

WORKING GROUP 2: MAKING THE LINK BETWEEN AMMONIA EMISSIONS, CONCENTRATIONS AND DEPOSITION.

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1. Introduction

The Working Group addressed the issues involved in making the link between estimated national NH₃ emissions and measurements of NH_x concentrations and deposition. The impetus for the working group was the need to quantify independently the effectiveness of NH₃ emission abatement, since it is impossible to monitor national NH₃ emissions directly.

The discussion was informed by the case studies provided in the workshop background document. Particular attention was given to the Netherlands, where questions have been raised over the effectiveness of the abatement measures. The Dutch case study was compared with data from other countries where NH₃ emissions should have changed on the basis of altered source sector activity. The Working Group addressed the uncertainties in the analysis in relation to current national emission reporting, process understanding, generalization and current monitoring activities. For each of these, recommendations were agreed which are reported here (with # denoting a priority recommendation). It was recognized that there are institutional limitations to implementing the recommendations (that in some cases have been made before) and an urgent need to review these limitations is identified. Finally, the Working Group made a first review of the requirements to assess compliance with national ammonia emission ceilings.

2. Case Studies of expected emission cHanges

2.1. CONCLUSIONS

2.1.1 DUTCH 'AMMONIA GAP'

The example of the NH₃ abatement policy in the Netherlands is important since it raises questions regarding the efficacy of abatement measures and the extent to which this can be assessed from atmospheric monitoring. The lack of a measured decrease in NH₃ concentrations for 1993-1997 in the Netherlands led to concerns for some parties in negotiating emission ceilings under the Gothenburg Protocol. Given the costs of implementing abatement strategies, these parties felt that more certainty was needed before agreeing to major technical abatement programmes. In particular, much of the reduction under the Dutch NH₃ abatement policy was supposed to result from the use of low emission manure spreading methods. Hence, these methods come under particular scrutiny.

The 'ammonia gap' needs to be considered in relation to: a) reliability of the emission and abatement estimates, b) offsetting factors that may have affected the abatement efficiency, c) interactions which may mask the trends when evaluating atmospheric monitoring data. Further information as regards c) is provided from the other countries where emission reductions were expected due to altered livestock and fertilizer quantities.

a. Reliability of the emission abatement estimates

Abatement measures may not have been applied as widely as estimated, while the measures themselves may not have been as effective as anticipated. In recognition of

the ammonia gap, a survey of farmers practice was made. This revealed several differences between intended and actual practice. In particular, many farmers had misunderstood the definitions and had reported use of 'deep injection' in field application of slurry, when they actually used shallow injection. These factors have resulted in a recent revision of the expected Dutch abatement from 35% to 25%.

Full details of the assumptions in the Dutch calculations need to be made openly available for comparison by interested parties (e.g. extent of compliance assumed, assumed % reductions from the different component measures).

b. Off-setting factors

During the period in which the NH₃ abatement measures were introduced other changes in agricultural management have occurred which may have offset the expected reductions in NH₃ emissions. These include policies to reduce nitrate leaching, improve animal welfare and reduce car NO_x emissions.

The nitrate policy in the Netherlands led to the prevention of manure application during winter months. As a result, more manure is spread in spring and summer months. While this would tend to reduce nitrate leaching, the warmer conditions of spring and summer favour increased NH₃ emissions. While it is agreed that this analysis is qualitatively sound, currently no estimates have been made regarding the quantitative effect of this interaction on Dutch NH₃ emissions.

It is noted that traditional tied stalls tend to give lower NH₃ emissions than cattle housing systems that allow free movement of the animals. A move away from tied systems, on the basis of animal welfare, may therefore have tended to increase NH₃ emissions. However, it has been argued for the Netherlands that this tendency would have been offset since new cattle housing (e.g. green label systems) also incorporates low NH₃ emission features.

It is well known that cars fitted with 3-way catalysts to reduce NO_x emissions emit much more NH₃ than other vehicles. With full take-up of catalytic converters, vehicles may account for ~10% of a country's NH₃ emission. More complete and open reporting is necessary to ensure that these changes have been properly recognized in national emission estimates.

c. Atmospheric interactions

Even if NH₃ emission reductions had been fully successful in the Netherlands, several factors tend to mask the trends in NH_x concentrations and deposition. These include weather conditions, interactions with SO₂ and NO_x, and the diurnal variability in emissions.

