concentration. For the livestock farm the sign of the differences is in general similar, but the crossover point is reached already just before 50 km. A maximum sensitivity is found at a few kilometres from the source, leading to differences of about –6% and 3% for in- and decreased *dv*, respectively, and to –3% and almost 7% at 400 km.

For the industrial stack variations are highest near the source, between almost –30% and 15% for in- and decreased *dv*. Further away, variations decrease with distance, until a crossover point at around 300 km. The relative difference in NO_y wet deposition then becomes around ±1% at 400 km.

For the motorway the relative difference in wet deposition relative to the reference case follows the sign of the concentration differences. However, the impact is now highest near the source, about –4% and 20% for in- and decreased *dv*. Next, the impact decreases with distance until the crossover point at around 150 km. The relative difference then

increases again to about 2% and –7% at 400 km for increased and decreased *dv*, respectively.

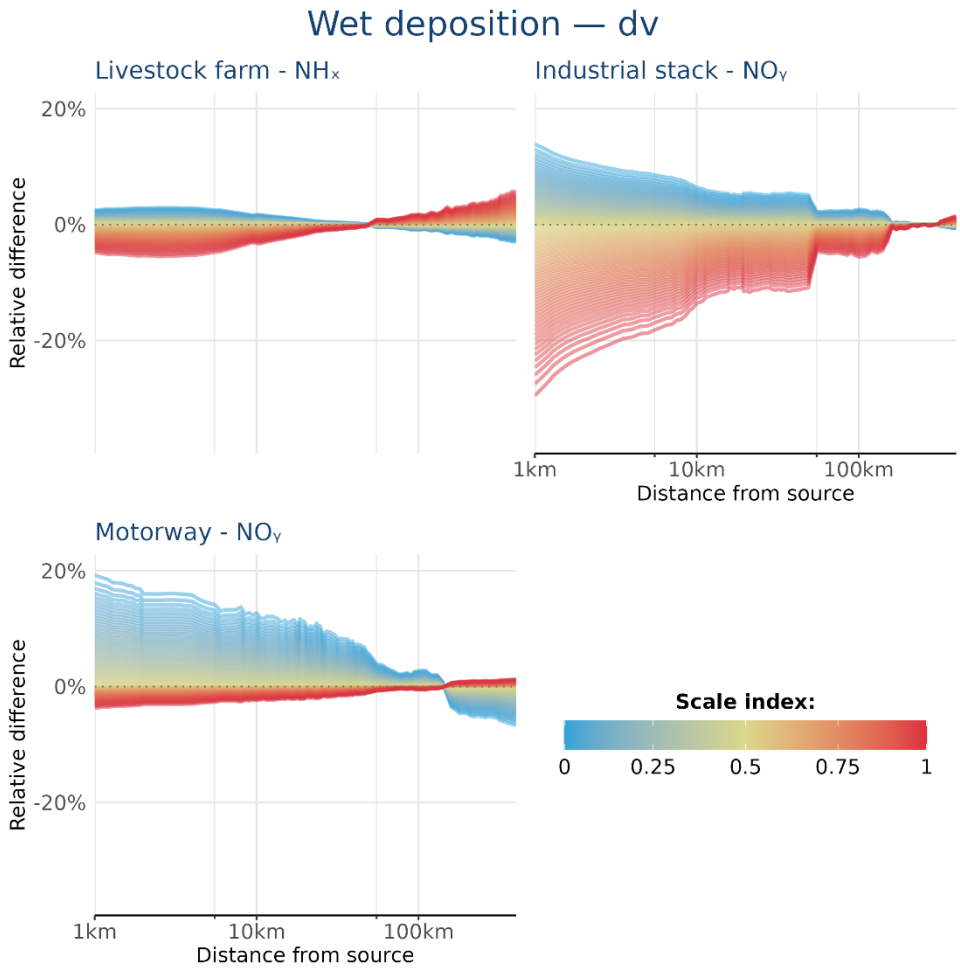

Figure A47 Relative difference (%) in wet deposition compared to the reference run (average over the various directions) when varying dv. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

1.12.4 *Total deposition – impact of dv*

When varying *dv*, the relative difference in total deposition of either NH_x or NO_y (Figure A48) is the result of the impacts on both dry and wet deposition described above. For NH_x from the livestock farm the impact is fairly similar to the one dry and wet deposition. Total deposition decreases with increasing *dv*, and vice versa, in the first 50 km. This is followed by an only very small impact, for which a 'wiggly' pattern is obtained because the crossover points for dry and wet deposition are found at different distances. The sensitivity is highest just before 5 km, with maximum variations of around –6% and 5% for increased and decreased *dv*, respectively. After the crossover point the sensitivity changes sign and variations between about 6% and –3% are reached at 400 km.

For the industrial stack the change in total deposition initially has the sign of the dry deposition changes, which initially shows an increase with increased *dv*. The wet deposition initially shows a decrease with

increased *dv,* and vice versa, which reduces the net impact on total deposition. Furthermore the crossover point is around 50 km for dry deposition while this is at about 300 km for wet deposition. Additionally, the relative impact of dry deposition decreases with distance, while the relative importance of wet deposition increases with distance. This results in some sort of crossover "plateau" around 10 km, and again a switch in sign just before 400 km. The sensitivity is highest at 1 km, with changes between around 17% and –8% for in- and decreased *dv*. It decreases towards the crossover point around 10 km. This is followed by an increase in impact up to about –13% and almost 7%, for in- and decreased *dv*, just before 50 km. The differences are only about half of these at 100 km, and less than ± a percent at 400 km.
For the motorway the total deposition of NO_y also follows the dry deposition of NO_y near the source. At greater distances the impact of varying *dv* is similar for both dry and wet deposition. As such, initially there is a decrease in total deposition with increased *dv* until the switch in sign at about 150 km. The impact is highest around 10 km, between about –5% and 23% for in- and decreased *dv*. At 400 km differences have decreased to between about 1% and –7%.

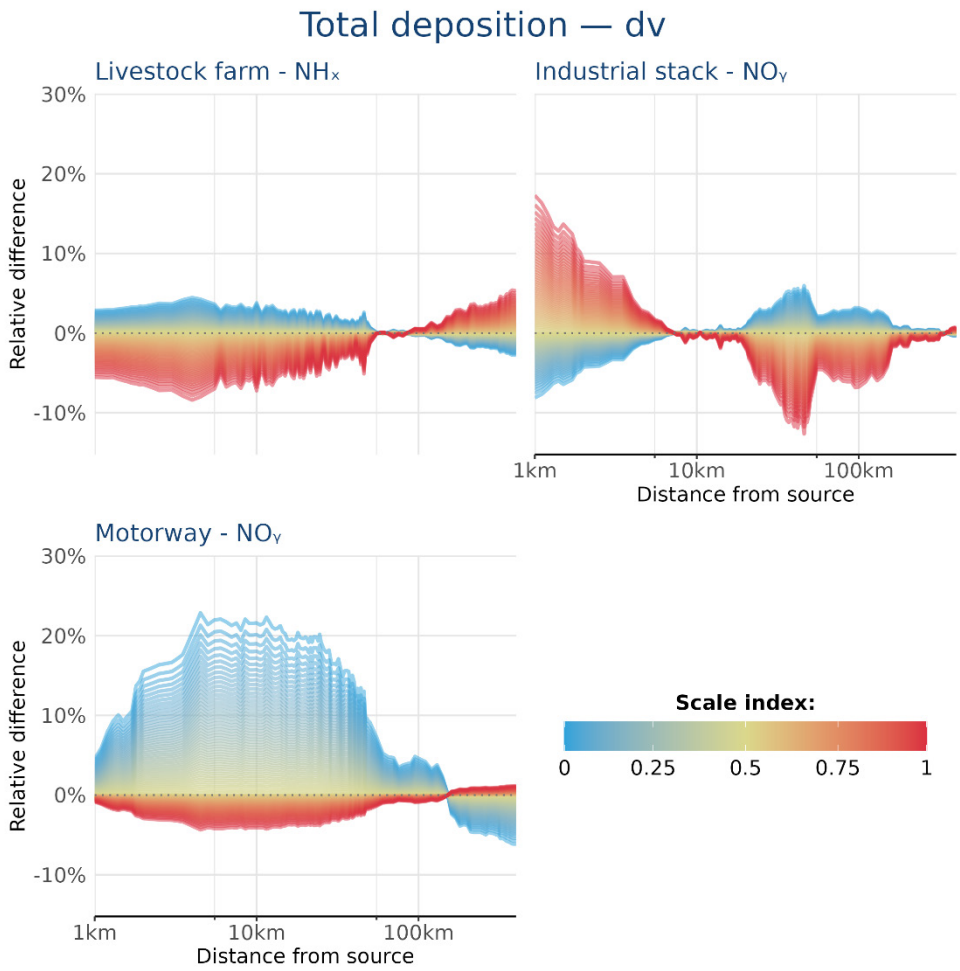

Figure A48 Relative difference (%) in total deposition compared to the reference run (average over the various directions) when varying dv. Top left: livestock farm NH_x; top right: industrial stack NO_y; bottom: motorway NO_y.

2 Adjustments in OPS-LT needed to perform sensitivity analyses

Some of the variations of parameters varied in the sensitivity analysis can be provided as input in the OPS-LT model. More specifically, the meteorological data is varied by selecting different meteorological files. Hence, variation is included by providing different control files (input file of OPS-LT) for which the meteorological file is varied (a pick from 7 different files). Variation in the source height was included by providing different source files for which the source height varies between the files. For most parameters however, this could not be done and alterations were needed on code level. This Section describes these alterations as well as the impact of the alteration on other parts of the code. Coded lines are indicated using the 'Courier New' format. Names of subroutines are indicated in blue.
The multiplication factors that are mentioned below are prescribed in a so called "ops_varin.txt" file which is provided as input to OPS. When this file is not provided, the default values are used, which give no perturbation. This particular implementation is not described here, as the focus is on the perturbation of the variables for the sensitivity analysis.

