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2019

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citation for published version (APA)

Schrader, F. (2019). *Challenges and perspectives in modelling biosphere-atmosphere exchange of ammonia*.

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SUMMARY

*Knowledge about the process being modelled starts fairly low,
then increases as understanding is obtained,
and tapers off to a high value at the end.*

— Harold Chestnut

6.1 KEY FINDINGS

In this thesis, I have explored the state-of-the-art of NH_3 biosphere-atmosphere exchange modelling. I have tried to highlight the key issues that need to be addressed in future developments, and to give recommendations on how to deal with them in the context of plot- and regional-scale inference of NH_3 fluxes from measured or modelled concentrations. In doing so, I have always assumed the perspective of a *practical model developer*, i.e., I have made an effort to find solutions that are ready to be implemented both in an ecosystem-integrated setting, such as micrometeorological monitoring sites, and in large-scale CTMs. This often comes at the cost of sacrificing a fully mechanistic description of all processes; however, suitable, science-based, steady-state approximations to complex and dynamic NH_3 exchange processes are something we desperately need to address the challenges that we are facing. That is, to identify key areas that are in need of immediate action to preserve our few remaining natural ecosystems; to create nation-wide and even global budgets of N_r emission, transport, and deposition; to back up our predictions with observational evidence and work hand in hand with the people who run representative monitoring efforts; and to clearly communicate the shortcomings of our current tools and the need for further research.

6.1.1 Deposition velocities

In Chapter 2, I have compiled a database of recent measurements and modelled estimates of the NH_3 deposition velocity. While the general idea of using a single deposition velocity per land-use class and over large periods of time is an inherently unsatisfying concept for process modellers, I have come to learn and experience first-hand that it is indeed still being widely used nowadays. This is especially the case in regulatory processes, where consultants and government agents alike need easy-to-understand and easy-to-use tools that leave little to no room for interpretation in their application. Using approaches that represent the scientific state-of-the-art, no matter how simple from a researcher's point of view, may already be so complex that they would indubitably lead to legal battles about operator bias. To quote the great mathematician John von Neumann: *With four parameters I can fit an elephant, and with five I can make him wiggle his trunk.* From the experience I have gathered in the past five years, it seems to me that this is also more often than not the case for modelling biosphere-atmosphere exchange of N_r compounds, where there is a plethora of different tools for the same purpose, which not all necessarily arrive at similar answers.

That being said, it is our responsibility as researchers to provide policy with the necessary tools to do their jobs, while being as objective as possible at this task. In Schrader and Brümmer (2014) we have made an attempt at doing so, by weighting results from a literature research based on an assessment of their validity and reliability. All the necessary tools to modify this weighting scheme, be they due to a disagreement with our weighting choices, or for specific applications, have been made publicly available. Key results found in this study reflect the overall expectation regarding the order of magnitude of deposition velocities for certain ecosystems, as well as their variability. Ecosystems with large receptor areas, such as coniferous or mixed forests, were found to show the highest deposition velocities, and agricultural areas, while having the lowest median deposition velocity, likely due to N saturation and thus large emission potentials, are overall very variable. Weighted average and median deposition velocities were 2.2 and 2.1 cm s^{-1} for coniferous forests, 1.5 and 1.2 cm s^{-1}

for mixed forests, 1.1 and 0.9 cm s⁻¹ for deciduous forests, 0.9 and 0.7 cm s⁻¹ for semi-natural sites, 0.7 and 0.8 cm s⁻¹ for urban sites, 0.7 and 0.6 cm s⁻¹ for water surfaces, and 1.0 and 0.4 cm s⁻¹ for agricultural sites, respectively.

Results of this study have since been used by numerous other researchers as a plausibility check for their experimental measurements and monitoring efforts (Adon et al., 2018; Häni et al., 2018; Hunova et al., 2016; Johnson et al., 2016; Thimonier et al., 2019; van der Graaf et al., 2018). Originally, this study emerged from the need for an update to the in Germany widely used tabulated deposition velocities by VDI (2006), and it was funded for that purpose (Schrader and Brümmer, 2013); however, it should be noted that at the time of writing this thesis, a task force responsible for updating the industry standard described in VDI (2006) is in the processes of exploring options for a simplified resistance model to be used in regulatory processes in the future instead.