Weather conditions. The atmospheric behaviour of NH₃ is closely coupled to weather conditions. Emissions, dispersion, atmospheric transport, wet and dry deposition are all affected, with the result that there is substantial intra- and inter-annual variability. The NH₃ policy in the Netherlands should have reduced emissions from 1993, but full monitoring datasets have only been analyzed up to 1997. Given the large weather-induced variability, this is a very short time to evaluate trends in NH_x. In particular, it should be noted that:

- No comprehensive monitoring of NH₃ concentrations was available in the Netherlands before the abatement measures were introduced
- 1996 and 1997 were dry years, which would have tended to increase NH₃ concentrations.

- A local area study in a source region of the Netherlands indicated more than expected reductions in NH₃ concentration in 1998, which was a very wet year. The reduction cannot be fully explained by increased scavenging of NH_x and it has been suggested that emissions were also less (wet conditions, less manure application to wet soils).

SO₂ and NO_x. At the same time as the NH₃ abatement policy has been implemented, SO₂ and NO_x emissions in the Netherlands and across Europe have decreased. This interaction explains part of the NH₃ gap:

- Modelling has shown how a decline in SO₂ and NO_x emissions reduces the rate of NH₄⁺ aerosol formation, thereby increasing NH₃ concentrations. One estimate suggests that since 1984 this has tended to increase NH₃ concentrations by 10%.
- Reliable data on NH₄⁺ aerosol concentrations are available in the Netherlands and these show the largest decrease. Measured NH₄⁺ concentrations decreased by 29% between 1993-97 compared with 12% expected from model calculations.
- The trends in NH₄⁺ aerosol and wet deposition also reflect transboundary fluxes and are complicated by changes in NH₃, SO₂ and NO_x emissions in other countries.

Diurnal profiles. The diurnal profiles of NH₃ emissions from manure spreading and animal housing are different. While nearly all spreading emissions occur during the day (fresh application, ground warmest and driest, increased turbulent mixing), emissions from animal houses vary much less. Due to the reduced atmospheric mixing at night, night-time emissions result in higher NH₃ concentrations. Animal housing therefore has a larger effect on average NH₃ concentrations than manure spreading (per kg emitted). As a result, it is harder to detect changes in NH₃ concentrations due to manure spreading policies than housing. This is important given that most of the estimated reduction in the Netherlands was due to spreading.

2.1.2 EXPECTED CHANGES IN NH₃ EMISSIONS FOR OTHER COUNTRIES

Analysis of the case studies from other countries re-enforce the conclusions regarding the atmospheric interactions. In particular it was noted:

- Although there is uncertainty in the emission reductions for Eastern Europe, the change in sector activity that should result in reductions appears to be real (observation of disused farms and uncultivated fields).
- In many instances, it is difficult to see a reduction in NH_x for east European sites despite the expected emission reduction. This emphasises the masking effect of atmospheric interactions.
- Most data are available for NH₄⁺ in precipitation and NH₄⁺ wet deposition. Trends in these quantities are particularly sensitive to weather differences, adding scatter to long-term trends. For measurements at one site, it should not be expected to detect a trend in regional NH₃ emissions of less than 50% in less than 5 years.
- Detailed site based analyses are important to address processes, but for trend analysis it is also essential to compare data from many sites.
- Downward trends in NH₄⁺ in precipitation have been measured in former East Germany, Russia, Denmark and to a lesser extent Switzerland and former West Germany. These trends are consistent with altered NH₃ and SO₂ emissions at a European level. In some cases the trends are statistically different from zero.
- Data from Denmark show the largest reduction in NH_x. Aerosol NH₄⁺ decreased by 34% between 1989-99, compared to 6% for NH₃ and 20% for NH₄⁺ in wet

deposition. The changes in aerosol are larger than Danish NH₃ emission abatement (~20%) over the period and may be explained by an SO₂ interaction and at least partly by transboundary fluxes.

In addition to these features, the data highlight that:

- Ammonia fluxes with vegetation are bi-directional, due to the existence of a 'compensation point' concentration. This may buffer NH₃ concentrations, particularly when monitoring at remote sites, so that less reduction is seen in the monitoring record. This effect introduces non-linearity into the emission-concentration-deposition relationship.
- As a secondary product, NH₄⁺ aerosol is not very spatially variable and can be adequately monitored at only a few sites per country. By contrast, NH₃ is very spatially variable, with the result that the area of influence at a monitoring station is very small (e.g. >50% influence in less than 1 km radius).