2.1 Plume rise due to buoyancy or momentum – *dh*

When the plume is emitted with a certain exit velocity or when it contains a temperature higher than the environmental temperature and therefore has a certain 'buoyancy' compared to the environment, the emitted plume may rise. The amount of plume rise depends on whether plume rise is caused by momentum (due to an exit velocity) or buoyancy and is furthermore dependent on the atmospheric conditions. When the plume contains both momentum and buoyancy, OPS uses the plume rise due to the dominant process as the final plume rise. In OPS this plume rise is only computed for vertical outflow, not for horizontal outflow.
To account for uncertainty in the calculation of the plume rise in OPS, the plume rise after determination of the final plume rise is perturbated in subroutine ops_plumerise with:

```
dh = dh*varin_unc%plrise_fact.
```

Here the original plume rise *dh* on the right hand side (m) is multiplied with a value prescribed in the structure *varin_unc%plrise_fact*. This factor is prescribed in the ops_varin.txt file.
The plume rise *dh* is added to the emission height to determine the initial plume height at the stack. OPS-LT determines plume rise twice. First a preliminary plume rise is determined based on the stability class and the preliminary wind sector to determine a preliminary initial plume height. This is needed to determine the wind sector and interpolation factor between wind sectors since the wind turns with height and as such the amount of turning, turning angle, depends on the initial source height. This is done in a preliminary way since the various meteorological variables that are needed further in the code are

dependent on the wind sector. Later in the code the plume rise is updated by using the actual meteorological data. For both calculations the subroutine ops_plumerise is used so for both methods the final plume rise is perturbated.

2.1.1 *Impact of altered plume rise*

The plume rise *dh* is used to determine the plume height at the source location by adding *dh* to the emission height. As stated above the plume rise is calculated twice. The first time this is done in a preliminary way to determine the wind sector and interpolation factor between wind sectors since the turning of the wind which depends on height needs to be taken into account. This impacts the meteorological variables which are determined based on the (interpolation between) wind sectors. Later in the code the plume rise is updated by using the actual meteorological data. Then both emission height and plume height including plume rise are subsequently used to determine the plume penetration through the inversion layer at the top of the boundary layer. Depending on the plume height and the boundary layer height, the emission may (partly) occur above the boundary layer such that the fraction of the plume inside the boundary layer (variable *onder*) may be less than 1. The concentration calculated using the total emission strength is corrected using this fraction. Note that OPS uses parameter *onder* also to incorporate the process of entrainment at the top of the boundary layer. This process mixes mass emitted above the boundary layer into the boundary layer gradually with increasing distance. Source depletion is also only performed on the amount of mass within the boundary layer.
Furthermore the plume height including plume rise at the source is used as initial height to determine the plume height at greater distances (where plume descent is taken into account for heavy particles). This is used in the equation for the concentration taking into account reflections at the surface and the top of the boundary layer. This also holds for the source depletion routines.
For point sources it is also assumed that there is extra vertical dispersion when plume rise due to buoyancy occurs. This is included by quadratically adding an extra vertical dispersion (*a*) to the vertical dispersion due to transport (*sigz*) and the initial vertical dispersion as part of the source characteristics (*sigz0*) in subroutine ops_conlt_par_puntbr:

```
sigz = SQRT((sigz*sigz) + (a*a) + (sigz0*sigz0)).
```

As such, an altered plume rise impacts the concentration and deposition in various ways. First the plume height is changed, which can affect the travel speed and direction of the plume as well as other meteorological variables. With respect to the receptor height, a higher plume with a reference height already above the receptor height, will result in lower concentrations due to that the plume axis is further away and more dilution occurs with higher wind speeds. On the other hand, for sources which penetrate the boundary layer, it is possible that with higher plume a (larger) part of the plume is now emitted above the boundary layer. This mass may enter the boundary layer at a greater distance due to entrainment/fumigation which could lead to higher concentrations compared to the reference run at this distance. The extra vertical

dispersion also causes further dilution of the plume. It depends on the distance of the receptor height and the plume axis whether this results in higher (when the plume now reaches the receptor height where it did not/less before) or lower concentrations (due to increased dilution).

2.2 Vertical dispersion length – σ_z

The vertical dispersion length σ_z (m) is the standard deviation of the concentration distribution of the Gaussian plume in the vertical direction at a certain distance of the source. In other words: it is a measure for the depth of the plume starting from the plume axis. Its value depends on the atmospheric conditions and the plume height and wind speed, amongst others.
Testing the sensitivity to uncertainty in σ_z can be done in two different ways. The first is to vary σ_z caused by atmospheric transport directly after its calculation. The second is by varying σ_z within the iteration in the separate transport routines ops_surface, ops_convec and ops_neutral. The latter takes into account the effect of a varying σ_z on reflections and therefore plume height and plume speed which again influence σ_z. The iteration methods however differ between the three transport routines while furthermore, depending on the source height, an interpolation occurs between the routine for the surface layer and the routines for the boundary layer above the surface layer. This makes that it is not clear to what extent a prescribed variation on σ_z ensures that σ_z is indeed varied by that amount. Therefore it was decided to go for the first option.
Note that it was decided to only vary the σ_z due to transport and not the initial dispersion length which is associated with area sources or as part of the source characteristics. This contribution is added quadratically outside the transport routines to the σ_z due to transport.
The σ_z due to transport is perturbated in subroutine ops_vertdisp:

```
sz = varin%varin_unc%sigz_fact*sz.
```

Here the σ_z after interpolation between the values calculated in the surface layer routine and the boundary layer routines (*sz* on the right hand side) is multiplied with a value prescribed in the structure *varin%varin_unc%sigz_fact*. This factor is prescribed in the ops_varin.txt file.
Subroutine ops_vertdisp is called twice in subroutine ops_stab_rek, once to calculate σ_z at the receptor using stability parameters from the source site (*sz_rcp_stab_src*), and once at the receptor using stability parameters from the receptor site (*sz_rcp*). It is assumed that the vertical dispersion length follows the formula ax^b. In other parts of the code, OPS still follows this equation. Hence the calculated σ_z due to transport, to be precise: the average of *sz_rcp_stab_src* and *sz_rcp*, is used to determine factor *dispg* (equal to *a* in ax^b) as follows:

```
dispg = (sz_rcp_stab_src + sz_rcp)*0.5 / (x**DISPH(istab)).
```

Here *DISPH(istab)* equals *b* in ax^b. This is a hard coded value dependent on the stability class. Variable *dispg* is also calculated separately for each stability class. Variables *dispg* and *DISPH* are used in other parts of the code together with the distance to recalculate σ_z. This happens in

subroutine ops_conlt_trans_disp when the plume is entirely above the mixing layer as this is still needed for wet deposition. Otherwise this happens in ops_conlt_par_oppbr for area sources, or in ops_conlt_par_puntbr for point sources. The initial dispersion length is added quadratically to the dispersion length calculated using *dispg* and *DISPH* in ops_conlt_par_oppbr and ops_conlt_par_puntbr. This final σ_z is used to calculate the concentration at the receptor.
Note that for area sources in ops_conlt_par_oppbr the final σ_z due to transport is a combination of two σ_z's (depending on the distance with respect to the edge of the area source), while additionally a constant value of 0.1 is included to calculate a new σ_z. This 0.1 is stated to represent the distribution of source heights within the area source and could be regarded as an additional minor initial vertical dispersion length. This causes that the actual multiplication factor differs slightly from the multiplication factor applied in ops_vertdisp. However, it was decided not to correct for this, since this 0.1 is not part of the σ_z caused by atmospheric diffusion / transport. Furthermore, this value is so small that the impact is expected to be negligible.
Furthermore, *dispg* is used in ops_depoparexp to calculate when the plume becomes fully mixed, while in that same subroutine σ_z is used to determine the source depletion for coarse particles. Within subroutine ops_brondepl both are also used inside the source depletion terms for dry deposition for all phases of the plume: inside the area source, when the plume is not yet well mixed and when the plume is well mixed.
Note that for area sources when the receptor is outside the area source, ops_vertdisp is called again to determine the σ_z at a distance of twice the radius *s2*. Subsequently a new *sigzr*, being the vertical dispersion length calculated for an area source, is calculated using both *s2* and the plume height at the receptor. Due to that the multiplication factor for the sensitivity analysis is applied inside ops_vertdisp and an area source representative *sigzr* is calculated using additionally the plume height at the receptor, not the entire *sigzr* is adjusted according to the multiplication factor. For the study presented here this is not an issue, since area sources are not included in the source types. However, for future research area sources may be wanted to be tested in a sensitivity analysis as well. As such, first *s2* was divided by *varin_unc%sigz_fact*. Subsequently, after the calculation of *sigzr*, *sigzr* was multiplied by *varin_unc%sigz_fact*:

```
s2 = s2 / varin%varin_unc%sigz_fact.
sigzr = s2 / alog((htot + s2) / htot).
sigzr = varin%varin_unc%sigz_fact * sigzr.
```

This *sigzr* is used to calculate the effective plume height of the area source (*hf*) as well as a wind speed representative for the plume over the area source (*uxr*) which is again used to calculate an average wind speed for the entire travelled distance (*ugem*). Within subroutine ops_brondepl, variable *sigzr* is used in the loss rate term for dry deposition inside the area source, while *ugem* is used inside the source depletion term for dry deposition when the plume is not yet well mixed. Variable *ugem* is furthermore used to calculate the average wind speed over the trajectory (*utr*) in ops_depoparexp which is used in the source depletion terms for chemical conversion and wet deposition.

Finally, the various terms also influence the source depletion terms of the secondary material, as well as the calculation of the concentration of the secondary material.

