Quantification of nitrogen deposition and its uncertainty with respect to critical load exceedances

Bleeker, A.

2018

document version
Publisher's PDF, also known as Version of record

Summary

This thesis deals with the quantification of nitrogen deposition and its uncertainty, with a focus on the relevance of these uncertainties for critical load exceedances. Current policies on protecting Natura2000 areas, in the Netherlands and other countries in Europe, build on these exceedances. Uncertainties in nitrogen deposition have a direct effect on the critical load exceedances and, thus, eventually on the evaluation of the effectiveness of measures to abate nitrogen emissions to protect nature areas. The overall question to be answered in this thesis is if the uncertainties in the deposition estimates are sufficiently known to merit their application for policy to combat nitrogen deposition. Knowledge about these uncertainties can then be used to determine if abatement measures should be taken based on the estimates, next to arguments like e.g. precautionary principles, effects, public concerns, etc.

The quantification of the deposition and critical load exceedance is the main topic of this thesis with a focus on uncertainty in the deposition. The uncertainty in critical loads is not addressed here. How can we estimate nitrogen deposition and what is uncertainty in deposition estimates? Do we, taking everything into account, know enough about the uncertainties in nitrogen deposition? Do these uncertainties justify extensive emission reduction measures based on (in this case) exceedances of critical loads. In the following paragraphs these questions will be answered through addressing the research questions in this thesis.

How can we determine nitrogen deposition levels and their trends in time?
Nitrogen deposition results from the emission, transport and atmosphere-biosphere exchange of reduced nitrogen components mainly stemming from agricultural sources and oxidised nitrogen stemming from burning of fossil fuels. Nitrogen can be deposited directly in the form of gasses and aerosols to the earth's surface as well as through precipitation as wet deposition. Nitrogen deposition can be determined using measurements, model calculations and the combination of the two. A detailed overview of different methods used for this is introduced in Chapters 2-5 of this thesis. For wet deposition rain samplers are used that measure both the amount and the chemical composition of precipitation. It is shown here that despite the long-term use of such samplers, the differences between the sampling methods can be up to 27% for ammonium and 25% for nitrate on the same plot. Dry deposition is much more difficult to measure and there is no single instrument that measures the dry deposition of different N-components directly. Therefore, different methods are used for different components.

Models are essential to quantify the deposition and to relate emissions to depo-

sition. Models are helpful to understand and explain processes, but also to determine the effect of scenarios and policy options. Models can be based on measurements, upscaling them or translating measurement derived parametrisations into deposition models. Generally, measurements are used for validation of theoretical or empirical models. Models differ in their application; there are small-scale and large-scale models (see Chapter 4).

Trends in ambient concentration and deposition are preferably derived from measurements. Chapter 5 discusses the trends in measurements and the effects of policies in Europe. The trend assessment for reduced nitrogen showed that for the interpretation of long-term trends an improved mechanistic understanding and modelling is required. This especially holds for a better generalisation on the bi-directional controls on NH_3 exchange, inclusion of chemical interactions when dealing with dry deposition and the advancement of the regional temporal modelling of NH_3 emissions in relation to environmental conditions.

The aim of the methods is to determine nitrogen deposition relevant for critical load exceedances and, therefore, be representative for an ecosystem scale on an annual basis. Both measurements and modelling may prove to be sub-optimal, because of several reasons related to e.g. logistics, financial possibilities or data availability. While the final setup of a measurements network is almost always a compromise because of these issues, the effect of them on the modelled or measured deposition is not always clear. Prior to establishing a measurement network, the effect on the overall uncertainty needs to be investigated. For the interpretation of measured trends, it is necessary to relate the observed changes to the right processes. E.g. changes in atmospheric chemistry alone were shown to decrease wet deposition of ammonia by 10% for the Dutch situation. A good understanding of the chemical interactions is therefore required to understand the potential 'masking effects' of changing emissions of potential reaction agents.

For both methods, measurements and modelling, uncertainties are potentially introduced through omission of relevant processes. There are consistent trends in measured concentrations and deposition, using similar measurement methods, that show changes in both nitrogen concentrations and deposition. Intercomparisons, both in the laboratory and in the field, have been undertaken, to validate these measurements as discussed in Chapter 3.

What is the uncertainty in nitrogen deposition on different spatial scales and what are the factors influencing it?

Both the levels of and trends in deposition of nitrogen are subject to a broad range of uncertainties. Systematic and random errors occur in trying to determine the deposition by either measurements or modelling (or a combination).

In Chapter 8 it is shown that atmospheric N deposition estimates generally have a significantly high level of uncertainty, at landscape scale commonly as large as a factor of ~2.