2.1.3. KEY FINDINGS FROM THE CASE STUDIES

Putting together the findings from the Netherlands and other countries where emissions have changed, it can be concluded that:

- a) Reductions in concentrations and deposition are sometimes difficult to detect from single site data, but are seen with sufficient time and area averaging.
- b) The abatement policies in the Netherlands and Denmark have been successful in reducing NH₃ emissions, even if the expected decreases may not have been fully realized.
- c) More than expected reductions are seen in NH₄⁺ aerosol, compared with less than expected reductions in gaseous NH₃, for example due to interactions with changing SO₂ and NO_x. This emphasizes the transboundary nature of NH_x pollution and the need for international agreement on emission controls.

2.2. RECOMMENDATIONS

In order to improve the assessment, improved reporting, monitoring, process description and modelling is necessary. Detailed recommendations are provided in later sections. However, it should in particular be noted that:

- The Dutch comparison between modelling and measurement needs to be re-run using latest emission reduction figures (25%), including all NH₃ sources directly in the model and covering the period up to 2000. #
- The trends in measured NH_x in Eastern Europe and elsewhere should be compared with model estimates, specifically addressing the reliability of emission abatement estimates, off-setting factors due to changes in agricultural management, the interactions with other pollutants and non-linearities such as the ammonia compensation point. In particular, further modelling of trends in Denmark is necessary since this provides a second example of a country with a major NH₃ technical abatement policy. #
- Regional monitoring of NH₃ concentrations must quantify site representativity, and ideally compare many sites. #
- Adequate monitoring of NH₃, aerosol NH₄⁺ and wet deposition should ideally be in place before NH₃ abatement measures are introduced. #

3. Reliability of national emission data

3.1. CONCLUSIONS

Detection of trends in monitored atmospheric concentration and deposition can only be made against a reference of reliable temporal trends in emission data. Although the absolute value of a national NH₃ emission is itself subject to uncertainty, it is even more important when considering trends to report emissions for different years using a consistent methodology and input data. At present, national submission of emission data is on major-sector basis (e.g. transport, agriculture, nature etc). This is not sufficient to evaluate fully the reliability of the temporal trends in emissions data submitted by parties to the Convention. Improved data are needed in relation to SNAP (Selected Nomenclature for Air Pollution) sub-classes, agricultural management and NH₃ emission factors.

3.1.1 SNAP SUB-CLASS DATA

Statistics on agricultural activity are a major basis for deriving NH₃ emission estimates. It is essential to quantify the reliability and uncertainties in the specific sectors contributing to NH₃ emissions as specified in the Atmospheric Emission Inventory Guidebook (AEIG). Currently, uncertainty estimates are not generally available for animal numbers (e.g. numbers of dairy cows, fattening pigs, laying poultry etc) and fertilizer use. To assess whether a trend between years is reliable, uncertainty estimates (as upper and lower confidence limits) need to be provided for each category and submitted on a yearly basis together with the sector activity data. These need to be provided as part of reviews to the Convention.

For example, it is noted that estimated NH₃ emissions in Eastern Europe decreased on average by 45% between 1989 and 1996. This change is estimated solely on the basis of changed sector activities for the different livestock numbers and amounts of fertilizer used.

The same principles apply when considering non-agricultural sources of NH₃. Parties should specify the SNAP sub-class uncertainties contributing to reported non-agricultural emissions.

3.1.2 AGRICULTURAL MANAGEMENT DATA

We need reliable data on: nitrogen excretion by animals, the distribution of animals between different animal types, the distribution of manure between different storage types (e.g. open vs. covered storage; lagoon vs. slurry tank), field application methods for each manure type. These data are necessary because of the substantial differences between countries in the feeding practices and substantial differences between housing, storage and spreading types in the emission coefficients.

These data are essential to demonstrate the extent of abatement measures being implemented under the Gothenburg Protocol.

3.1.3 EMISSION FACTORS

Parties should be encouraged to adapt the emission coefficients (AEIG default methodology) to reflect regional and national differences in emissions factors due to region specific agricultural practices, weather conditions and soil types. However, where countries deviate from the AEIG default values, scientific justification must be provided to accompany the annual submission of emission data. The national submissions of ammonia emission data and the accompanying scientific justifications should be collated centrally and published openly by the Convention. This should encourage scientific debate on the robustness of estimates provided by different parties to the Convention.

3.2. RECOMMENDATIONS

Annual submissions of national NH₃ emission data to the Convention should specify:

- ammonia emission per sub-sector activity (e.g. animal and fertilizer types) #
- the contribution of each major component source to the emission (e.g. housing, storage, spreading, grazing), except where the Emission Inventory Guidebook simple methodology is used. #
- the statistics of SNAP sub-class activity assumed in the calculations should be stated in submission of emission estimates, giving upper and lower uncertainty limits. #
- the % occurrence estimates (with upper and lower uncertainty limits) of different agricultural management practices, including abatement options, that are assumed for the emission calculations
- scientific justification should be provided for emission factors that deviate from default values in the Emission Inventory Guidebook. A similar justification is needed to explain changes in methodology compared with reporting in previous years. #
- The national submissions including scientific justification should be collated and published openly (including on the internet). #

4. Process understanding

The extent of process understanding of atmospheric NH_x was recognized as a limitation to quantifying the link between emissions, concentrations and deposition. Although much is known about these issues, there are several key uncertainties which require experimental measurement and process modelling.