2.3 Boundary layer height

This Section describes the adjustments in OPS-LT to study the sensitivity to uncertainty in the boundary layer height. A meteorology pre-processor (METPRO) is used before reading the processed meteorology OPS-LT. In METPRO the boundary layer height is derived from primary meteorological data and further analyzed for statistical use in OPS-LT.
Reading this data and processing it for proper use within OPS-LT occurs in OPS-LT subroutine ops_readstexp by reading it with subroutine ops_readdata and scaling it inside a loop over the various meteorological variables using the scaling factor linked to the specific meteorological variable[1] to obtain the final meteorological value. Hence it was decided to perturb the boundary layer height right after the scaling occurs with:

```
astat(:,2,:,:) = varin_unc%unc_meteo%xl_fact *
                 astat(:,2,:,:).
```

where *astat(NTRAJ, NCOMP, NSTAB, NSEK)* is the array used to store the meteorological parameters with *NTRAJ* the number of distance classes, *NCOMP* the number of components in the meteorological input data, *NSTAB* the number of stability classes and *NSEK* the number of wind sectors. The boundary layer height is indicated with *NCOMP* = 2. The original boundary layer height is multiplied with a value prescribed in the structure *varin_unc%unc_meteo%xl_fact*. This factor is prescribed in the ops_varin.txt file.
Later on in the code (in subroutine interp_sek), the *astat* data (directly or via *stt* which is the variable for which meteorological parameters have been interpolated between distance classes) is used to determine some specific boundary layer height variables:

- *xl*: the maximal mixing height over the transport distance
- *xloc*: the local mixing height near the source
- *xl100*: the mixing height at 100 km

When the boundary layer height from the provided meteorological data equals zero, the local boundary layer height *xloc* may be set to a default value. This is prescribed in subroutine interp_sek in variable *MENGH* with:

```
INTEGER, PARAMETER :: MENGH(NSTAB) =
                  (/ 300, 985, 302, 537, 50, 153/ ).
```

As such, this also needs to be multiplied with the multiplication factor of the uncertainty analysis. Since *MENGH* is defined as a constant parameter in the attributes list, a new scaled variable is defined (*mengh_scaled(NSTAB)*) and varied in a similar way as *astat(:,2,:,:)*, using FLOAT to convert an integer to a real:

[1] For instance, wind speed is provided in 0.1 m/s and will be scaled to m/s. Scaling factor can also be 1.0 as is the case for the boundary layer height.

```
mengh_scaled = varin_unc%unc_meteo%xl_fact * FLOAT(MENGH(:)).
```

Subsequently, when the read meteorological data provides a 0-value, the code for determining *xloc* is changed from `xloc = FLOAT(MENGH(istab))` to `xloc = mengh_scaled(istab)`.Furthermore, the effective transport height for the well mixed plume is set at `0.5·xl`. However, this is limited to the value of parameter *HUMAX* being 500 m. As such, altering the boundary layer height without altering *HUMAX* would result in an unaffected maximal plume height when the multiplication factor of *xl* > 1, while this 500m is relatively high for runs with a multiplication factor < 1. Therefore it was decided to also define a new scaled variable for *HUMAX*, called *humax_scaled* which is varied similarly as *mengh_scaled*. Since *HUMAX* is defined in a module where various constants are set and not in a subroutine with supplied arguments, we cannot provide the ops_varin-variable to this module. Hence, it was decided to vary *HUMAX* directly in the subroutines where it is used with:

```
humax_scaled = varin%varin_unc%unc_meteo%xl_fact * humax.
```

This is done in ops_conlt_trans_disp where transport and dispersion parameters are calculated, in ops_depoparexp where parameters needed for dry deposition, wet deposition and chemical conversion are calculated and in ops_brondepl where the source depletion terms are calculated.

Note that the boundary layer height was perturbed in the code of OPS-LT only after reading the meteorological data derived by the preprocessor METPRO. This statistical meteorological data also contains tables with the distribution of the stability classes over the day, both source and receptor oriented. These give the percentage of the amount of hours that a certain stability and mixing layer height combination occurs in 2-hourly blocks, which are used to account for diurnal variation in emissions. With a higher or lower mixing layer height around the predetermined 'threshold' of mixing layer height associated with the respective stability class, it may occur that the frequency of hours would need to be distributed differently. This however is not adjusted in the here presented sensitivity analysis.

2.3.1 Impact of altered boundary layer height

Variables *xl* and *xloc* are used in subroutine ops_stab_rek where they are used in ops_plumerise (in OPS-lib, renamed to *zmix* and *zmix_loc*) in ops_plume_penetration. This subroutine determines variable *onder* (part of the plume below the mixing height) and *hemis1* (emission height including plume rise due to buoyancy or momentum; this value is adjusted to *zmix* when the original *hemis1* is higher than *zmix* AND *onder* > 0). As such, besides that another mixing height influences the mixing volume, it also influences the fraction of the plume inside the mixing layer and hence directly the concentration since the concentration due to transport and dispersion is multiplied with *onder* to only take into account the dispersion and transport of the material inside the mixing layer. Furthermore it may impact the height of the emission

point with respect to the part of the plume inside the mixing layer. Both are mostly of importance for relatively higher sources.
Variable *xl* is furthermore used in the call to ops_vertdisp (where *xl* is renamed to *zi*) in ops_stab_rek. In ops_vertdisp, it is used to determine the interpolation factor between surface layer and mixing layer (*fm*) and the interpolation factor between the convective and near neutral layer (*fs*). Subsequently, *zi* is used in routines ops_convec, ops_neutral en ops_surface to determine the plume height *zu*, the wind speed at plume height at distance *dsx* and the vertical dispersion coefficient *sigz*. In all routines variable *zi* is used directly in the determination of *sigz*, while it is used to determine when the plume is reflected against the boundary layer height as well. When the plume touches both the ground and the mixing height and/or when the plume becomes well mixed, the plume height is set equal to half the mixing height.
After ops_stab_rek, *xl* is stored in *xlm* since later in the code *xl* may be overwritten. Firstly, this may occur in ops_conc_ini for heavy particles (when the settling velocity of that particle size class > 0.02 m/s in OPS-LT, not relevant for this study). When the mixing layer height is less than the source height including plume rise due to buoyancy/momentum and including plume descent due to settling (*htt*), the mixing layer height is set to 2**htt*. The reason behind this is that it is assumed that for course particles the boundary layer height should not be of importance since the settling process is not hampered by the inversion layer. As such, this increase of boundary layer height is used as a sort of 'work around' to ensure that the plume is entirely within the mixing layer.
The other time that *xl* may be overwritten is in ops_conlt_trans_disp in ops_conltexp (in ops_conc_ini). When the plume is entirely above the mixing layer (*onder* = 0), the concentration at the ground level is 0, but some plume information is still needed to determine the wet deposition which takes into account the entire column. When the vertical dispersion length *sigz* is less than 40 m, *xl* is set to 80 m, otherwise *xl* is set to 2**sigz*.
When the plume is not entirely above the mixing layer, *xl* is used to determine the phase of the plume and an index into different transport distance regimes. When *sigz* > 1.6·*xl* the plume is assumed 'well mixed'. Furthermore it is used to distinguish between the intermediate and the short distance. For the short distance there is no reflection at the mixing layer, for the intermediate distance this is taken into account. Furthermore it is used to determine the effective transport height for the intermediate distance.
After the call to ops_conlt_trans_disp in ops_conltexp, subroutine ops_conlt_gauss_zrcp is called where *xl* is used directly in the concentration calculation when the plume is well mixed, and is otherwise used to determine the reflection terms at the surface and the boundary layer which are used within the concentration calculation. With higher/lower boundary layer height, reflection at the boundary layer will start at a further/shorter distance respectively.
In short regarding the transport routines: the boundary layer height at the receptor *xl* is used to determine the phase of the plume (not yet well mixed or well mixed), to determine the effective transport height when the plume is well mixed or at intermediate distance, to determine when or if reflection at the boundary layer and the surface takes place and to determine the concentration due to transport and dispersion. With

higher/lower boundary layer height, reflection at the boundary layer will start at a further/shorter distance respectively while furthermore there will be more/less dilution.

The next use of *xl*, *xloc* and *xl100* is in subroutine ops_depoparexp after ops_conc_ini. Here the average dry deposition velocity (*vd)* over the trajectory is calculated (*vd_uncorr_trj_zra)*. The initial *vd* needs to be corrected to take into account the amount of time that the plume is above the mixing height and no deposition takes place. The local mixing layer height *xloc* is compared with the plume height at the source to determine whether a correction for high sources or for low sources needs to be applied.
In subroutine par_nat called within ops_depoparexp the maximum mixing layer height at the receptor *xl* is used as thickness over which the wet deposition takes place for plumes (partly) inside the mixing layer for in-cloud scavenging. The loss rate due to wet deposition decreases with increased mixing height. Variable *xl* is also used in a help variable to determine the distribution factor between washout (below cloud) and rainout (in cloud) and inside the determination of *cratio*. This is the ratio between the concentration at the surface and the concentration occurring with full mixing in the boundary layer and is used for reversible wet deposition.
Back in ops_depoparexp *xloc* and *xl100* are used to determine at which distance the plume touches the ground (*xg*) for stable conditions. Otherwise, only *xloc* is used to determine when the plume touches the ground.
All three mixing layer variables and *xg* are used in the call to ops_brondepl, the subroutine which calculates the source depletion factors for dry deposition:

- For distances past *xg* the plume is assumed fully mixed. In that case *xl* and *xg* are used in the calculation of the source depletion factor of dry deposition for when the plume is well mixed. Distance *xg* is then also the furthest distance of phase 2 for which then a *sigzxg* as well as a mixing layer height *xlxg* are determined using *xg* and an interpolation of *xloc* and *xl100*. The effective transport height *hf* at *xg* is *xlxg*/2, limited by *humax_scaled*. This is then used to determine the wind speed at *xg*, and the average wind speed over the trajectory for phase 3, *ugem*. Variables *xl*, *xg* and *xlxg* are used to determine the undepleted concentration at distance *xg*, which is later used in the source depletion term for dry deposition for phase 2 of the plume. Finally, *ugem* is used to correct the vertical profile due to the extraction of material in the atmosphere at the surface due to dry deposition. For phase 3 of the plume, this equals the above mentioned *ugem* which is influenced by the mixing layer height.
- After the if-statement for a well-mixed plume, a new if-statement is entered, now for when there is an area source. When the distance is further than the edge of the area source, subroutine ops_vertdisp is called with *xl* (renamed to *zi*) where it is used determine the vertical dispersion length, plume height and wind speed at the radius distance of the area source. This is used to determine the effective plume height of the area source, the effective wind speed of the area source, and the average wind speed over the trajectory (hence taking into account the distance

and wind speed inside and past the area source). When the vertical dispersion length for the area source *sigzr* is larger than the mixing height at 100 km, *xl100*, *sigzr* = *xl100*. This *sigzr* is used to determine the dry deposition rate for the area source.
- The source depletion for phase 2 of the plume (past the area source, but not yet well mixed) uses a vertical dispersion length as well which is possibly influenced by the mixing layer height via reflections. This equals the *sigzxg* mentioned above in cases that the total travelled distance > *xg*.
- Finally the source depletion for phase 1 (inside the area source) for all depletion processes is determined using several of the variables influenced by the mixing layer height: the dry deposition rate for the area source, the loss rate due to wet deposition and the wind speed representative for the plume over area source.