6.1.2 *Non-stomatal exchange*

While Chapter 2 was the result of my first steps into the field of NH₃ research, Chapter 3 emerged from a deeper dive into the intricacies of modelling biosphere-atmosphere exchange of trace gases. Starting with the parameterisation of a two-layer canopy compensation point model (Nemitz et al., 2001) after Massad et al. (2010a), I noticed consistently low predicted non-stomatal deposition fluxes even at very humid measurement sites. A deeper investigation of the reasons for this revealed that this was very likely due to a too high minimum non-stomatal resistance prescribed in the parameterisation, especially at sites with large average acid-to-NH₃ ratios in the ambient air (Schrader et al., 2016). Observed average acid-to-NH₃ ratios ranged from 0.1 to 0.7 at the sites analysed in our study, leading to minimum non-stomatal resistance values at 100 % relative humidity between 45 to 315 s m⁻¹ before further corrections – considerably larger than values found in a meta-analysis by Massad et al. (2010a). This was further amplified by a too strong temperature response parameter in an empirical correction that essentially accounts for an increasing compensation point with rising temperatures, even

though this pathway is modelled unidirectionally in Massad et al. (2010a)

At the same time, I looked at alternatives for modelling exchange with non-stomatal surfaces, and I was intrigued by the ideas of Wichink Kruit et al. (2010). They have recognised the modeller's dilemma of having to find static approximations for processes that we know very well are dynamic in nature (Wentworth et al., 2016). While very sophisticated models exist that have been shown to successfully simulate phenomena such as early morning emission of NH_3 from leaf surfaces (Flechard et al., 1999; Sutton et al., 1998), they have not found widespread application in regional modelling due to limits in computational power for sub-grid processes and specific requirements about their input data. Wichink Kruit et al. (2010) attempted to solve these problems with the introduction and empirical parameterisation of a non-stomatal compensation point that does not require dynamically solving model equations at small time-steps. However, comparing this model to observational data I noticed a tendency to overestimate deposition under certain circumstances only. Their best parameterisation for the non-stomatal (and stomatal) emission potential included an instantaneous temperature response, accounting for seasonality in the emission potentials, which acts in the opposite direction of the temperature response that converts emission potentials into compensation point concentrations. This conversion is based on solubility and dissociation equilibria and can under some circumstances be considerably weaker than the seasonality function in the Wichink Kruit et al. (2010) model. In fact, as outlined in section 3.2.3, the equilibrium reaction temperature response may be completely countered by the seasonality response when temperatures increase. Consequently, under very warm conditions, when one would generally expect stronger volatilisation of NH_3 and thus a larger compensation point, it can actually fall to zero.

Comparing the two models against each other, as well as against micrometeorological measurement, led to an admittedly somewhat vague answer to the question which of the two parameterisations performs better in practice: *It depends*. The key lesson to learn from this observation is that there is, in fact, still a lot to do in terms of parameterising even these fairly simple

models, and that we should work towards a consensus on an appropriate framework to do so. The widely used unidirectional approach may work well in practice and be better validated on long-term budgets, but on finer temporal scales the approach of Wichink Kruit et al. (2010) allows for more realistic predicted fluxes and meaningful parameters, given that it will be updated with some modifications in the future, such as a delayed temperature response accounting for seasonality in the emission potential parameterisations.

