

## Chapter 23

# Detecting Change in Atmospheric Ammonia Following Emission Changes

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### 23.1 Summary

The Working Group discussed the progress on the state of knowledge on deriving trends from measurements and their use to verify abatement measures or other causes for decrease in emissions of ammonia to the atmosphere. The conclusions from the 2000 Berne meeting (Menzi and Achermann 2001), the background review (Bleeker et al. 2009) and presentations during the session (Horvath et al. 2009; Tang et al. 2009; Webb et al. 2009), as well as the discussions served as input for the conclusions of this report.

We have seen some clear advancement in closing the gap between the observed and expected values for reduced nitrogen, where we do get a better understanding of the reasons behind it. The long-term measurements that are available follow the emission trend. Current measurements make it possible to evaluate policy progress on ammonia emission abatement. Especially in those countries where there were big (>25%) changes in emissions, such as in the Netherlands and Denmark the trend is followed quite closely, especially when meteorology is well taken into account. In order countries, such as the UK, the trend was much smaller, but there was no gap between measurements and model estimates. In the Netherlands there still is an ammonia gap: a significant (30%) difference between emissions based ammonia concentrations and measurements. The trend is the same. The difference might be due to either an underestimation of the emission or an overestimation of the dry deposition. It is recommended to further explore this gap, especially by investigating the high temporal resolution measurements, improving the emission/deposition modeling, by having a model intercomparison with countries that use models that do not show a gap and finally by doing a thorough uncertainty analysis.

On the European scale it is difficult to follow the emission changes, both because of lack of measurements, especially in the Eastern part of Europe and because of the confounding factor of the SO<sub>2</sub> emission reductions, affecting the ammonium

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concentrations in aerosol and in rain water. It is recommended to fully implement the EMEP monitoring strategy and to improve the models in order to quantify the influence of a changing chemical climate.

The EMEP monitoring strategy can be a good starting point for development of a strategy that is focused on the right questions. Therefore first it is necessary to evaluate policies and the indicators derived from them that need to be assessed (time and space). Using existing models a pre-modeling study should be done to select the monitoring sites that eventually will give you the answer to the basic (policy) question using improved models and assessment tools. The best and economic feasible instrumentation should be selected with an extensive QA/QC program to make the measurements comparable. After implementation, especially for trend evaluation, the monitors used should not be changed.

## 23.2 Introduction

The Working Group addressed the issues involved in making the link between estimated national  $\text{NH}_3$  emissions and measurements of  $\text{NH}_x$  concentrations. Monitoring  $\text{NH}_3$  emissions directly is not possible and therefore there is a need to quantify the effectiveness of  $\text{NH}_3$  emission abatement in a more independent way by using measurements of  $\text{NH}_x$ .

The Working Group addressed the following objectives:

- To quantify the extent to which estimated regional changes in ammonia emissions have been reflected in measurements of ammonia and ammonium in the atmosphere
- To distinguish cases where the estimated changes in ammonia emission are due to altered sectoral activity or the implementation of abatement policies and thereby assess the extent to which atmospheric measurements verify the effectiveness of ammonia abatement policies
- To make recommendations for future air monitoring and systems for assessing the national implementation of ammonia abatement policies

During the discussions within the working group the following items were addressed, which will be described in more detail in the following sections:

- Update the current scientific understanding based on new datasets and assessments
- Is there still an 'ammonia gap' in the Netherlands?
- Does such a gap exist in other countries of Europe?
- Are we confident about the effectiveness of ammonia mitigation policies?
- How can we best address the relationships between emission and deposition using atmospheric modelling and improved monitoring activities?

Update the current scientific understanding based on new datasets and assessments

The update of the scientific understanding is presented in the Background Document prepared for this workshop. The document builds on the Berne Background Document (Sutton et al. 2001), which was used to facilitate the discussion about following trends by means of measurement data at the UNECE Ammonia Expert Group meeting in Berne (Switzerland) in 2000. It is now several years since the Berne Workshop and major new datasets on European  $\text{NH}_3$  and  $\text{NH}_4^+$  monitoring and their relationship to estimated  $\text{NH}_3$  emissions have become available for the following countries: UK, Germany, Hungary, Switzerland, Denmark, The Netherlands, North Carolina (USA), Slovak Republic, Norway and Croatia. Bases on these datasets the findings of the previous workshop were evaluated, updating our current scientific understanding about the different issues that were addressed in the previous document. In particular, input is given to questions like: is there still an 'Ammonia Gap' in the Netherlands, does such a gap exist in other countries, can we be confident of the effectiveness of ammonia mitigation policies and how can we best address the relationships between emission and deposition, using atmospheric modelling and improved monitoring activities?