In short regarding the routine to calculate deposition parameters, the boundary layer height is used:
- To correct the dry deposition velocity over the trajectory for the fact that the plume is sometimes higher than the boundary layer height such that dry deposition does not take place. In other words: the effective dry deposition velocity over the trajectory becomes smaller when the plume height at the receptor exceeds the mixing layer. With higher mixing layer height, this will occur less often, and effective dry deposition velocity overall will be larger.
- As thickness over which the wet deposition takes places for plumes (partly) inside the mixing layer for in-cloud scavenging. The loss rate due to wet deposition decreases with increased mixing height.
- In the (smooth) distribution factor of washout (pr = 0) and rainout (pr = 1). The combined below- and in-cloud scavenging rate is usually much higher than the below cloud scavenging rate, but in-cloud scavenging can only have affect if the pollutant is able to penetrate clouds. Cloud base height is assumed equal to the mixing height. So for higher mixing layer heights, it will take longer for a plume emitted at a certain height to reach the mixing layer height, and the distribution factor will remain smaller for a longer traveling distance resulting in less efficient scavenging.
- For calculating the ratio between the concentration at the surface and the concentration occurring with full mixing in the boundary layer which is used for reversible wet deposition. This ratio increases with increased mixing height when the plume is not yet fully mixed (though the mixing layer is also used to determine the vertical dispersion length in this case). Otherwise the mixing layer term is found in both nominator and denominator and there is no impact. This ratio is multiplied directly with the washout and rainout scavenging and hence directly found in the loss rate due to wet deposition.
- To determine when the plume touches the ground which determines which phase the plume is in (fully mixed or not yet fully mixed) while additionally it is used in the various source depletion terms for dry deposition.

    - The source depletion term for the well mixed phase increases with increased mixing height, which would result in lower concentrations at the receptor. However, it also depends on the distance travelled in the well mixed state, which will be smaller with higher mixing layer height, since a fully mixed phase will only be reached at greater distances. This can give non-linear results.
    - The source depletion term for the not yet well mixed phase depends on the vertical dispersion length and wind speeds at effective transport height and average over the trajectory so far which may be impacted by the mixing height and can give non-linear results.
    - The source depletion inside the area source for all depletion processes is determined using several of the variables influenced by the mixing layer height: the dry deposition rate for the area source, the loss rate due to wet deposition and the wind speed representative for the plume over area source and as such can give non-linear results as well.
    - For coarse particles source depletion also depends on the vertical dispersion length which may be impacted by the mixing height and can give non-linear results.
- The average wind speed impacted by the mixing layer height is used to correct the gradient factor used for the vertical profile impacted by dry deposition over the trajectory.

After the call to ops_depoparexp, both *xl* and *xloc* are used in ops_conc_rek where they are only used in ops_seccmp. Here *xl* is used for area sources to set the vertical dispersion length to half the mixing layer height when the original vertical dispersion length is larger than half the mixing layer height. This is subsequently used to determine the wind speed at transport height. Furthermore *xl* is used for the distribution factor between washout and rainout for the secondary material, similar as for the primary material. It is also used as thickness over which the rainout of secondary material occurs, similar as for the primary material. The loss rate due to in-cloud scavenging decreases with increased mixing layer. The dry deposition velocity averaged over the transport distance is also corrected for the fact that the plume is sometimes higher than the mixing layer such that deposition does not take place, using *xloc*. With higher *xloc*, this will occur less often, and effective dry deposition velocity overall will be larger.
In subroutine seccd, used to compute the cross wind integrated mass fluxed for primary and secondary material, variable *xl* is used as well. For area sources, when 1.5 times the vertical dispersion length is larger than the mixing layer height, the effective plume thickness is set to the mixing layer height. This is used in the source depletion term for dry deposition for area sources. This source depletion increases with increased mixing height. For area sources, this source depletion term is used as a help variable in determining the distance over which the production of the secondary species takes place. For point sources this equals the effective travel distance. Then *xl* is also used in the source depletion term for dry deposition of both the primary and secondary material with increased source depletion for increased mixing layer. This is used to calculate how much primary and secondary material is lost

per time step, which is subsequently used to calculate the cross-wind integrated mass flux of the secondary species. Back in subroutine ops_seccmp, *xl* is used to compute the concentration of secondary species using the Gaussian plume in not well mixed conditions (for the reflection terms) as well as fully mixed conditions (for the mixing volume).

2.4 Chemical conversion – k_{chem}

For 'chemical conversion', the sensitivity to uncertainty in chemical conversion over the trajectory was tested. Hence, the relevant term in OPS-LT is the term for source depletion due to chemical conversion. Source depletion accounts for the effect of removal of material over the trajectory on the atmospheric concentration. In other words, it is the reduction of the source mass due to loss over the trajectory, in this case removal due to chemical conversion. This effect is amongst others dependent on the distance from the source. The depletion factor due to chemical conversion is determined in subroutine ops_depoparexp with:

```
cch = cch * EXP( - (a/utr * vchem/360000.)).
```

Here *cch* on the left hand side is the source depletion term which accounts for the loss of mass due to chemical conversion. The *cch* on the right hand side equals 1.0 in this part of the code, i.e. no form of source depletion has been accounted for yet. Parameter *a* (m) represents the effective distance over which chemical conversion occurs, *utr* (m/s) is the average wind speed over the trajectory, *vchem* (%/h) is the chemical conversion velocity of the emitted substance and the factor 360000 converts %/h to fraction/s. Furthermore, *vchem* is used directly in the source depletion term representative for depletion inside the area of an area source *cq1* (since there is no sensitivity analysis for area sources in this study, this is not elaborated on further).
Since OPS-LT version 5.0.0.0 the chemical conversion velocity for SO_2, NO_x and NH_3 can be calculated based on the output of the EMEP model. When this is desired, the flag *iopt_vchem* must be set to 1 (set by setting input variable 'CONVRATE' to 'EMEP' in the control file). For *iopt_vchem = 0* or other substances than SO_2, NO_x and NH_3 a standard parametrisation is used based on meteorological conditions (including light intensity), season and part of the day. In the case of user defined substances, the user must provide a fixed conversion value. In all cases the value of *vchem* represents an average over the trajectory. Hence, after the calculation of *vchem* this value is perturbed to study the impact of uncertainty in chemical conversion.
Since *vchem* can be determined in 3 ways, this value must be perturbated in 3 places in OPS-LT:

1. for when EMEP conversion rates are used (in subroutine ops_par_chem)
2. for when the standard parametrisation for the acidifying substances SO2, NOx or NH3 is used (in subroutine ops_vchem)
3. for non-acidifying / other substances (in subroutine ops_vchem).

Perturbation occurs as follows:

```
vchem = varin_unc%unc_sourcedepl%vchem_fact * vchem
```

Here the original *vchem* is multiplied with a value prescribed in the structure varin_unc%unc_sourcedepl%vchem_fact. This factor is prescribed in the ops_varin.txt file.
Besides that *vchem* is used for the source depletion terms over the trajectory (*cch* and *cq1* for outside and inside area sources respectively) which influences the concentration of the primary species via the depleted source strength, *vchem* is also used in the production term of the secondary species and the loss term of the primary species used in the calculation for the secondary species.

2.5 Dry deposition – $v_{d,pri}$ and $v_{d,sec}$

For 'dry deposition', the sensitivity to uncertainty in the dry deposition velocity over the trajectory used for the source depletion was tested. Source depletion accounts for the effect of removal of material over the trajectory on the atmospheric concentration. In other words, it is the reduction of the source mass due to loss over the trajectory, in this case removal due to dry deposition. This effect depends, amongst other things, on the distance from the source. The deposition velocity at a specific receptor point only depends on the surface properties and the meteorological conditions at the location of the receptor. Since local deposition velocity is independent of the source receptor distance, the local v_d was initially deemed not relevant for determining the uncertainty in concentration and deposition at a specific location for a specific source. Yet, in order to use uncertainty information on v_d over the trajectory and at specific locations in a consistent way, it was decided to vary the local dry deposition as well. This enables a study of the full sensitivity to dry deposition, also in comparison with the sensitivity to other parameters. For practical reasons this was done in a post-processing step (i.e. after the simulations have been finished). In similar future efforts local v_d might be perturbed within OPS as well.
The amount of source depletion due to dry deposition of the primary species depends on the phase of the plume: when the plume is well mixed (phase 3), when the plume is not yet well mixed (phase 2) and (if present) within an area source (phase 1). Note that for a plume in a phase beyond phase 1, also the source depletion for the earlier phases is taken into account. For instance, for a plume in the well mixed state, also source depletion when the plume was not yet in the well mixed state needs to be accounted for, as well as source depletion in the area source when the emission is not a point source.
Adjusting the source depletion directly influences the concentration since the concentration resulting from dispersion and transport is multiplied with the source depletion term. It influences the secondary material as well, since the amount of primary material in part determines how much can be converted to the secondary material. The OPS-LT adjustments for source depletion due to dry deposition of the primary material are described in Sections 2.5.1, 2.5.2 and 2.5.3 for the specific plume phases.
Source depletion due to dry deposition of the secondary material is also accounted for. This is described in Section 2.5.4.
The post-processing methodology for perturbing the dry deposition at the receptor points consistent with the perturbation of $v_{d,pri}$ applied for the source depletion over the trajectory, is described in Section 2.5.5.