4.1 CONCLUSIONS

4.1.1 EMISSION

- In general the understanding of the NH₃ emission process is good, though there are problems in the process understanding of emissions from solid manures. In particular, it is difficult to predict the extent of composition of composting and hence NH₄⁺ production.

4.1.2 DISPERSION AND TRANSPORT

- Quantification of atmospheric dispersion is poor in stable night-time conditions. Improvements are necessary given the importance of dew to local NH₃ dry deposition.
- The influence of complex terrain on NH₃ dispersion is poorly quantified, in particular including the effects of roughness changes such as woodland edges and transport of NH₃ over hilly/mountainous areas. Improved descriptions of the advection of air masses into tall canopies are necessary.

4.1.3 DEPOSITION

- NH₃ exchange with vegetation is recognized as being bi-directional. This means a) that the net flux at a given time is a result of upward (emission) and downward (dry deposition) components, b) that net fluxes with emission and deposition occur over a single canopy type at different times (on hourly, diurnal and seasonal scales).
- While many of the mechanisms of NH₃ dry deposition/exchange are understood (e.g. cuticular exchange, stomatal exchange, stomatal compensation point), there

are major uncertainties regarding the extent of soil and leaf litter emissions, and the extent to which these are recaptured in relation to canopy structure and environmental conditions.

- The controls on the plant stomatal compensation point are currently poorly understood. Initial measurements and modelling demonstrate a close link to management and plant phenology, but improved understanding is necessary in relation to ecosystem functioning to address management interactions with more certainty.
- Local NH₃ dry deposition is poorly quantified, with key issues being the potential saturation cuticular uptake near point sources in relation to environmental conditions and the advection of NH₃ into tall canopies.
- The quantification of advective transport of NH₃ from farms into nearby forest stands is poorly quantified. This is relevant when considering the potential for local recapture of NH₃ emissions.

4.1.4 CHEMISTRY

- Important atmospheric chemistry interactions of NH_x include interconversion between gaseous NH₃ and aerosol NH₄⁺, reactions with clouds, precipitation scavenging and interactions between pollutants in the dry deposition process.
- SO₂ was formerly the most important pollutant for NH₃ interactions, given the high emissions and lower vapour pressure of ammonium sulphates compared with ammonium nitrate. However, with decreasing SO₂ emissions the interactions between NH₃ and HNO₃ become even more important. As NH₄NO₃ is volatile altered atmospheric transport distances may be expected.
- While the chemistry of pure ammonium sulphate or ammonium nitrate is reasonably well understood the rules appear to break down for mixed chemistry. Sulphate may lower the vapour pressure of associated ammonium nitrate affecting balances, although this effect is currently poorly quantified.
- Although ammonium sulphate is usually considered to be stable, interactions in clouds in the presence of nitrates can re-form ammonium nitrate, allowing re-volatilization of NH₃. This effect has been demonstrated in the field, but is currently poorly quantified.
- The time scales of gas – aerosol inter-conversion are currently poorly quantified and are relevant in defining both local deposition and regional transport. In particular, the consequences of aerosol size distributions are poorly quantified.
- The same chemical interactions that occur on aerosols and in clouds apply to leaf surfaces and therefore affect the dry deposition process. Although the SO₂ interaction with NH₃ cuticular uptake is partly understood, improved quantification is essential if trends in NH₃ deposition are to be explained in relation to changing SO₂ concentrations.

It should be recognized that most chemistry is based on north European experimentation and that atmospheric transformations of NH₃ may be different in warmer climates such as southern and eastern Europe. To some extent these reflect the chemical situation in California which has been well studied.