Modelling

The main uncertainties in modelling the deposition are the limited understanding of bi-directional fluxes of N_r and the still largely unknown sources and forms of organic N deposition. The uncertainties in the modelled values are the result of errors that can be random and systematic. While systematic errors are mostly the result of insufficient knowledge about the processes involved in deposition, or due to neglect of processes or variables, random errors are the result of variability in measured quantities and/or parameters, as also discussed in Chapter 5. These can be caused by e.g. measuring errors and parameterisations. Overall, knowledge of the reliability of models for deposition estimates is limited. This is mainly due to the fact that there are only few observations of many key deposition parameters, which hampers the assessment of the total N_r deposition estimates, especially its dry deposition component. More specifically there is a lack of measurements on some of the key compounds such as gaseous nitric acid, HNO_3, coarse-nitrate and ammonia, NH_3, without which a comprehensive model evaluation is hardly possible. Other sources for the uncertainties in the modelled deposition relate to: partitioning between gaseous HNO_3 and either fine of coarse nitrate aerosol, estimates of NH_3 emissions (+ relation with meteorological factors, as well as agricultural practices and coupled with an understanding of biosphere-atmosphere exchange), dry deposition of particles, not including all nitrogen species (e.g. organic N), omission of bi-directional NH_3 transport, sub-grid fluxes of NH_x compounds, but also the effects of e.g. topography on wet deposition.

At different spatial scales, different aspects play a role. Going from a low to high resolution, processes included in atmospheric transport and deposition models will need to describe the local scale in more detail. This obviously puts more pressure on data availability and their quality and may eventually hamper an optimal setup. Representativeness of the modelled results may then prove to be a real problem, potentially introducing large uncertainties. For models, in general, the uncertainty increases with an increasing resolution. At a low resolution, i.e. averaging over a larger region, the chance that the actual deposition values are covered is relatively large when there are only random biases. In the case of systematic biases the uncertainty is scale independent. At a higher resolution, the representativeness issue becomes more important and introduces a higher uncertainty in the deposition estimates when using high resolution data that do not accurately represent a local situation. Acquiring high-quality data (at a high resolution), that are needed to prevent an increase of the uncertainty in the modelling outcome at high resolutions, may be hampered by financial and/or privacy restrictions.

Measurements
The representativeness issue also holds for nitrogen deposition measurements. A potentially high density of measurement locations is needed to adequately capture steep gradients in air concentration/deposition of nitrogen when using measurements alone. Financial restrictions often limit the effort, possibly resulting in a less dense monitoring network. In that case, it needs to be clear beforehand what the effect of this restriction is on the capacity of the network to adequately capture the spatial and temporal distribution of concentration and/or deposition. New methods, such as satellite observations might overcome this problem, but further development is needed.

How does the uncertainty in the nitrogen deposition affect the relationship with effects like biodiversity and exceedance of critical loads?
Chapter 6 shows that about 11% of the area of the world's protected areas under the Convention on Biological Diversity receives more than 10 $kgN.ha^{-1}.y^{-1}$, which indicates a situation where negative effects on the ecosystems are likely to occur. For the year 2030 the situation is not expected to change.

In principle, every aspect of the cause-effect relation in such an assessment has its own (sometimes considerable) uncertainty, combining them is not a straightforward process. The way in which the errors propagate along the emission/deposition/critical load/exceedance chain may give room for error compensation within the chain. Because of this, it is not easy to tell what the overall uncertainty of such a system will be. An example of this is the mixing of oxidised nitrogen during its longer travel distances, compared to reduced nitrogen. While individual sources, based on an activity and an emission factor, have their own uncertainties, there is an increasing potential for error compensation due to the long travel distance in the air (compared to reduced nitrogen), which reduces the uncertainties in total deposition estimates. Another example is that due to the error compensation the estimates of the deposition of total nitrogen, are more certain than those of the individual nitrogen compounds.

The uncertainty can be determined for the situation where the items in the chain are known through e.g. comparison with available measurements. However, the influence of systematic biases, such as not taken a nitrogen component into account, when determining an empirical critical load for a particular species, a problem may arise further along the chain. For determining the empirical critical load the total (known) input of nitrogen is related to an observed effect of the species. However, when there is also a contribution of an unknown species (e.g. organic N), relating the effect to the known species may eventually result in policy makers taking more drastic actions focussing on reducing a critical load exceedance based on the available information about the known compounds. For example, when ammonia emission reduction measures don't show the expected result in terms of reducing exceedance of critical loads, an easy solution would be to reduce ammonia emission even more. However, in the meantime, the unknown nitrogen compound might influence the visible effects on biodiversity and thus erroneously known compounds like ammonia could be blamed for this.

Is there evidence for the hypothesis of this thesis, or can it be falsified?