Next to country-specific case studies, also an overview of the European situation is given with respect to the link between emissions of  $\text{NH}_3$  and the modelled and measured concentration/deposition of reduced nitrogen. Changes in chemistry, especially due to decreasing  $\text{SO}_2$  emission, are evaluated in terms of its effect on the levels of reduced nitrogen over European both temporal and spatial, providing a better understanding of the observed levels in  $\text{NH}_3$  and  $\text{NH}_4^+$  over Europe.

In general, the conclusions from the Berne Background document (Sutton et al. 2001) are (to a large extent) still valid. Any further elaboration on the update of our current scientific understanding of the problems addressed in the background document and the conclusions made is not given here, but will be worked out in more detail when addressing the other items in the following sections. However, looking at the differences between Berne Background document and the current review (Bleeker et al. 2009), we can distinguish some major advances in our understanding of the different issues we mentioned before:

- We have seen some clear advancement in closing the gap between the observed and expected values for reduced nitrogen, where we do get a better understanding of the reasons behind it. This especially builds on case studies from the UK (study on Foot and Mouth Disease) and the longer term datasets that became available for the Netherlands and Denmark.
- The long-term measurements for reduced nitrogen follow the emission trend. The extended datasets presented in the background document made it clear that following emission trends by means of measurements can only adequately be done when long-term measurements are available (longer than, e.g. 10 years).
- Current measurements make it possible to evaluate policy progress on ammonia emission abatement. This is especially true for the situation in Denmark and the Netherlands, where the  $\text{NH}_3$  ammonia reduction is followed by the monitoring results from the national monitoring networks. However, this is only possible since both these networks were designed to follow the expected changes in

emissions, based on extensive pre-studies used for developing the measuring networks. The UK also performed such a pre-study for developing their national monitoring network. Clear trends from this network could not be found there, but this is mainly due to only small changes in emission levels. Another case in the UK, where it was possible to detect changing emissions in measured concentrations of  $\text{NH}_3$ , was related to a study following an event of Foot and Mouth Disease in different regions in the UK. Also here a modelling pre-study was used for designing the layout of the measurement network, in order to detect effectively the expected emission changes within the different regions.

- In recent years the instrumentation (i.e. models, monitoring equipment) to evaluate the link between ammonia emission and the concentration and/or deposition of reduced nitrogen has improved.

### **23.3 Reflection of Emissions and Changes Therein in Monitoring Data**

#### **23.3.1 Conclusions**

##### **23.3.1.1 Is There Still an Ammonia Gap in the Netherlands?**

Originally, the ammonia gap existed in two parts: (i) an absolute difference between measurements and concentrations based on modelling and (ii) a difference in trend in measurements and modelled data. There have been several studies done focusing on explaining these two gaps (see Bleeker et al. 2009). There is no difference between the measured and modelled trend. By extending the monitoring period, improving the emission estimates and taking the meteorological conditions into account this is solved. However, the absolute systematic difference is still in the order of 30%. The explanation of the difference can be by two factors, probably contributing both: (i) underestimation of certain emissions and (ii) parameterisation (overestimating) the dry deposition in agricultural areas. In source areas advection might play a significant role, but this is of more importance for the very small scale. However, when selecting monitoring sites this aspect has to be taken into account.

##### **23.3.1.2 Does this Ammonia Gap Exist in Other Countries in Europe?**

Apart from the Netherlands and Denmark, where emissions decreased by about 30%, in most countries where monitoring takes place the concentrations did not change much (UK, Switzerland). In these countries the emission reductions were only very limited. However, no systematic gap such as in the Netherlands is signalled (UK, DK) or clearly detected by deposition or concentration measurements (Switzerland).

For the whole of Europe it is questionable if the decrease in emissions can be detected that took place in the early nineties, during the transition period (to market economy) when significant structural changes occurred leading to vast improvements in efficiency of production in the Eastern part of Europe. This is troubled by the fact that at the same time the SO<sub>2</sub> emissions strongly decreased affecting the lifetime and transport distance of ammonia (Horvath et al. 2009).

The EMEP monitoring sites that have been in place are not aimed to detect ammonia from agriculture and therefore the signal is not detectable. Furthermore, for the new EMEP monitoring strategy there is lack of implementation especially in Eastern Europe. It is therefore concluded that the evaluation of the absolute emissions in Europe and the changes therein is difficult because of lack of monitoring data covering the whole of Europe.

### **23.3.2 Recommendations**

Even though several studies and sensitivity analysis on the Dutch ammonia gap have been done, we suggest that the high temporal resolution site data might be used to interpret difference between model and measurements. Furthermore, emission modeling and the effect of meteorology and/or the net surface exchange need further improvement. Gradually the emission-deposition modeling should be integrated because they are strongly related and influenced by the same factors (meteorology). A quantification of the uncertainty in model input (emissions), parameters and output, together with a quantification of the uncertainty in the measurements is necessary. Because Denmark and the UK could not detect a significant gap the Working Group felt that it would be beneficial to exchange models and apply the Dutch model on the Danish and UK data and vice versa. Furthermore, modeling experience from the US could be taken on board.