4.2. RECOMMENDATIONS

Further work is required to improve process understanding in relation to the following:

- the mechanisms of NH₃ emission from solid manures, including interactions with composting. (emission)#
- improved description of local dispersion under stable night-time conditions. (dispersion)
- the influence of complex terrain on NH₃ dispersion including roughness changes between short/tall canopies and complex topography. (dispersion) #
- the mechanisms of dry deposition/bi-directional NH₃ exchange with different plant canopies, in particular the extent of soil/litter emissions and their recapture by an overlaying canopy. (deposition) #
- the biological controls on the NH₃ compensation point in relation to ecosystem functioning and management interactions in defining net dry deposition. (deposition) #
- the saturation of NH₃ dry deposition to plant cuticles near point sources and the advection of NH₃ into forest stands. (deposition) #
- the interactions of NH₃ with SO₂ and NO_x chemistry, in particular dealing with the effect of chemical mixtures including SO₄²⁻ and NO₃⁻, and the effect of cloud processing in remobilizing NH₃ from ammonium sulphates. (chemistry) #
- the time-scales of gas-particle inter-conversion processes in relation to aerosol size distributions. (chemistry)
- the quantitative influence of SO₂ and NO_y on NH₃ dry deposition rates including the interaction with other ions on leaf surfaces. (chemistry) #

5. Generalization

Generalization of processes is an important limitation to quantifying the atmospheric controls on NH₃. This includes for example: going from detailed process based models at fine spatial and temporal scales to more general models considering larger areas and longer time-scales, with a concurrent development of parametrizations for the key processes.

5.1. CONCLUSIONS

5.1.1 EMISSION

- The dynamic link between emission and dispersion models is understood, but not sufficiently and further work is required in actually developing detailed approaches to link these two components.
- Further data are necessary regarding detailed statistics of manure practices, as simple annual NH₃ emissions are not sufficient when considering NH₃ abatement. In particular, seasonal variation in agricultural practices in relation to climate is necessary.
- Measurements of NH₃ emission need to be interpreted in relation to the measurement approach used (e.g. wind tunnel estimate of urine emission vs. micrometeorological assessment of field level fluxes). Most atmospheric transport modelling uses net (top of canopy) emissions at a field level and from whole buildings/stores. The results of finer scale measurements need to be generalized accordingly.

5.1.2 DISPERSION AND TRANSPORT

- The methods in dispersion modelling for upscaling (going to larger areas) and zooming (going to smaller areas) for ammonia are currently uncertain bearing in mind the non-linearities specific to ammonia.
- Many parameters in atmospheric dispersion and deposition models are rather uncertain. Improved description requires additional input data in many cases.
- Models range from highly detailed deterministic approaches to simple statistical treatments of dispersion and deposition. Both approaches have validity, with the latter in particular useful for considering spatial variability.
- The effects of topography and patchiness (spatial pattern of ground cover) are currently uncertain at the process level and substantial work is necessary before these effects can be generalized. Nevertheless, there is an urgent need to incorporate simple estimates of the effects into generalized dispersion models.

5.1.3 DEPOSITION

- There is a growing body of information on compensation points for specific vegetation and management. The effect of temperature is well understood allowing the temperature normalized compensation point parameter Γ to be estimated. However, substantial work is required to generalize databases for the full range of relevant agricultural and semi-natural plant canopy types.
- Improved generalization is necessary for agricultural crops in defining bi-directional exchange in relation to management activities, while appropriate land cover databases are necessary to allow scaling up.
- While monitoring of throughfall (precipitation wash-off) under forest canopies is often made for NH_x , it must be recognized that canopy exchange processes mean that this is not the same as total NH_x deposition. Improved methods are necessary to allow interpretation of trends in these measurements in relation to likely changes in canopy exchange with time (due to e.g. changing SO_2 concentrations).

5.1.4 CHEMISTRY

- The findings of process studies on atmospheric chemistry need to be incorporated into generalized atmospheric transport models. In particular, the potential for re-volatilization of NH_3 from ammonium sulphate (following cloud interactions with nitrate) should be recognized, as well as the effect of mixed chemistry on the balances.
- Improved descriptions are necessary of aerosol chemistry in relation to gas-particle inter-conversion time-scales and size distributions. The influence of these effects on atmospheric transport distance needs to be assessed.
- Initial dynamic models of the interaction of NH_3 with SO_2 , NO_y and other ions on leaf surfaces are available. A major challenge is to generalize the results from such detailed modelling into atmospheric transport models to allow both the influence of cloud chemistry and dry deposition changes to be addressed when considering the effect of regional reductions in SO_2 .