6.1.3 *Stomatal exchange*

In Chapter 4 – part opinion piece, part case study – I describe first results on the way to a more mechanistic inter-compound coupling in biosphere-atmosphere exchange modelling. The desire to work towards such coupled models emerged from the goals of the junior research group I started my work in, called NITROSPHERE, in which we aimed to shine light on the interactions between N_r deposition and greenhouse gas exchange using both novel measurement techniques and biosphere-atmosphere exchange models. With regards to the modelling part, I found that there is relatively little overlap between those communities who model CO_2 exchange, and those who model NH_3 exchange. This came as a surprise, since they are fundamentally linked through a shared exchange pathway, as well as possible effects of acidity / alkalinity that are yet to be researched. Furthermore, many CTMs do model CO_2 exchange, albeit independent from other trace gases, and many, if not most sites at which NH_3 flux measurements are being made are already equipped with CO_2 flux measurement instrumentation.

In Schrader et al. (in preparation) we argue there are many potential benefits from modelling NH_3 and CO_2 exchange in a coupled manner, i.e., by linking them through the stomatal conductance via the well-known and well-understood model of Ball et al. (1987). This may be done in a number of different ways, e.g. either by directly inferring stomatal conductance from measured fluxes of CO_2 , or by modelling photosynthesis and ecosystem respiration, validating them on measured CO_2 net ecosystem exchange, and deriving a gap- and noise-free dataset

of stomatal conductance from modelled photosynthesis. This is useful for both the modelling and measurement community: By deriving stomatal conductance directly from CO₂ fluxes, models gain the ability of directly reacting to rising atmospheric CO₂ concentrations and the associated effects on stomatal opening (Ainsworth and Rogers, 2007). The monitoring community can benefit from having valid, data-driven and site-specific estimates of the stomatal conductance instead of generalised *one-size-fits-all* empirical parameterisations for few ecosystem types, that in addition often require potentially unavailable ancillary data and / or complex sub-models that are prone to errors in their implementation. Finally, we demonstrated the advantages for the experimentalist through an increased confidence in the validity of NH₃ flux partitioning via resistance models on a case study using eddy-covariance measurements.

6.1.4 Temporal resolution

The question of *mean* versus *effective* deposition velocities, i.e., the difference between the mean of the flux divided by the concentration and the mean flux divided by the mean concentration, was the topic of conversations with many different colleagues throughout my academic journey. It was always clear, both numerically and logically, that they cannot be the same, but at the same time, it was almost never discussed in the literature. In fact, as the reader may have noticed, I was not fully aware myself of the full scope of this issue at the time of publishing Schrader and Brümmer (2013, 2014) / Chapter 2 in this thesis, although it should be noted that in the vast majority of studies that went into our results only one of the two was mentioned anyway. I therefore worked on trying to quantify, and later correct for it, not only with focus on deposition velocities, but especially on bidirectional models with low-resolution concentration measurements as input data – which would need an *effective* modelled exchange velocity, but have usually been applied with *mean* exchange velocities in the literature available at the time of writing.

Eventually, I found that the magnitude of the error can be quantified precisely: It directly depends on the correlation of ambient concentrations and the NH₃ exchange velocity, which, unfortu-

nately, is unknown when working with low-resolution concentration measurements. However, as demonstrated in Chapter 5 / Schrader et al. (2018), site-specific correction functions can be calibrated on temporary parallel measurements, and they allow for a remarkable reduction of bias in long-term NH_3 deposition budgets (e.g., from 86 % to less than 1 % in one example). Since it appears as though the correlation is not constant throughout the year, these parallel measurements need to be taken either over a longer period, or at least temporarily during the different seasons. As of writing this thesis, it is unknown how long exactly this period needs to be, and whether or not the correlation is significantly different if calculated over shorter time periods than the *target resolution*, i.e., the sampling resolution of the low-resolution concentration measurements. From a sensitivity study outlined in Appendix C.2 it seems that there is relatively little change in the magnitude of the error (and therefore the correlation) between one and four weeks, which indicates that probably more than one data point per month may be used for the calibration of correction functions for a monthly target resolution.

6.2 UNCERTAINTIES

Rigorous assessments of both model and measurement related uncertainties in the estimation of NH_3 dry deposition fluxes are extremely rare in the recent literature, likely due to considerable difficulties in estimating fundamental uncertainty ranges necessary for a reliable error propagation. Generally speaking, uncertainty results from a combination of random errors (*noise*) and systematic errors (*bias*), although in practice these two types often cannot be differentiated easily, and the same source of error may appear as either of them depending on the researcher's point of view and the scales of interest.