## **23.4 Are We Confident About the Effectiveness of Ammonia Mitigation Policies?**

### **23.4.1 Conclusions**

Abatement options are usually tested on their effectiveness in the lab or under controlled conditions. The controlled conditions should reflect the practice in the field. These experiments form the basis of the abatement effectiveness. In practice the efficiency can differ because the practical applicability might be different. This is seen in the Netherlands for example in the case of application techniques of manure. The effectiveness of abatement options is also addressed in the Cross-Cutting Group A on the reliability of ammonia emission data and abatement efficiencies (Section 6.5).

Current monitoring is not focused on evaluating individual abatement options. The monitoring is focused on evaluating the changes in concentration/deposition to see if exposure of ecosystems improves. There are however, special case studies which focus on the abatement efficiency, such as the STOP program and the Veld study in the Netherlands, the food and mouth disease (FMD) measurement campaign in the UK, etc. Sectoral changes or individual abatement options can therefore currently not be detected with existing monitoring data. Local studies in this case are relevant to find sectoral changes.

Big changes in emissions have been detected using monitoring data. These changes either result from implementation of abatement options, such as in the Netherlands or Denmark, or as the result of reducing animal numbers due to the economic situation (Eastern Europe). The trends in emissions (including abatement measures) are in agreement with trends in measurements.

In Europe, where there is only limited monitoring of wet deposition and aerosol concentrations, there are confounding factors that result in deviating trends, such as the effect of the reduction of SO<sub>2</sub> emissions in Europe. This resulted in changes in ammonium deposition and concentrations. Furthermore, year to year changes in meteorology can have effects on observed trends and need to be filtered out with models.

### **23.4.2 Recommendations**

It is recommended to initiate local studies when large changes are expected (e.g. FMD). Furthermore, we need to quantify the chemical effects better in models. Also the meteorology effects on emissions and depositions have to be quantified better. For the monitoring it is useful to follow a monitoring strategy aimed at asking the question and resulting indicators.

## **23.5 Recommendations on Improvement of Modelling and Monitoring Activities**

### **23.5.1 Conclusions**

From the background document and the Working Group discussions it was concluded that evaluation of emissions and the changes therein need a modelling and monitoring strategy. Currently, the monitoring activities are not suitable to evaluate emissions because they have not started aiming for evaluating agricultural data. Two exceptions are the Netherlands and the UK. However, EMEP modelling and monitoring activities were started in a time when sulphur and oxidized nitrogen emissions caused impacts and protocols were agreed upon to reduce its emissions.

The sites that have been established at that time not necessarily are suitable for evaluating agricultural emissions, such as ammonia.

### **23.5.2 Recommendations**

A good modelling/monitoring strategy therefore starts with the basic question we want to address. This starting point is driven by the current policy items based on worries about impacts, such as impacts on biodiversity, human health and climate change. These impacts lead to questions about the contribution of ammonia, its emissions, abatement options to reduce emissions and validation and verification options. These form the starting point for the modelling/monitoring strategy. The first activity would therefore be to evaluate CLRTAP/EU/national policy objectives, based on impacts (biodiversity, PM and human health, climate change) and to determine the expected changes at the different scales (emission, concentration, deposition). The policies might, e.g. be focused on the protection of ecosystems (biodiversity) through decreasing critical load exceedances. This means that deposition needs to be monitored for different ecosystems. However, since deposition monitoring, especially of dry deposition, is not possible, the concentration monitoring provides a good alternative, provided issues like the changes in surface affinities are taken into account.

We therefore have to take into account what is already in place; a monitoring strategy builds on current facilities. Therefore it is strongly advised to implement the EMEP monitoring strategy and improve and extend it by focusing on:

- Monitoring of the spatial variations in ammonia emissions
- Detect the expected changes in emissions
- Focusing on all output parameters of the model relevant for changes in ammonia (e.g. N-balance)

The way to move forward with this is to do a pre-study based on current knowledge. The aim of such a study would be to optimise the spatial and temporal resolution of monitoring data, given the current monitoring sites and policy questions, the current emission, transport and deposition modelling, the impacts and the level of integration (e.g. pollutant swapping, climate change, etc.). Once the pre-study has been done the locations are selected and the temporal resolution, the required precision and accuracy and the representativeness are known. It is essential to use monitoring equipment with enough and known quality for specific applications; harmonised, intercompared, QA/QC, etc., not changing over the years. The past years instrumentation has been improved and new methods might be considered for monitoring depending on their application (MARGA, photoacoustic, lasers, passive samplers, DELTA, etc.). Finally, resources should be reserved for improvement and application of models to do good assessment of monitoring results (emissions, dry deposition, atmospheric chemistry, dispersion, transport) and answer the policy questions. Additional to this local studies and

impact assessment for special issues might be initiated, e.g. in cases where large emission reductions are expected (e.g. the FMD study in the UK).

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