2.5.1 *Source depletion phase 3: well mixed plume*

The source depletion term for dry deposition for phase 3, *cdn*, is determined in subroutine ops_brondepl with:

```
cdn = EXP( - ((disx - xg)/ueff * vd_eff_trj_zra/xl)).
```

Here *disx* is the source receptor distance, *xg* the distance at which the plume is assumed to have become well mixed, *ueff* the windspeed at the effective transport height, *xl* the boundary layer height and *vd_eff_trj_zra* the effective deposition velocity over the trajectory taking into account the amount of time that the plume is above the mixing height and no deposition takes place. Variable *vd_eff_trj_zra* depends on the source height, as well as the substance (e.g. coarse particles or gasses).
To ensure that for future sensitivity analyses all substances can be calculated, it was decided to adjust the deposition velocity over the trajectory directly after the determination of *vd_eff_trj_zra* (in subroutine ops_depoparexp) with:

```
vd_eff_trj_zra =
varin%varin_unc%unc_sourcedepl%vd_drydep_pri_fact *
vd_eff_trj_zra.
```

Here the original *vd_eff_trj_zra* is multiplied with a value prescribed in the structure varin%*varin_unc%unc_sourcedepl%vd_drydep_pri_fact*. This factor is prescribed in the ops_varin.txt file.
Besides that *vd_eff_trj_zra* is used for the source depletion term over the trajectory for phase 3 (*cdn*) which influences the concentration and hence dry and wet deposition of the primary species via the depleted source strength, it is used in the subroutines where the secondary species are determined (ops_seccmp and seccd). Here it is also used for the source depletion factor of the primary species, which is used for the loss term of the primary species, which is in its turn used for the cross-wind integrated mass flux of the primary species (*qpri)*. This is input for the production term of secondary species as well as the loss term of the secondary species. Both production and loss terms are of influence on the cross-wind integrated mass flux of the secondary species (*qsec*) and hence concentration of the secondary species (*consec*). Variables *consec* and *qsec* are used in subroutine ops_conc_rek to determine respectively the dry and wet deposition of the secondary species at the receptor.
In short: a variation in *vd_eff_trj_zra*, used for source depletion due to dry deposition of the primary material, impacts both primary and secondary species. For instance, when there is less deposition of the primary substance over the trajectory, more primary material remains which can be converted to the secondary species. Since dry and wet deposition are linked to the concentration or the mass in the atmospheric column, both deposition terms are impacted, both for the primary and secondary material. However, the source depletion term for dry deposition over the trajectory and the deposition terms for deposition at the receptor of the *secondary* material are NOT directly influenced by varying *vd_eff_trj_zra*.

2.5.2 *Source depletion phase 2: plume not yet well mixed*

The source depletion term for dry deposition of phase 2 of the plume (not yet well mixed) is also determined in subroutine ops_brondepl:

```
cq2 = EXP( -(2.*al/qbstf*1.e-6*vd_trj_z0/12*2*PI*(xx +
virty)*cxx*ueff/ugem/onder*(xx - radius)*(1.-cgt))).
```

Here *al* is a help variable[2], *qbstf* the source strength, *vd_trj_z0* the deposition velocity over the trajectory calculated at z_0 (roughness length), *xx* the representative distance for phase 2 of the plume (within phase 2 this is the source-receptor distance, for plumes in phase 3 this equals *xg*), *virty* the distance between a virtual point source and the center of an area source (for point sources *virty* = 0), *cxx* the representative concentration for phase 2 of the plume (for phase 2 it holds that *cxx* = *c*, being the concentration due to dispersion and transport without taking into account removal processes and without correction for the vertical gradient, for phase 3 cxx is the undepleted concentration at distance *xg*), *ueff* the wind speed at the receptor at effective transport height, *ugem* the average wind speed dependent on the phase of the plume, *onder* the fraction of the plume below the boundary layer height, *radius* the radius of the area source (0 for point sources) and *cgt* is 1 minus the concentration gradient between 4 and 50 m height. This concentration gradient is determined at the receptor but is corrected for the possibility that the gradient has not yet fully developed (at shorter distances for instance).
For the sensitivity analysis variable *vd_trj_z0* is varied. This variable is determined in subroutine ops_depoparexp and dependent on the substance (e.g. coarse particles or gasses). To ensure that for future sensitivity analyses all substances can be calculated, it was decided to adjust the deposition velocity over the trajectory directly after the determination of *vd_trj_z0* with:

```
vd_trj_z0 = varin%varin_unc%unc_sourcedepl%vd_drydep_pri_fact
* vd_trj_z0.
```

Here the original *vd_trj_z0* is multiplied with a value prescribed in the structure varin%*varin_unc%unc_sourcedepl%vd_drydep_pri_fact*. This factor is prescribed in the ops_varin.txt file.
Variable *vd_trj_z0* is only used for the source depletion term over the trajectory for phase 2 (*cq2*) which influences the concentration and hence dry and wet deposition of the primary species via the depleted source strength. Though an adjusted primary concentration impacts the secondary material by that another amount of primary material can be converted, varying this parameter has no consequences for other parts of the code.

[2] `al = 8./PI*sh/((1.+SQRT(1.+8/PI*sh))**2)` with `sh = (sigzxg/htot)**2` where *sigzxg* is the vertical dispersion length σ_z at $x = xg$, with *xg* the distance at which the plume is assumed to be júst well mixed, and *htot* the plume height at the receptor. PI = 3.14159265358979.

2.5.3 *Source depletion phase 1: area source*

The source depletion term for dry deposition of phase 1 of the plume (inside an area source) is also determined in subroutine ops_brondepl. The total depletion factor is:

```
cq1    = EXP( -(k_drydep_src*dxeff/uxr)).
```

Here *k_drydep_src* is the conversion velocity for dry deposition (1/s), *dxeff* the effective distance over which the deposition takes place and *uxr* the wind speed representative for the area source. Variable *dxeff* is of importance since this is determined with a help variable *kt_area* which contains information on the chemical conversion and wet and dry deposition:

```
kt_area = ((vchem + vnatpri)/3.6e5 +
      k_drydep_src)*diameter/uxr.
dxeff = diameter/4.*EXP( -kt_area/18.).
```

Here *vchem* is the chemical conversion rate (%/u), *vnatpri* the loss rate due to wet deposition of the primary compound (%/u) and *diameter* the diameter of the area source. Factor 3.6e5 is the conversion from percentage per hour to fraction per second.
For the sensitivity analysis, variable *k_drydep_src* is varied. This variable is determined locally in subroutine ops_brondepl. As such is wat decided to adjust the loss rate due to dry deposition within the area source directly after the determination of *k_drydep_src* with:

```
k_drydep_src =
varin%varin_unc%unc_sourcedepl%vd_drydep_pri_fact *
k_drydep_src.
```

Here the original *k_drydep_src* is multiplied with a value prescribed in the structure varin%*varin_unc%unc_sourcedepl%vd_drydep_pri_fact*. This factor is prescribed in the ops_varin.txt file.
Variable *k_drydep_src* is only used for the source depletion term over the trajectory for phase 1 (*cq1*) which influences the concentration and hence dry and wet deposition of the primary species via the depleted source strength. Though an adjusted primary concentration impacts the secondary material by that another amount of primary material can be converted, varying this parameter has no consequences for other parts of the code. Furthermore, this is only of influence for area sources. For point sources, as is the case in this study, *cq1* equals 1.0, i.e. no source depletion.

2.5.4 *Source depletion secondary species*

Note that the variables *vd_eff_trj_zra*, *vd_trj_z0* and *k_drydep_src* are used for the primary species only. As such, source depletion for the secondary species is not accounted for. This must be done separately. The source depletion for dry deposition of the secondary species is also determined using a dry deposition velocity though the equation is somewhat different since a numerical time stepping scheme is used here. In the loop over the time steps, the loss term is calculated and the concentration of secondary species is updated. The amount of time

steps for which this is done depends on the distance. The source depletion factor due to dry deposition for secondary species *e1_sec* (-) is calculated in subroutine seccd with:

```
e1_sec = e1_sec*exp( - dt*vd_sec_eff_trj_zra/xl).
```

On the right hand side *e1_sec* equals 1.0 (no source depletion yet), on the left hand side this is only source depletion due to dry deposition. Variable *dt* is the time step (s), *vd_sec_eff_trj_zra* is the effective deposition velocity (m/s) for the secondary compound for the trajectory at height *zra* (the height at which it is assumed that the concentration profile is undisturbed by deposition, 50m), taking into account the time that the plume is above the mixing layer and there is no deposition. Variable *xl* is the maximal mixing height over the transport distance (m). The source depletion due to dry deposition for secondary species determined in seccd does not distinguish between the various plume phases. Therefore it is sufficient to only vary *vd_sec_eff_trj_zra* for the sensitivity analysis, in a similar way as for the primary species. Since variable *vd_sec_eff_trj_zra* depends on the source height, it was decided to adjust this value directly after these calculations have occurred in subroutine ops_seccmp with:

```
vd_sec_eff_trj_zra =
varin_unc%unc_sourcedepl%vd_drydep_sec_fact *
vd_sec_eff_trj_zra.
```

Here the original *vd_eff_trj_zra* is multiplied with a value prescribed in the structure varin_unc%unc_sourcedepl%vd_drydep_sec_fact. This factor is prescribed in the ops_varin.txt file. Note that after the call to subroutine seccd, a correction is made using the 'exact' depletion factor for primary species to account for source depletion for the various phases of the plume.
Variable *vd_eff_trj_zra* is only used for the source depletion term of the secondary material which influences the concentration and hence dry and wet deposition of the secondary species via the depleted source strength. Varying this parameter has no consequences for other parts of the code.

2.5.5 *Dry deposition at the receptor*

For consistency, we wish to apply the same perturbation of deposition velocity v_d locally, at the receptor, and over the trajectory. This Section describes the pragmatic solution in the present research, where the "disturbed" dry deposition flux at the receptor point, $F_{d\_rcp,disturbed}$, is computed in a post-processing step. This was done as follows.
We first acknowledge the fact that the local dry deposition at a receptor point has no influence on the depletion and hence the local atmospheric concentration. Only after integration over longer distances, v_d (and the related dry deposition flux F_d) will affect atmospheric concentration. We also acknowledge the fact that the concentration in air arriving at a specific receptor point C_{rcp}, includes depletion effects and hence effects of the perturbed v_d over the trajectory between the source and the receptor. Third, we note that the local deposition from the output of OPS is based on the product of C_{rcp} and a local, internally computed and unperturbed value of v_d, here called v_{d_rcp}. Fourth, we assume that

(relative) effects of the dry deposition flux on the concentration profile during transport over the trajectory between the source and the receptor and those at the receptor are roughly the same on average, so that possible differences may be ignored.
These considerations imply that the local, disturbed dry deposition $F_{d\_rcp,disturbed}$, consistent with the perturbation of depletion over the trajectory, can be obtained as:

$$F_{d\_rcp,disturbed} = f_{vd} * v_{d\_rcp} * C_{rcp},$$

in which f_{vd} is the perturbation factor applied to dry deposition over the trajectory. Concentration C_{rcp} is output from OPS. We would obtain the implied value of $v_{d,rcp}$ as:

$$v_{d\_rcp} = F_{d\_rcp} / C_{rcp},$$

where F_{d_rcp} is the local value of the dry deposition at the receptor point, which is also output from OPS and which only includes effects of (disturbed) depletion over the trajectory. Hence, we simply use:

$$F_{d\_rcp,disturbed} = f_{vd} \times F_{d\_rcp}.$$

We apply this post-processing step to both primary and secondary compounds. For the primary and secondary material f_{vd} equals *vd_drydep_pri_fact* and *vd_drydep_sec_fact* respectively from structure varin%varin_unc%unc_sourcedepl defined in the OPS_varin.txt file.