Random error in measurements mainly results from limited precision of the applied measurement techniques, but can be quantified to a certain degree from careful characterisation of individual instruments. However, it should be noted that laboratory characterisations in a controlled environment are not always necessarily representative of an instrument's behaviour in long-term field campaigns. Secondary sources of random uncertainty

are found in both temporal and spatial scales: Multiple point-measurements (e.g., with chambers) are likely to exhibit larger variation than those from field-integrated micrometeorological approaches (although each individual point-measurement may be regarded as systematically biased). Similarly, due to intra- and inter-annual variation in meteorological and chemical conditions, individual years may not be representative of a long-term deposition climatology, and week-long measurement campaigns may not be representative of a seasonal average, whereas an average over multiple such campaigns is likely to be.

Systematic measurement uncertainty, on the other hand, is related to accuracy instead of precision, and much more difficult to quantify. Sources include, but are not limited to, instrument bias (e.g. due to calibration errors, or actual quantities of interest below the limit of detection), bias in data-processing related parameters (e.g., stability correction functions), errors in measurement setups, neglect of advection errors, and vertical flux divergence due to chemical reactions between the reference height and the NH_3 sink level. Likewise, temporal and spatial issues can introduce systematic error as well: Sampling bias originating from generally higher data availability during daytime may subsequently lead to biased gap-filling results and ignorance of night-time production and consumption processes, as is known to be the case for CO_2 (Moncrieff et al., 1996). Spatially related sources of uncertainty in measurements are an individual measurement site's representativeness for the ecosystem of interest, and heterogeneity in the flux footprint.

Both random and systematic errors in the measurement directly translate into errors in modelled fluxes, since models are typically calibrated to fit our measurements, and, if applied outside of CTMs, driven with measured input variables. As a consequence, for example noisy concentration measurements will inevitably result in noisy modelled fluxes. Regarding fixed, land-use specific parameter values, it is debatable whether or not data availability for all ecosystem types of interest is enough to bridge the gap from systematic representativeness errors to random variation in parameterisations. For instance, the acid-to- NH_3 ratio in the non-stomatal resistance parameterisation of Massad et al. (2010a) is based on four to five individual data

points for three of the four land-use classes they consider, and the uncertainty range around the relative humidity response parameter they provide leads to coefficients of variation of up to 89 % for short semi-natural ecosystems. Given the exponential nature of the parameterisation, this leads to enormous (random and very likely systematic) uncertainties in non-stomatal fluxes. However, at the time of publication, it was virtually impossible to estimate the systematic proportion of it, since the extensive literature review behind the Massad et al. (2010a) parameterisation covered the majority of available literature on measured NH_3 fluxes at the time, leaving little room for independent validation. Note that this is not meant as criticism of their approach, but rather a demonstration of a dilemma that necessarily emerges from limited data availability.

In comparison to measurements, modelled fluxes likely suffer from even larger systematic uncertainties: In this thesis, I demonstrated physical implausibilities in state-of-the-art parameterisations of the non-stomatal exchange pathway (Chapter 3), as well as errors arising from temporal averaging (Chapter 5). Note that the latter can directly be translated into the same type of systematic error we found in Schrader et al. (2018) when averaging in space, although the extent of that is yet to be investigated. With regard to regional modelling, both random uncertainty and systematic uncertainty in the representativeness of grid-cell averages naturally arise from implicit mixing errors and systematic errors in land-use and parameter maps, model resolution issues in orographically variable regions, and low granularity of emission maps due to data protection regulations. That being said, perhaps the most significant sources of systematic uncertainty are unresolved physical and chemical processes, such as within-canopy gas-particle interconversion, or leaf-level chemical processes like cuticular desorption. These are often impossible to parameterise outside of specialised field trials, or limited in their application due to computational constraints (e.g., Flechard et al., 1999, 2013; Sutton et al., 1998). Last but not least, there may still be significant *unknown unknowns* left in our understanding of NH_3 exchange processes, which would naturally fall in the same category.