5.2. RECOMMENDATIONS

Further work is required to improve generalization in relation to the following priorities:

- Improved datasets need to be collected regarding seasonality of agricultural practice and manure handling. (emission). #

- the further development of coupled emission, dispersion and deposition models incorporating the coupled dynamic link between emission and dispersion/deposition. #
- improved methods for upscaling and zooming for ammonia allowing a dynamic link between scales to deal with non-linearities. (dispersion)
- the development of initial (even if uncertain) approaches to generalize the influence of topography and patchiness in atmospheric dispersion models (dispersion).
- the parametrization of dry deposition for NH₃ incorporating stomatal and cuticular exchange pathways for application atmospheric transport models. #
- the collation of dry deposition compensation point parameters for vegetation types and management conditions, together with appropriate land cover data (deposition). #
- the incorporation of cloud interactions into atmospheric transport models, including the potential for re-volatilization of NH₃ from ammonium sulphate. (chemistry). #
- the revision of gas-particle interactions in atmospheric dispersion models to account for time-scales and size distributions for NH_x. (chemistry)
- the generalization of dynamic models of NH₃, SO₂ and NO_y interactions with leaf surfaces as simple steady state approximations for incorporation in atmospheric transport models. (chemistry) #
- the assessment of scenarios of changing SO₂ emission separating the effect of cloud/aerosol chemistry and dry deposition chemistry on NH₃ concentrations and deposition. (chemistry/deposition). #

6. Monitoring of NH_x

6.1. CONCLUSIONS

Atmospheric pollutant monitoring is generally conducted as a reference against which to assess environmental impacts. In the case of NH_x, these impacts are acidification, eutrophication and secondary aerosol formation. Since acidification and eutrophication effects are a response to long-term deposition, which varies spatially, the priority is for long-term data at sufficient sites to detect spatial patterns. For this purpose, the measurements can have a low temporal resolution. The contribution of NH₃ to aerosol formation is an issue of increasing interest. Here, effects are related to a) long-term increases (global temperature balance) and b) high concentration 'episodes' (human health, visibility). Hence monitoring in relation to ecosystem effects will follow a different strategy to monitoring to assess human health and visibility effects.

It is noted that different countries in Europe have varying NH_x monitoring capability in terms of species considered, temporal resolution of sampling and density of sites. In the context of the Convention, countries contribute to the measurement network of the EMEP programme. Historically, the EMEP network had a strong emphasis on measurements of precipitation chemistry. However, this is a derived quantity (from NH_x concentrations and precipitation amount) and it is recognized that monitoring gases and aerosols is even more important.

It should be noted that in 1997 a workshop was held at Aspenäs Herrgård, Sweden under the auspices of EMEP and WMO to revise atmospheric monitoring strategies

for regional air pollutants (Schaug and Uhse 1997). In terms of the effects of NH_x the following issues were raised (Lövblad and Sutton 1997):

1. Much effort is placed under EMEP in monitoring on a daily basis (air concentrations and wet deposition). In most cases, this is more than necessary, since weekly to monthly data is sufficient to interpret long-term trends.
2. Few sites in the EMEP network currently distinguish between gaseous NH_3 and aerosol NH_4^+ . (A filter sample of total NH_x is made at most sites). This is a key limitation to interpreting the results since the spatial and temporal patterns of NH_3 and aerosol NH_4^+ are very different.
3. Studies using active sampling methods have been used exclusively in the EMEP programme. When continuously calibrated, passive samplers for ammonia can provide a cheap and reliable method for examining spatial variability of ammonia in source regions with a low temporal resolution (e.g. monthly).
4. Site selection for monitoring of NH_3 concentrations must consider the underlying vegetation and management type, since vertical concentration gradients can affect values. To standardise measurements, these should be carried out over short grass without fertilizers or grazing
5. By reducing effort given to high temporal resolution daily data, more resources could be allocated to distinguishing gaseous NH_3 and aerosol NH_4^+ and increasing spatial coverage.
6. It is necessary to quantify the detailed temporal dynamics of NH_3 concentrations for model development. Similarly, continuous NH_3 dry deposition monitoring may be achieved by monitoring at several heights together with appropriate micrometeorological equipment. However, both these tasks are expensive, and can only be conducted at a limited number of 'super sites'.

It was agreed by the Working Group that in general terms these conclusions still hold true. However, the following caveats and conclusions were made:

- A limited number of daily monitoring sites should be retained, bearing in mind the new need for aerosol monitoring. However, in this case separate sampling of NH_3 and NH_4^+ is essential.
- Long term measurements of NH_3 and NH_4^+ should avoid use of filter packs, which are affected by sampling artefacts. Denuders provide a more reliable separation but must be supported by adequate quality assurance methods.
- Passive sampling can be adequate with care and continuous calibration. However, replication is essential and some approaches are not suitable for low concentrations ($<2 \mu\text{g m}^{-3}$)
- Care is needed to site normal sampling away from point sources (e.g. $>100\text{-}200$ m), although representative sampling should consider both near source and remote sites. Ideally measurements should be accompanied by meteorological data (wind direction, wind speed, temperature, humidity, solar radiation).
- It is essential to establish the long term stability of sampling protocols, preferably against an independent reference.
- With regard to super-sites, there is a need to improve dry deposition monitoring methods, including detailed spectroscopic methods as well as low cost techniques. Such super-sites need to be located in high and low NH_3 emission areas, as well as in north and south Europe. Currently, it is concluded that the challenges of

measuring fluxes mean that reliable NH₃ flux monitoring over forest is not possible at present.