2.6 Wet deposition

For 'wet deposition' the sensitivity to wet deposition over the trajectory, used for the source depletion, as well as at the receptor was tested. Source depletion accounts for the effect of removal of material over the trajectory on the atmospheric concentration. In other words, it is the reduction of the source mass due to loss over the trajectory, in this case removal due wet deposition. The source depletion term for wet deposition is determined in subroutine ops_depoparexp with:

```
cch = cch*EXP( -(a/utr*vnatpri/360000.)).
```

Here *cch* on the right side is the source depletion term in which the effect of mass loss due to chemical conversion is already accounted for. The *cch* on the left hand side is the source depletion term including the mass loss due to wet deposition. Parameter *a* gives the effective distance over which wet deposition occurs, *utr* is the average wind speed over the trajectory. Parameter *vnatpri* gives the mass loss rate due to wet deposition of the primary substance (%/h), and 360000 the conversion factor from %/h to fraction per second.
Contrary to passing the uncertainty through the dry deposition velocity for dry deposition, the parametrisation for wet deposition does not allow a variation through a wet deposition velocity term. Hence, it was decided to vary the mass loss rate *vnatpri*. This is calculated in subroutine par_nat with:

```
vnatpri = cratio*(vnatwash*(1. - pr) + vnatrain*pr).
```

Here *cratio* is the ratio between concentration at the surface and concentration at full mixing in the boundary layer (used for reversible wet deposition, otherwise *cratio* = 1), *pr* represents the distribution between washout (below cloud) and rainout (in cloud) and is dependent on the plume height and boundary layer height combined with the vertical dispersion length. Below-cloud scavenging is important for scavenging from plumes close to sources in situations where there is no interaction with clouds yet (*pr* = 0). At greater distances, when the plume is vertically well mixed and has had the opportunity to penetrate into the cloud base, scavenging occurs only through in-cloud scavenging (*pr* = 1) with a bulk scavenging ratio which integrates all processes within and below the cloud. The terms *vnatwash* and *vnatrain* are the loss rates due to washout and rainout respectively (%/h). Sections 2.6.1 and 2.6.2 discuss the adjustments in OPS-LT needed for the sensitivity analysis for the washout and rainout scavenging rate respectively, for the primary material. Section 2.6.3 describes similar adjustments for the secondary material.

2.6.1 *Below-cloud scavenging (washout, close to the source)*

Variable *vnatwash* is determined in subroutine par_nat with:

```
vnatwash = regenk*100./twt*(1 - EXP( - twt*lambda_b)).
```

Here *regenk* represents the chance of rain and *twt* represents the average duration of the precipitation event dependent on the source – receptor distance. Variable *lambda_b* gives the below-cloud scavenging rate (1/h) and is determined separately for gasses and for particles. Hence it was decided to vary *lambda_b* for the uncertainty analysis in two parts of the code (for gasses and for particles, both in subroutine par_nat):

```
lambda_b = varin_unc%unc_sourcedepl%washout_pri_fact *
lambda_b.
```

Here the original *lambda_b* is multiplied with a value prescribed in the structure *varin_unc%unc_sourcedepl%washout_pri_fact*. This factor is prescribed in the ops_varin.txt file.
Variable *lambda_b* is only used for the *vnatwash* term which in its turn is only used for the *vnatpri* term described at the start of Section 2.6. Variable *vnatpri* is used for the total source depletion term over the trajectory for area sources (inside *cq1*, see Section 2.5.3) and of course for the source depletion term for wet deposition for the distance past the area source (*cch*, start of Section 2.6). Both influence the concentration and hence dry and wet deposition of the primary species via the depleted source strength. Furthermore *vnatpri* is used to calculate the wet deposition flux (µg/m^2/h) of the primary compound at the receptor (*dnatpri*). Finally, *vnatpri* is also used in the subroutines where the secondary species are determined (ops_seccmp and seccd). Here it is also used for the source depletion factor of the primary species, which is used for the loss term of the primary species, which is in its turn used for the cross-wind integrated mass flux of the primary species (*qpri)*. This is input for the production term of secondary species, as well as the loss term of the secondary species. Both production and loss terms are of influence on the cross-wind integrated mass flux of the secondary

species (*qsec*) and hence concentration of the secondary species (*consec*). Variables *consec* and *qsec* are used in subroutine ops_conc_rek to determine respectively the dry and wet deposition of the secondary species at the receptor.
In short: a variation in *vnatpri* impacts both primary and secondary species. For instance, when there is less deposition of the primary substance over the trajectory, more primary material remains which can be converted to the secondary species. Since dry and wet deposition are linked to the concentration or the mass in the atmospheric column, both deposition terms are impacted, both for the primary and secondary material. For the primary material, also the wet deposition flux at the receptor is varied. However, for the secondary material the source depletion term for wet deposition over the trajectory and the deposition terms for deposition at the receptor are NOT directly influenced by varying *vnatpri*.

2.6.2 *In-cloud scavenging (rainout, for greater distances)*

Variable *vnatrain* is determined in subroutine par_nat with either:

```
vnatrain = regenk*100./twt*
            (1 - EXP(-scavcoef/100.*twt))
```

or

```
vnatrain = regenk*100./twt*(1. -
            EXP( - routpri*twt*ri/1000./h)).
```

Here a distinction is made between whether a scavenging rate (*scavcoef,* %/h) is provided, or a scavenging ratio (*routpri*, -). Variable *regenk* represents the chance of rain, *twt* represents the average duration of the precipitation event, *ri* gives the rain intensity (mm/h, hence the extra factor of 1000 to convert from mm to m), and *h* is the thickness over which wet deposition takes place (m).
Determining the amount of scavenging by rainout can occur in three ways:

1. A scavenging rate (%/h) can be provided directly in the input file.
2. A scavenging ratio (-) can be provided directly in the input file.
3. For acidifying compounds (SO_2, NO_x and NH_3) the scavenging ratio is calculated within OPS-LT (in subroutine ops_init and ops_resist_rain_out_scav_ratio).

The scavenging rate for in cloud scavenging, Λ_{in} (%/h), is hence determined with either the predefined $\Lambda_{in} = scavcoef$ or from $\Lambda_{in} = \frac{routpri \cdot ri}{h}$. For non-acidifying gases both *scavcoef* and *routpri* are set in subroutine ops_init, while *routpri* for acidifying compounds and particles is determined in subroutine ops_resist_rain_out_scav_ratio. Hence, to enable a sensitivity analysis, in these locations *scavcoef* and *routpri* are varied with:

`scavcoef = varin%varin_unc%unc_sourcedepl%rainout_pri_fact * scavcoef` (1 location).
`routpri = varin_unc%unc_sourcedepl%rainout_pri_fact * routpri` (2 locations).

Here the original *scavcoef* and *routpri* are multiplied with a value prescribed in the structure (varin%)*varin_unc%unc_sourcedepl%rainout_pri_fact*. This factor is prescribed in the ops_varin.txt file.
Variables *scavcoef* and *routpri* are only used for the *vnatrain* term which in its turn is only used for the *vnatpri* term described at the start of Section 2.6.. The impact of changing variable *vnatpri* is already described in Section 2.6.1.

2.6.3 Secondary species

The source depletion for wet deposition of the secondary species is also determined using a loss rate due to wet deposition similar as for the primary species though the equation is somewhat different since a numerical time stepping scheme is used here. In the loop over the time steps, the loss term is calculated and the concentration of secondary species is updated. The amount of time steps for which this is done depends on the distance. The source depletion factor for secondary species *e1_sec* (-) is calculated in subroutine seccd with:

```
e1_sec = e1_sec*exp( - dt*vnatsec/3.6e5).
```

On the right hand side *e1_sec* equals the source depletion factor due to dry deposition, on the left hand side the source depletion due to wet deposition is also taken into account. Variable *dt* is the time step (s), *vnatsec* is the loss rate due to wet deposition of the secondary compound (%/h), factor 3.6e5 the conversion from %/h to 1/s. Variable *vnatsec* is, as for the primary species, the loss rate which is interpolated between washout (*vnatwashv*, below cloud) and rainout (*vnatrainv*, in cloud) of the secondary species. It is determined in subroutine ops_seccmp with:

```
vnatsec = vnatwashv*(1. - pr) + vnatrainv*pr.
```

Here *pr* represents the distribution between washout and rainout.

2.6.3.1 Washout

For below-cloud scavenging, the washout loss rate *vnatwashv* is determined with:

```
vnatwashv = regenk*100./twt*(1. - EXP( -lambda_b*twt)).
```

Here *regenk* represents the chance of rain (-) and *twt* represents the average duration of the precipitation event (h). Variable *lambda_b* gives the below-cloud scavenging rate (1/h) and is determined right above *vnatwashv*, only for secondary aerosols. As for primary species it was decided to vary *lambda_b* for the uncertainty analysis:

```
lambda_b = varin_unc%unc_sourcedepl%washout_sec_fact *
lambda_b.
```

Here the original *lambda_b* is multiplied with a value prescribed in the structure *varin_unc%unc_sourcedepl%washout_sec_fact*. This factor is prescribed in the ops_varin.txt file.
The varied *lambda_b* for the sensitivity analysis for washout is only used for the washout loss rate for the secondary material. This is, via *vnatsec*, used for the source depletion which influences the concentration and hence dry and wet deposition of the secondary species via the depleted source strength. Furthermore *vnatsec* is used to calculate the wet deposition flux (µg/m^2/h) of the secondary compound at the receptor (*dnatsec*). Varying this parameter has no consequences for other parts of the code.