It is evident from this discussion how difficult it is to provide general estimates of uncertainty that are representative for the combined effects of all different types and sources, and that can be applied on different scales. All uncertainty estimates in this field need to be linked to a clearly defined reference frame in space, time, and application. Micrometeorological measurements at a given site can have low systematic uncertainty if the measurements are aimed at testing hypotheses about a specific site only, but very little certainty for general statements about an ecosystem type as a whole. To my knowledge, there has not been an attempt at quantifying uncertainties of state-of-the-art NH_3 biosphere-atmosphere exchange models at different scales in the recent literature. Erisman (1993), in one of the few simultaneous assessments of model- and measurement related uncertainties, estimated the total random uncertainty in the yearly average of modelled NH_3 dry deposition to be 130% and the systematic uncertainty to be 58% on a 5 by 5 km grid over the Netherlands. For the whole country, his random uncertainty estimates reduced to 17% and systematic uncertainty to 45%. Note that these numbers are based on the (likely not always valid) assumption of uncorrelated errors. In the *worst case*, perfectly correlated errors, systematic errors for total (wet and dry) reduced nitrogen deposition increase up to 89–98%.

Strategies for a new comprehensive uncertainty analysis could be based on reanalysing instrument comparison field trials to obtain good estimates of measurement-related errors, following the example of Nemitz et al. (2009). Random uncertainty in models might be estimated by simultaneous parameter optimisation on existing flux datasets, potentially using Bayesian methods that yield credibility intervals and parameter distributions instead of single *best fit* values, or at best, approximate confidence intervals as obtained from traditional frequentist statistics. Forward error propagation from noisy input data and parameters whose distribution is known can nowadays easily be done with simple *brute force* Monte Carlo approaches for one-dimensional applications, thanks to abundant computing power. Systematic uncertainty should be analysed using model ensembles similar to the works of Flechard et al. (2011), but with higher temporal-resolution of the input data, and compared to direct flux measurements

obtained with micrometeorological methods. For sites where the necessary data are available, comparing mechanistically accurate models (or model-parts) (e.g., Flechard et al., 1999; Massad et al., 2010b) with the simpler compromises used in CTMs might be instructive with regard to their physical plausibility, and a first step towards developing approximate, easy-to-parameterise meta-models for missing processes.

In the work at hand I have not done any uncertainty calculations that are remotely as comprehensive as, e.g., shown in Erisman (1993) myself. However, the numbers Erisman (1993) arrives at are generally in agreement with my own expectations and experiences in field-scale application of NH_3 biosphere-atmosphere exchange models more than two decades later. Given that running existing state-of-the-art models in parallel with the same input data often yields a surprisingly large span of predicted fluxes, sometimes even in opposite directions for individual pathways, I generally assume that at the field scale, absolute total uncertainty in annual estimates of dry deposition is at the very least on the order of $1 \text{ kg ha}^{-1} \text{ yr}^{-1}$, or in relative terms, usually a significant two-digit percentage. Note that it is generally accepted that micrometeorological measurements are usually only accurate to around 10–20% anyway, due to natural variability in terrain, turbulence, and instrumentation (Moncrieff et al., 1996).

In a recent long-term measurement campaign at a remote site in the Bavarian Forest National Park, we compared eddy-covariance measurements of total N_r biosphere-atmosphere exchange (Brümmer et al., in press; Zöll et al., under review), the majority of which consisted of NH_3 , with modelled estimates using different variants of the dry deposition module DEPAC (van Zanten et al., 2010), based on the parameterisation of a two-layer canopy compensation point model of Wichink Kruit et al. (2010). Depending on the specific scenario (inside the CTM LOTOS-EUROS and a measurement-driven 1D variant; with and without land-use corrected for site-specific conditions), modelled cumulative total N_r fluxes before gap-filling were within 20–70% agreement with the measurements, although almost all variants showed a tendency to overestimate deposition (Brümmer et al., in press).