For protecting Dutch nature areas from nitrogen deposition, a drastic reduction of the ammonia emission of more than 70% compared to the 1980 situation is required, according to calculations presented in Chapter 7. Possibilities to decrease the emissions and their effects were investigated, using information on calculated depositions, critical loads and their exceedances. Such an assessment combines the different steps in the cause-effect relation, as is also shown in Chapter 6, with uncertainties influencing the individual aspects of this relation.

For this thesis, the following hypothesis was postulated: *Nitrogen deposition contributes to negative societal effects on environment, biodiversity, climate and human health. Policy development to reduce nitrogen deposition and its effects requires reliable estimates at different spatial and temporal scales based on measurements and models. Such estimates are necessary to determine cause-effect relationships, such as the estimates of critical load exceedances. Policy to reduce exceedances is associated with costs to sectors and society. Therefore, it is essential to know the uncertainty in deposition. Nitrogen deposition is, however, still poorly understood and a full quantification of uncertainties is too limited to merit application of estimates for policy. Unless the gap between the estimates and policy goals is large, the estimates cannot be used, both for describing deposition trends and for absolute estimates at specific locations, without identification of uncertainties.*

As shown in this thesis, there are methods available to quantify deposition on different scales. A combination of measurements and models is needed to provide the best estimate. However, spatial and temporal aggregated nitrogen deposition is still uncertain due to the nature of the sources, transport and landscape dependent deposition. Issues remain about both measured and modelled deposition. The following issues play a major role, as shown in Chapter 8, resulting in systematic or random bias:

- Lack of detailed and accurate activity data (random),
- Source specific emission factors (systematic),
- Missing components (systematic),

- Omission of processes (systematic),
- Lack of measurements (random).

Modelling studies, based on models that were validated using state of knowledge information of that time, have been widely used for e.g. exceedances of critical loads and policy evaluations. While current models are able to evaluate the effect of measures, which was already shown for the spatial emission optimisation studies, the uncertainty is, however, not fully quantified. Difficulty of this lies in the nature of the problem: large spatial and temporal variation of emissions and deposition, but also by a seemingly simple atmospheric chemistry that is, however, difficult to capture in models due to the presence of different chemical equilibria in the atmosphere.

A good example of the effects of the uncertainties is given by the Dutch ammonia gap. This systematic bias is caused by missing sea emission and some other smaller sources missing, but also incomplete deposition parameters for e.g. fertilised grassland. Another example is the mis-interpretation of the decrease in SO_2 and the effect of long-range transport and their focus on ammonia concentration, rather than on the total mass balance. Random bias is introduced in situations where e.g. reported emission locations do not correspond to the actual situation and thus the contribution of the deposition from these sources to critical load exceedances is, therefore, over- or underestimated.

Although some attempts have been made to quantify uncertainties in the estimates of nitrogen concentrations and deposition, a full identification of the uncertainties is lacking because the listed uncertainties have not all been quantified. When such an identification is not complete, a true answer about nitrogen deposition loads is not possible.

Can policies still be implemented in such a situation? Is it acceptable that extensive emission reduction measures are implemented, based on (in this case) exceedances of critical loads? While the overall chain from emission to deposition and exceedance of critical loads has a long list of uncertainties associated with it, from a scientific point of view these extensive measures are not fully justified, since there are simply too many unknowns with large uncertainties.

However, from a precautionary perspective it is not that simple. If no action is taken until all the uncertainties are identified, then a potentially endangered situation will only get worse. In the field there are observations of the effects of e.g. nitrogen on biodiversity and groundwater quality, showing that the gap between current concentration and/or deposition estimates and the policy goals (e.g. critical loads) is still large. For this reason, although uncertainties remain, it is still justified to take mitigation action, so long as ongoing efforts are taken to continue to reduce uncertainties and to review progress in the results of the mitigation measures.

For this it is recommended to (re-)establish a monitoring programme focussing on key components of the source-effect chain described here. Such a program needs to address the knowledge acquisition on processes involved in the chain and their interactions, with the aim to reduce the uncertainties in measuring and modelling of both nitrogen deposition and critical loads. Another important goal of the programme is its ability to specifically monitor the effects of mitigation measures related to nitrogen deposition. Relevant scientific developments for such a programme start with a clear definition of nitrogen (deposition) to be used throughout the whole source-effect chain. Methods for measuring and modelling this nitrogen then need to be lined up with this definition, which can require models to include new processes for which parameterisations and emissions have to be obtained. This also has consequences for the measurements: what measurement method and setup is needed to measure nitrogen deposition? Recent developments with respect to satellite observations can help solve challenges with respect to spatial and temporal variables related to nitrogen emission and deposition, but more work is needed on developing parameters related to the nitrogen pathway. Future research needs to focus on these aspects to reduce the uncertainties in the chain. Hopefully, this thesis contributes to an improved understanding of nitrogen deposition and reducing its uncertainty.