2.6.3.2 Rainout

For in cloud scavenging, the rainout loss rate *vnatrainv* is determined with:

```
vnatrainv = regenk*100./twt*(1. -
             EXP( -routsec*twt*ri/1000./xl)).
```

Variable *ri* gives the rainfall intensity (mm/h, hence the extra factor of 1000 to convert from mm to m), and *xl* is the thickness of the atmospheric layer over which wet deposition takes place (m). Contrary to the primary species, no distinction is made between whether a scavenging rate or scavenging ratio is provided. Only the in-cloud scavenging ratio for the secondary compound (*routsec*) is provided. This value is hard coded in subroutine ops_init and dependent on the substance that is calculated. Hence is was decided to vary *routsec* after the checks on substance with:

```
routsec = varin%varin_unc%unc_sourcedepl%rainout_sec_fact *
routsec.
```

Here the original *routsec* is multiplied with a value prescribed in the structure *varin_unc%unc_sourcedepl%rainout_sec_fact*. This factor is prescribed in the ops_varin.txt file.
The varied *routsec* for the sensitivity analysis for rainout is only used for the rainout loss rate for the secondary material. The impact of varying *routsec* is described in the previous Section (2.6.3.1).

2.7 Meteorological conditions

Meteorological conditions used as input for OPS are derived from weather observations performed by the Royal Netherlands Meteorological Institute (KNMI). These observations are turned into input suitable for OPS by means of two preprocessors. These preprocessors are intimately linked with OPS. Hence, we describe the procedures followed to obtain meteorological input suitable for application in the present sensitivity in this Chapter of the appendix, although they do not require code in OPS itself.
The meteorological input used by OPS actually represents statistical properties of meteorological conditions. These properties are required

for six meteorological regions or districts as well as for national averages of weather conditions. Regions 1-6 are distinguished mainly according to wind regime. The dataset containing national averages of meteorological conditions is referred to as "Region 0". See Sauter et al. (2023) for further information.
The meteorological conditions are derived from measurements performed at automatic weather stations run by the Royal Netherlands Meteorological Institute (KNMI). Regional statistical properties are obtained from these data in two steps.

1) The pre-processor MPARKNMI is used to derive from the measurements average meteorological conditions per district (Regions 1-6) and for the Netherlands as a whole (Region 0). Depending on the stations' location, local and regional roughness, wind speed and direction are transformed to a standard roughness and height, and subsequently interpolated among the stations and regions, using the stations' position relative to the location of the centre of the regions.
   Output includes hourly wind speed, wind direction and precipitation duration averaged per district and at the national scale. National averages of wind speed and direction, used for the construction of the trajectories, are also incorporated in the hourly output per region. Furthermore, the hourly values of solar radiation, air temperature and relative humidity are provided. In addition, daily averages of the intensity and duration of rainfall events are given at the national scale, as well as the regional daily average of rainfall duration. At present, preprocessing by MPARKNMI yields the same timeseries for solar radiation, air temperature and humidity for each region, based on the average of the Netherlands. Only precipitation characteristics and local wind speed and direction differ among the regions.
2) The pre-processor METPRO is used to derive important secondary variables like mixing layer height and friction velocity. Per region, hourly meteorological data and secondary data derived from them are distributed over each of the meteorological classes distinguished in OPS. These classes are based on 12 wind sectors and six stability regimes. The frequency, i.e., the number of hours for which each class occurs, is determined. This is done for four trajectory distance classes (0, 100, 300 and 1000 km). National averages (Region 0) are used to construct the trajectories from wind speed and direction as well as their properties in terms of primary and secondary meteorological variables. During an OPS run, local meteorological conditions at the position of sources and receptors are determined based on their location and either on an interpolation of meteorological data from the three nearest regions or on a prescribed meteorological dataset. Interpolation between wind sectors and trajectory distances classes also takes place, depending on the source-receptor distance and direction.

When testing uncertainty due to meteorological conditions, variation of one single specific meteorological variable should be avoided since this may lead to inconsistencies. A simple example is that a change in air temperature implies a change in absolute humidity if relative humidity is

left unchanged and vice versa. To avoid such inconsistencies we chose to vary meteorological datasets as a whole.
We expect the main model uncertainty due to meteorological conditions to be related to the spatial distribution of the weather conditions, e.g., country average versus regional, or coastal versus inland regions. Because at present the same national averages of temperature, humidity and global radiation are applied to each region, and national averages of wind speed and direction are used to construct the trajectory paths and their base properties, differences between regional sets are comparatively small and are not expected to represent such spatial variation. Hence, we decided to proceed as follows.
For each region, one representative KNMI weather station was selected. Measurements from the selected station were then assumed to represent the weather conditions of the entire Netherlands. For example, the weather conditions observed in Maastricht also apply to Groningen, Vlissingen, *etcetera*. See Table A1, or Table A2 and 6 in the main text, for the list of weather stations used to define meteorological conditions for the sensitivity runs.
Measurements from the selected weather stations (Year 2015) were first converted by MPARKNMI, applying the meteorological conditions from the selected station for every other weather station in MPARKNMI as well. This results in six new meteorological data sets, each containing meteorological data for districts 0-6, as before. Whereas the same meteorological conditions are used across the country, small differences among the regions remain due to correction of wind speed for regional roughness length and the related spatial interpolation.
The resulting hourly timeseries from MPARKNMI are then processed by METPRO to obtain statistical properties of primary and secondary meteorological conditions for each region (0-6) within each dataset. This includes, for example, stability classification and determination of wind sectors and their frequency distribution, wind speed, temperature humidity, friction velocity and maximum mixing layer height. Within each dataset, differences among regions will even be smaller than in the default dataset. However, the differences among the new datasets as a whole will be comparatively large and represent differences across the country.
In addition to the six new datasets based on observations at six different, individual weather stations, the standard OPS set based on multiple stations was retained. Thus, we have seven meteorological sets in total. The standard (default) set for the year 2015 defines the reference situation. During the sensitivity runs, one of the seven sets was selected randomly. Of the selected set, always the meteorological conditions for district 0, i.e. the national averages, are prescribed in the present sensitivity analysis. Each set is formatted in the same way and can be used as input in OPS without further code adjustment.

2.8 Diurnal variation

This Section describes the adjustments made in OPS-LT to test the sensitivity to daily variations imposed on the emissions. Since the required methodology for the implementation has already been extensively described in the main text (Sections 3.1.5 and 3.2.2), we will limit ourselves here to only a small description of the purpose of the diurnal variation followed by the technical implementation in OPS-LT.

OPS-LT uses a yearly averaged emission strength as input. In reality however, most emissions are not constant in time. For instance, the traffic intensity is higher at the start and the end of the work day than during the night and around noon; NO_x emissions increase and decrease accordingly. In OPS, the yearly averaged emission strength is multiplied with so-called emission correction factors to account for such effects. The industrial stack and the motorway source have a diurnal variation correction prescribed in a diurnal variation file. The diurnal variation is a distribution of emissions over a single day for a particular source type. Twelve emission weights, each representing a two-hour block, are provided for each source type. Typically, the diurnal variation of emissions and hence of emission factors roughly resembles a 'sinusoid', for example for industrial activities with more emissions during the day and less during the night. The shape of this prescribed variation is scaled in the sensitivity analysis, while taking care that:

- emissions do not become negative
- the prescribed signal is not inverted, i.e. the 'sinusoid' is not flipped.

Note that the default 'sinusoid' differs among the various source types. This implies that the bounds on these variations will differ. This implies the following, which can also be deduced from Figure A49 which shows the default weighting factors, the maximum scaled weighting factors and the continuous emission (which equals the minimum scaled weighting factors):

- In the case of a specific perturbation factor, its relative effect on the diurnal variation change may depend on the source type. For instance, a scale index of 1 does not give the same perturbation for source type A versus source type B.
- This also introduces asymmetric effects in the sensitivity to the change in diurnal variation. For instance for the motorway source using the maximum scaled factor cannot deviate as much from the default as the minimum scaled factor, since the lowest default emission correction factor is already close to 0% (16%) and the amplitude cannot increase much further to prevent negative emissions.

The implementation of this scaling methodology in OPS-LT is given in Section 2.8.1. The default diurnal distribution factors used in OPS-LT are provided in Table A2. Code *dv* = 1 for average industrial activity and *dv* = 3 for average traffic intensity. For livestock farms, half the prescribed diurnal variation of average industrial activity is applied as emission correction factor in the code as an estimate to take into account the day/night rhythm of animals on the emissions.

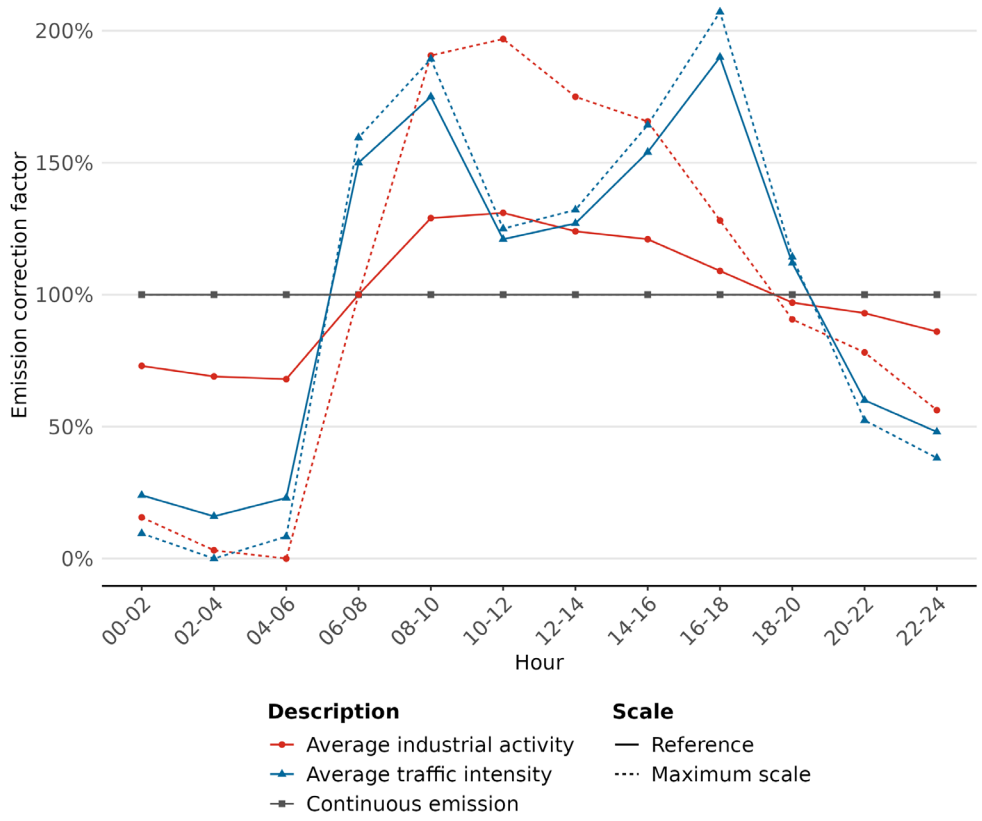

Figure A49 Overview of the default emission correction factors (solid lines) based on the two-hour blocks used in this study as well as the weighting factors using the maximum scale (scale_index = 1). Weighting factors using the minimum scale (scale_index = 0) all equal a uniform distribution (100%).