- Regional flux sampling is desirable, but it is not currently feasible for NH₃.
- Strategies for monitoring depend on the objective. Within the Convention, the initial objective was to monitor transboundary pollution, with a consequent focus on remote sink areas. Now that the Gothenburg protocol is signed, focus will increasingly shift to monitoring compliance. More effort is therefore needed to monitor NH_x in source regions, which are most closely affected by national emissions.

6.2. RECOMMENDATIONS

6.2.1 SAMPLING FREQUENCY

- Detailed temporal data (<hourly) is only necessary at a few sites, with the focus being to understand process interactions, or to monitor dry deposition to key sites. As this activity is expensive for NH₃, it can only be made in some countries at a few sites per country. #
- A primary requirement is to detect long-term trends and spatial patterns. For this purpose cheap monitoring methods are needed. Sampling with denuder-filter combinations has been shown to operate reliably at weekly to monthly levels. #
- Where protocols are validated, passive sampling methods can be adequate at a monthly or fortnightly frequency. Continuous reference against active sampling is essential.
- Less effort should be given to monitoring wet deposition on a daily basis, as weekly precipitation data is sufficient to assess long term trends. #

6.2.2 WHAT CHEMICAL COMPONENTS TO MEASURE

- Sampling for total NH_x in air should be replaced throughout the EMEP network with separate sampling for gaseous NH₃ and aerosol NH₄⁺. #
- Sampling using a denuder-filter combination can provide cheap, reliable speciation between NH₃ (collected on the denuder) and NH₄⁺ (collected on a subsequent filter).
- Passive sampling is the cheapest method to characterize the variability of NH₃ in source areas, where concentrations are sufficient and methods are validated.
- It is necessary to continue records of ammonium in precipitation and wet deposition. In high NH₃ emission areas, wet-only sampling is necessary.

6.2.3 WHERE MEASUREMENTS SHOULD BE MADE

- Since NH₃ is much more spatially variable than NH₄⁺ aerosol, measurements at many more sites are needed for NH₃ to quantify spatial patterns in relation to NH₃ source sink areas and regions with different source types. #
- Monitoring for NH₃ should be made with dense networks and a low sampling frequency to identify seasonal and long-term trends. Sampling should be stratified to cover regions dominated by different source types. Sampling density should depend on expected gradients, with more sites required in source areas. #
- In low ammonia regions, active denuder sampling is recommended to provide sufficiently precise data. In source regions, passive samplers may be applied. Since passive samplers do not require electricity, they may be installed more flexibly in dense networks.

- In addition to national distributions of NH₃ concentrations, demonstration local studies should be established in different countries to demonstrate a) the spatial variability of NH₃ at a local level (<5 km resolution) and b) the representativity of monitoring stations.
- Detailed monitoring at ‘super sites’ should focus on key source and sink regions of countries to characterize temporal dynamics and dry deposition fluxes. #
- Given the smooth distribution of aerosol NH₄⁺, sampling is necessary at only few (5-30) sites per country. #
- As records of NH₄⁺ in wet deposition are in most cases abundant, the need is to maintain the existing sites to observe long term trends. Siting criteria for wet deposition measurements have been discussed elsewhere (EMEP/CCC Report 1/95).

6.2.4 REQUIRED MONITORING DURATION

- To detect a significant trend in NH_x components would typically require >5 years due to the effect of within and between year meteorological variations. With this time perhaps only a change of 50% would be detectable from measurements at a single site. Since in many cases the expected changes are much smaller than this, it is necessary to assess trends by combining the results from several sites. #
- To detect small changes in NH_x components (<10-20%), in general would require a combined trend analysis using several sites with at least 10 years of measurements.
- The reliability of long-term protocols must be established, ideally against independent references. In particular, if methods change a period of 1 year overlap is necessary. #

6.2.5. IMPROVING THE ABILITY TO DETECT TRENDS

- Combining datasets improves the statistics and helps identify trends. Further consideration needs to be given to estimating national patterns of NH_x concentrations and deposition to improve trend detection. #
- More work is necessary to consider the benefits and limitations of statistical data analysis when assessing long term trends of data for many sites. In particular, methods to define trend confidence limits for entire national networks need further attention.
- Analysis of NH_x monitoring data needs to consider the distinct needs of a) monitoring transboundary pollution and b) monitoring to assess compliance.