Despite not directly quantifying them, this thesis contributes to the overall reduction of uncertainty by isolating sources of systematic error in the non-stomatal pathway (Chapter 3), promoting the use of mechanistic, direct, and data-driven methods (Chapter 4), and providing a correction method for systematic errors arising from temporal averaging (Chapter 5).

6.3 IMPLICATIONS

In this thesis, I probably ask more new questions than I give answers to existing ones. Nevertheless, if we look at the overarching aim of developing practical tools for the assessment of N_r deposition, the better understanding of state-of-the-art model parameterisations and the errors that are associated with them can directly be put to good use. Most, if not all of the shortcomings that have been discussed may be revised with reasonable effort, possibly even without additional measurements, as further outlined in the final section of this summary. Others may directly be addressed on the local scale.

There is a clear need for large-scale N_r deposition monitoring, which, in many countries is not met. In Germany, apart from throughfall measurements in forests, only very few experimental measurement sites exist, much less any kind of nation-wide deposition monitoring efforts. However, the results presented in this thesis may be helpful to fill this gap. For instance, continent-wide research infrastructures like ICOS (Franz et al., 2018) in Europe or NEON (SanClements et al., 2014) in the US, which routinely measure turbulent fluxes of CO_2 using the eddy-covariance technique, may be used as a modular platform for low-cost NH_3 deposition monitoring. Their existing flux towers may be equipped with low-cost, low-effort concentration measurement devices like passive samplers, and complemented by individually calibrated biosphere-atmosphere exchange models. In such a monitoring network, the stomatal pathway would be parameterised through CO_2 flux-derived stomatal conductance, as well as estimates of the stomatal emission potential based on historical modelled or measured N inputs (Massad et al., 2010a), or directly on long-term average ambient NH_3 concentrations (Wichink Kruit et al., 2010). Site-specific parameterisations of the non-stomatal

resistance (in a unidirectional framework) or its emission potential (following the works of Wichink Kruit et al., 2010) may be based on a one-time calibration on nighttime flux measurements using a roving system. Finally, the same system would be used to calibrate correction functions for the lowered temporal resolution of passive samplers (which actually only needs concentration measurements instead of fluxes) and for site-specific model validation. The initial effort for setting up such a hybrid model-measurement-network would be largely outweighed by the benefits of an already existing infrastructure, finished site exploration, and low additional long-term cost of operation.

6.4 RECOMMENDATIONS

Finally, I would like to conclude this thesis with a few recommendations for future research, both related to NH_3 modelling in general, and to the specific questions that emerge from my research.

CONSENSUS ON AN OPTIMAL FRAMEWORK Finding the optimal balance between realism and applicability is probably the biggest challenge for modellers in every scientific discipline. As Flechard et al. (2013) noted, the level of complexity needs to be tailored to the specific application, but overall the two-layer canopy compensation point model after Nemitz et al. (2001) has been recognised as a good compromise between realism and accuracy on different scales. However, for site-level applications the unidirectional treatment of the non-stomatal pathway remains somewhat unsatisfying. Personally, I am convinced that a hybrid model, i.e. a steady-state approximation of cuticular desorption following the example of Wichink Kruit et al. (2010), should be the way to go in the future, as there is no clear justification for treating both stomatal and ground-layer fluxes bidirectionally, but fluxes from other non-stomatal surfaces (e.g., wet leaves and stems) not.