Table A2 Overview of the diurnal distribution with weighing factors in %.

hours→ code ↓	0-2	2-4	4-6	6-8	8-10	10-12	12-14	14-16	16-18	18-20	20-22	22-24	Description
0	100	100	100	100	100	100	100	100	100	100	100	100	Continuous emission
1	73	69	68	100	129	131	124	121	109	97	93	86	Average industrial activity
2	33	33	35	80	150	155	120	116	122	135	145	77	Average heating behaviour
3	24	16	23	150	175	121	127	154	190	112	60	48	Average traffic intensity
31	8	5	43	157	141	126	143	172	190	113	67	35	Light duty vehicles
32	12	17	82	158	161	163	167	175	131	73	41	22	Heavy duty vehicles
33	3	1	54	184	149	137	149	174	165	89	69	26	Buses (public transport)
7	33	33	35	80	150	155	120	116	122	135	145	77	Average heating behaviour (without seasonal correction)

For livestock farms an additional emission correction based on temperature is prescribed by default in OPS. Emissions increase for temperatures higher than 10 °C and decrease for temperatures below 10 °C. As such, this default correction also needs scaling for the sensitivity analysis. The implementation of this scaling method in OPS-LT is given in Section 2.8.2.

2.8.1 *Scaling the twelve 2-hour blocks*

For scaling the diurnal variation prescribed in the twelve 2-hourly blocks, a new module was programmed containing the subroutine scale_dverl, which scales the 'sinusoidal' shape using a *scale_index*. This new subroutine is called within subroutine ops_read_emis. First, the subroutine read_variation is called which reads the file with the prescribed twelve 2-hourly blocks and selects the line matching the prescribed diurnal variation code (i.e. depending on the source type and preset in the source file). This is followed by the scaling routine scale_dverl to perturb the diurnal variation strength.
Subroutine argument *scale_index* equals the scaling factor prescribed in varin_unc%diurn_scale_index prescribed in the ops_varin.txt file. A *scale_index* of 0.5 gives no change in the diurnal variation. A *scale_index* moving towards 0 pulls the distribution closer to a uniform distribution (less variation during the day). A *scale_index* moving towards 1 pulls the distribution's extremes (peaks and valleys) apart (more extreme variation during the day). This is displayed in Figure 5 in the main report.
The new module is provided below, preceded by the call to the new subroutine. Here *dverl* is the array containing the 12 diurnal distribution weights which is perturbed in this subroutine, and *dv* the number of diurnal variations.
The adjusted *dverl* is used in subroutine ops_init to determine the weighted average of diurnal emission variation, ecvl(nstab, ntraj, :ndv), for each stability/distance class for all the diurnal variation codes, *ndv* (local variable in ops_init and identical to *dv* in ops_read_emis and scale_dverl; nstab and ntraj give the number of stability and distance classes: 6 and 4 respectively). Variable *ecvl* is subsequently used in subroutine ops_stab_rek where it is multiplied with the source strength to perform the emission correction.

Call to the new subroutine scale_dverl within subroutine ops_read_emis:

```
call scale_dverl(dverl, varin_unc%diurn_scale_index, dv)
```

New module containing the new subroutine scale_dverl:

```
! The function `scale_dverl` is used to randomly scale a
diurnal distribution in m_ops_read_emis::ops_read_emis for
use in sensitivity analyses.
!
module m_diurnal_perturbation
   implicit none

contains
   subroutine scale_dverl(dverl, scale_index, dv)
      ! Scale a diurnal distribution without losing
        normalisation. A scaling index (or q-value) is
        transformed to a scaling factor using the quantile
        function of the scaling probability distribution (see
        comment below). A `scale_index` of 0.5 results in no
        change to the distribution.

      use m_commonconst_lt, only: NHRBLOCKS
```

```
integer, intent(in) :: dv  ! Number of diurnal
  distributions.
real, intent(inout) :: dverl(NHRBLOCKS, dv)  ! Array
  containing diurnal distribution weights.
real, intent(in) :: scale_index  ! Scale index between
  0 and 1, where 0 pulls the distribution closer to a
  uniform distribution and 1 pulls the distribution's
  extremes (peaks and valleys) apart. Can also be
  interpreted as a percentile of the scale's
  distribution.

integer :: i_distr  ! Distribution index.
real :: pivot  ! Mean distribution value around which
  weights are scaled.
real :: minimum_weight  ! Smallest weight in
  distribution.
real :: upper_bound  ! Upper scaling limit.
real :: scale_factor  ! Actual applied scaled.

do i_distr = 1,dv
   pivot = sum(dverl(:,i_distr)) / NHRBLOCKS
   minimum_weight = minval(dverl(:,i_distr))

   ! If the mean equals the minimum, scaling is not
      possible, so this distribution is skipped.
   if (pivot == minimum_weight) then
      cycle
   endif

   ! The upper scaling bound is de maximum value by
      which the diurnal distribution can be scaled
      without ending up with negative values. ! At this
      scale, the smallest weight becomes 0.
   upper_bound = pivot / (pivot - minimum_weight)

   ! The scale factor distribution is defined as a
      piecewise linear function where the probability
      falls of as the scale factor diverges from 1 and
      with the median fixed at 1.
   !
   ! p(x) ^       *
   !      |       :\
   !    1 +      .o \
   !      |    ." :  \
   !      |  ."   :   \
   !      |."     :    \
   !      +--------------> x
   !      0       1     φ

   ! Evaluate the quantile function of the distribution
      described above.
   if (scale_index < 0.5) then
      ! The outcome lies below the median (scaling
        down).
      scale_factor = sqrt(2 * scale_index)
```

```
            else
               ! The outcome lies above the median (scaling up).
               scale_factor = upper_bound - sqrt(2 * (1 -
               upper_bound) ** 2 * (1 - scale_index))
            endif

            ! Scaling the distribution around a 'pivot' is
               equivalent to a linear interpolation between the
               original distribution and its mean value. Scaling
               up is then equivalent to extrapolating.
            dverl(:,i_distr) = max(0., scale_factor *
             dverl(:,i_distr) + (1.0 - scale_factor) * pivot)
         enddo
      end subroutine
   end module
```

2.8.2 *Scaling the temperature correction*

The default emission correction performed for livestock farm emissions in OPS is based on a diurnal distribution prescribed in the twelve 2-hour blocks as well as an additional emission correction (*EC*) based on temperature via:

$$EC_{livestock\ farm} = \max\left(1 + 0.0294\left(T - T_{avg}\right), 0.2\right).$$

Here *T* is the outdoor temperature of the current stability class, wind direction and distance class and T_{avg} the long-term average outdoor temperature which is set at 10 °C. The temperature correction is not allowed to be lower than 0.2 °C, which is the case for temperatures below -17.2 °C. It is unlikely that this occurs in the meteorological classes in OPS for which multiple hours have been averaged in the meteorological pre-processor. Note that for yearly runs, this function implies that the emission total for a year with an average temperature other than 10 °C, changes. In general this function gives a higher emission correction factor for temperatures exceeding 10 °C and a lower emission correction when temperatures are below 10 °C.
This emission correction function also needs a perturbation, as otherwise only half the diurnal variation (being the 12 two-hour blocks) is perturbed for the livestock farm. It was decided to do this by perturbing the slope of the temperature correction function with a maximum variation of 20% in either direction (up to 20% steeper or 20% less steep). This is done be rescaling the temperature which is used in the temperature correction function.
The code adjustments are done in subroutine ops_stab_rek where the yearly average emission (source strength) is adjusted by multiplying it with the emission correction factor. The temperature correction is only performed when the prescribed diurnal variation code equals 4 (*dv* = 4, used for livestock farms only, note that this is not a line in the Table A2 on purpose because of the different emission correction method). The temperature correction *tcor* is subsequently multiplied with the original source strength to perform the emission correction.
The methodology is given in the code provided below and adjustments are implemented in ops_stab_rek. Similar as for the 12 two-hourly blocks, a scale index of 0.5 gives no change in the diurnal variation. A scale index of 1 increases the slope of the temperature correction by

20%, and a scale index of 0 decreases the slope by the same amount. As such, this either strengthens or reduces the temperature impact on the emission.

```
! The temperature correction `tcor` can be scaled for the
    purpose of sensitivity analyses. For its initial use,
    this scale is limited to 20% in either direction.
    Ideally, this limit should be imposed by the source that
    sets the value in the varin file. Currently this constant
    is used to perform inverse sampling to transform a random
    variate from U(0, 1) to U(0.8, 1.2).

real, parameter :: DIURNAL_SCALE_RANGE = 0.2  ! Maximum
    deviation of the slope of the temperature correction
    factor.
real, parameter :: Tavg = 10.0  ! Point around which to scale
    the temperature.

...

IF  (ibtg .EQ. 4) THEN
    ! Map (0, 1) to (1 - DIURNAL_SCALE_RANGE, 1 +
       DIURNAL_SCALE_RANGE), see the comment above the
       declaration of `DIURNAL_SCALE_RANGE`. A scale index of
       0.5 results in no change.
    temp_scale = (1.0 + 2.0 *
     varin%varin_unc%diurn_scale_index * DIURNAL_SCALE_RANGE
     - DIURNAL_SCALE_RANGE)
    ! Scale the temperature to allow performing sensitivity
       analyses.
    temp_C_rescaled = (temp_C - Tavg) * temp_scale + Tavg

    ! Emissions from animal housing; temperature correction
       for NH3 emissions from animal housing systems;
    ! Tavg = 10 C
    ! Temperature correction tcor = 1 + (T - Tavg)/f = 1 +
       T/f - 10/f = (1-10/f) + T/f = (f-10)/f + T/f = (T + f-
       10)/f;
    ! Here f = 34, corresponding with a factor 1/34 = 0.0294
    tcor=max((temp_C_rescaled + 24) / 34, 0.2)

...
```

References

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