7. Limitations to implementation

7.1. CONCLUSION

The working group noted with concern that few countries appear to have responded to the recommendations of the Aspenäs Herrgård Workshop regarding NH_x monitoring strategies. Similarly, a number of the recommendations regarding reporting of emissions data have been made before under the Task Force on Emission Inventories of the Convention.

Since this WG re-enforces those recommendations, it is important to address what are the institutional limitations that prevent the adoption of the recommendations.

7.2. RECOMMENDATION

- A review is necessary to address the fate of the recommendations of the Aspenäs Herrgård Workshop regarding NH_x monitoring.
- A review is necessary to address the limitations to adopting existing recommendations of the TFEI regarding emission data for NH_3 .

These reviews need to establish the institutional limitations to adopting the previous recommendations, considering the response by various Convention bodies and by Parties to the Convention. They are essential if further progress is to be achieved according to the recommendations made here. #

8. Reporting and compliance

With the signing of the Gothenburg Protocol, the first national ceilings for NH_3 have been established, together with revised ceilings for other pollutants. Within this context, much more attention will be given within the Convention to assessing compliance to the Protocol commitments. In the past, compliance assessment was simply based on national reporting of emission data. Under the new structure of the Convention, the Implementation Committee is investigating improved methods to assess compliance. This is likely to depend on improved reporting procedures, as well as utilize independent approaches based on monitoring data. These are new areas and the Working Group included a first discussion of the issues.

8.1. CONCLUSIONS

- Existing reporting of NH_3 emissions to the Convention is inadequate to assess compliance of national NH_3 emission ceilings.
- Improved reporting is essential. Reporting needs to include the breakdown to component sources of national ammonia emission, detailed activity statistics and assumptions used regarding the extent and abatement achieved by different measures. The recommendations of section 3.2 are all appropriate for assessing compliance.
- Monitoring of NH_x concentrations and deposition provides the only independent means to assess compliance of NH_3 emission ceilings, but depends on a) sound measurement networks and b) interpretation of the measured concentrations and deposition bearing in mind transboundary fluxes.
- Aerosol NH_4^+ and wet deposition are the most spatially representative, but closely reflect transboundary fluxes and interactions with other pollutants. Air concentrations of NH_3 reflect largely sub-national patterns of NH_3 emissions, but are also affected by interactions with other pollutants (to a lesser extent) and are highly spatially variable.
- Measured trends need to be contrasted with modelled trends, both to provide support to the interpretation of trends and to explain interactions with other pollutants.
- There is an urgent need to establish adequate NH_x monitoring before NH_3 emission abatement strategies are put in place. The target date for the Gothenburg Protocol is 2010. This allows a decade of measurements from now, which should be sufficient to detect the main trends. However, it will take 2-3 years for ratification, when the Protocol requirements come into force. Therefore improved monitoring for NH_x needs to be established as soon as possible before ratification of the Protocol.

- Gridded emissions at 50 km x 50 km are already a requirement of the Convention, and are currently only rarely provided. As the needs of monitoring move towards compliance assessment these spatial inventories become even more important. They are necessary to provide a closer match between monitoring and modelling, especially since the possibilities to abate NH₃ emissions are not distributed evenly across countries.

8.2. RECOMMENDATIONS

The following components are necessary to improve the assessment of compliance for national NH₃ emission ceilings:

- Reporting of national NH₃ emissions should follow the recommendations of Section 3.2. in order for compliance to be assessed more reliably. #
- Measurements of NH₃, NH₄⁺ aerosol and wet deposition are all essential to address compliance. Model studies are necessary to interpret trends in relation to a) interactions with other pollutants, b) meteorological variability and c) transboundary fluxes.
- Monitoring of NH_x components is necessary in source regions to assess compliance. This complements the traditional focus of the Convention of monitoring in remote areas. Monitoring of NH₃ must be sufficiently representative to assess national patterns. This will require a combination of measurements and spatial modelling. #
- Adequate national monitoring of NH₃, aerosol NH₄⁺ and wet deposition must be established as soon as possible, if at all possible before NH₃ emission abatement strategies are put in place. The target should be to obtain at least 8 years monitoring data at several sites before completion of the Gothenburg Protocol in 2010. #
- Gridded emissions at 50 km x 50 km need to be provided to the Convention to allow better comparison with measurement data in assessing trends.

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