RECALIBRATION OF EXISTING MODELS As illustrated in Chapter 3, both the parameterisations of a two-layer model after Massad et al. (2010a) and Wichink Kruit et al. (2010) suffer from

some shortcomings in specific details of their implementation. I am convinced that feasible solutions to these can be found based on existing data and theoretical considerations. The temperature response parameter in Massad et al. (2010a) may be set with an effective one derived from equilibrium constants (cf. Section 3.3.1). The non-stomatal resistance can easily reach values of more than 1000 s m^{-1} at relative humidities as high as 90 % at sites with low acid-to- NH_3 ratios, and it should be tested whether more suitable minimum values can be obtained through a recalibration on high-frequency flux measurements instead of campaign averages. Regarding the quasi-bidirectional approach of Wichink Kruit et al. (2010), an effort should be made to parameterise it on direct flux measurements from a larger variety of ecosystems. The issue of the seasonality countering thermodynamic equilibria needs to be addressed, as it affects not only the non-stomatal pathway discussed in Schrader et al. (2016), but also the stomatal pathway through a similar functional relationship. I propose researching options for smoother response functions in the seasonality parameterisation, e.g. by basing them on average instead of instantaneous measurements of the air temperature, or alternative variables with a distinct annual course with less intensive short-term fluctuations.

DISENTANGLING NON-STOMATAL EXCHANGE Presently, non-stomatal exchange pathways are often treated in an unsatisfying manner. For example, the parameterisation of Massad et al. (2010a) is run in a one-layer configuration for unmanaged ecosystems and managed ecosystems outside of management events, but in a two-layer configuration e.g. after fertilisation. While generally the motivation to do so is reasonable, as the ground-layer and other non-stomatal sources can only be separated in flux measurements when the majority of the signal can safely be attributed to one source, it leads to an awkward shift in the meaning of certain parameters. When there is no clear ground-layer signal, its resistance is set to infinity and exchange with it is lumped into the non-stomatal pathway, which is otherwise often interpreted as representing leaf-surfaces only. This may lead to misunderstandings and possibly bias in future parameterisations based on meta-analyses and should be addressed by carefully

unraveling the two pathways, e.g. through the help of chamber measurements.

MECHANISTIC TREATMENT OF CO-DEPOSITION Known parameterisations of co-deposition processes are mainly based on empirical relations (e.g., Massad et al., 2010a; Wichink Kruit et al., 2017). Future research should explore more mechanistic solutions, including their applicability to other trace gases than SO₂. Practical options worth exploring would be to derive effective co-deposition functions from acidity / alkalinity calculations, or attempting to build a steady-state meta-model based on the dynamic leaf-surface chemistry simulations of Flechard et al. (1999).

COMPARISON OF MEASUREMENT TECHNIQUES While the aerodynamic gradient technique has been the de-facto standard for NH₃ measurements in the past, the recent literature has seen an increased use of novel methods primarily using different optical spectroscopy systems (Ferrara et al., 2012; Hansen et al., 2015; Volten et al., 2012; von Bobruzki et al., 2010; Whitehead et al., 2008; Zöll et al., 2016, and others). These should carefully be evaluated against each other and against gradient measurements in order to assess the uncertainties related with the techniques, and NH₃ flux measurements in general.

HARMONISED DATABASE OF FLUX MEASUREMENTS During my work on this thesis, I found that getting access to existing observational data is not a trivial task and can be extremely tedious when planning multi-site comparisons, and these data naturally come in an enormous variety of different formats and quality levels. There should be a global effort to build a harmonised database of not only NH₃, but all N_r exchange measurements, following the example of FLUXNET for eddy-covariance data. A better availability and accessibility of measured fluxes and other necessary variables would easily have one of the biggest impacts on boosting model development in the future.

MODEL VALIDATION A natural consequence of data scarcity is that models are often calibrated on virtually all available data,

which leaves little to no room for independent model validation. Model developers should try to address this, e.g., by running cross-validation analyses and exploring other options to obtain validation data that were not used for the respective parameterisations.

As this list is by far not exhaustive, and only discusses the most pressing issues from the point of view of someone mainly working with very simple one- or two-layer models, it is evident that the field of NH_3 biosphere-atmosphere exchange modelling still has a lot of room to grow. We should embrace these challenges and do our best to work towards building good and practical models, as in light of data scarcity they are necessary tools to identify key areas at which we need to focus our environmental protection efforts.

I hope that my humble contribution to this exciting field of research can be of value for the scientific community.