ESTABLISHING THE LINK BETWEEN AMMONIA EMISSION CONTROL AND MEASUREMENTS OF REDUCED NITROGEN CONCENTRATIONS AND DEPOSITION

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Abstract. In the context of international efforts to reduce the impacts of atmospheric NH_3 and NH_4^+ (collectively, NH_x), it is important to establish the link between NH_3 emissions and monitoring of NH_x concentrations and deposition. This is equally relevant to situations where NH_3 emissions changes are certain (e.g. due to changed source sector activity), as to cases where NH_3 abatement technologies have been implemented. Correct interpretation of adequate atmospheric measurements is essential, since monitoring data provide the only means to evaluate trends in regional NH_3 emissions. These issues have been reviewed using available measurements and modelling from nine countries. In addition to historic datasets, the analysis here considers countries where NH_3 source sector activity changed (both increases and decreases) and countries where NH_3 abatement policies have been implemented. In The Netherlands an 'ammonia gap' was identified between the expected reduction and results of monitoring, and was attributed initially to ineffectiveness of the abatement measures. The analysis here for a range of countries shows that atmospheric interactions complicate the expected changes, particularly since SO_2 emissions have decreased at the same time, while at many sites the few years of available data show substantial inter-annual variation. It is concluded that networks need to be established that speciate between NH_3 and aerosol NH_4^+, in addition to providing wet deposition, and sample at sufficient sites for robust regional estimates to be established. Such measurements will be essential to monitor compliance of the international agreements on NH_3 emission abatement.

Keywords: ammonia, ammonium, dry deposition, emission abatement, international protocols, wet deposition

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1. Introduction

It has been known for some time that the atmospheric transport and deposition of reduced nitrogen (NH_x), either as ammonia (NH_3) or ammonium (NH_4^+), leads to eutrophication and acidification of ecosystems (e.g. Grennfelt and Thoernelof, 1992; Fangmeier *et al.*, 1994; Hornung *et al.*, 1995; Heij and Erisman, 1995). Recognizing this, and the transboundary nature of the problem, the UNECE has spent the last six years developing the first international protocol that includes limits to NH_3 emissions, alongside reductions for sulphur dioxide (SO_2), nitrogen oxides (NO_x) and volatile organic compounds (VOCs) (Grennfelt *et al.*, 1994; Bull and Sutton, 1998). The Gothenburg Protocol, which was signed in December 1999, sets national ceilings for NH_3 emissions that should be achieved by 2010, as well as identifies a number of specific measures that must be taken (UNECE, 1999). In parallel, the countries of the European Union have recently agreed the National Emissions Ceilings Directive (NECD), which establishes targets for NH_3 which are binding in European law (EC, 1997, 1999).

With these new agreements in place, it becomes of paramount importance to demonstrate that the planned emissions reductions for NH_3 are both achievable and measurable. With most of the NH_3 emissions resulting from agricultural sources (e.g. Asman *et al.*, 1998), the burden for NH_3 abatement falls on the agricultural sector. Emission reductions may be sought partly through expected reductions in animal numbers and fertilizer consumption. However, in many cases substantial resources are being given to developing and applying technical measures to reduce NH_3 emissions. While emissions reductions due to reduced sector activity should not be in question, it is necessary to demonstrate the effectiveness of any technical measures.

In the case of heavy industries, it is often possible actually to measure emissions, for example as emitted from stacks. By contrast for NH_3, emissions result from many small, complex sources, such as naturally ventilated animal houses, manure stores, slurry spreading, grazing and fertilized crops. In the case of NH_3, it is therefore impossible to monitor or even sample emissions coherently, and emission estimates are based on the results of experimental studies for a range of situations. The alternative to monitoring emissions that is usually applied is to establish networks to monitor atmospheric NH_x concentrations and deposition, with the intention to assess emission changes from temporal patterns in the monitoring record. In this approach it is necessary to have sufficient understanding of the controls on atmospheric concentrations, while comparison of expected trends with models becomes extremely helpful.

The challenge to link NH_3 abatement to atmospheric monitoring of NH_x species has recently become even more apparent as questions have been raised regarding the effectiveness of NH_3 emissions controls in The Netherlands (Erisman *et al.*, 1998; Erisman and Monteny, 1998). In this example, a number of reasons were put forward to explain why an expected 35% abatement of national emissions could not be detected in the NH_3 concentration monitoring network. These included both interactions with atmospheric processes and the suggestion that abatement measures may not have been as successful as anticipated. In such situations there is the tendency for any discrepancy that cannot be explained by atmospheric processes to be attributed to ineffectiveness of the abatement measures. This places increased attention on the ability to understand and model the atmospheric interactions of NH_x. A second example illustrates this: following the end of the communist era in eastern Europe, animal numbers and fertilizer consumption decreased substantially, so that NH_3 emissions must have reduced (Strogies and Kallweit, 1996; Tsibulski *et al.*, 1996). With monitoring data from a background station in Hungary, Horváth and Sutton (1998) showed that there was no parallel trend in either NH_3 or NH_4^+ components between 1981 and 1994, even though national NH_3 emissions should have decreased after 1989 by >50% from a peak in 1983.

There are therefore two distinct challenges:

1. to quantify the link between NH_3 emission changes and monitored atmospheric NH_x in situations where emissions have definitely changed;
2. bearing in mind the uncertainties in 1., to assess the effectiveness of NH_3 emission abatement policies.

It should be noted that 1. is not just an issue that affects NH_x. For example, there are still many questions regarding the linearity between SO_2 and NO_x emissions reduction and monitoring data. Certain non-linearities are incorporated in the atmospheric transport models (e.g. Bartniki, 2000), while other interactions remain poorly quantified. Such effects may, for example, explain why deposition of sulphur in remote regions has declined more slowly than predicted in models (RGAR, 1997).

This article addresses the link between NH_3 emission abatement and atmospheric measurements in these two stages. Firstly, it considers a series of case studies from Europe and America to illustrate inherent uncertainties in linking emissions to concentrations and deposition. Secondly, it considers examples from countries where NH_3 abatement policies have been implemented and uses these to address the extent to which monitoring data can determine the effectiveness of abatement measures. Finally, this article suggests where improvements are needed in: (a) field measurements, to improve process quantification, (b) atmospheric modelling, to integrate process understanding and (c) monitoring networks, to detect temporal and spatial changes.

2. Historic Datasets

Before considering recent examples of NH_3 emission reduction, it is important to demonstrate that longer-term changes in NH_x deposition have indeed occurred.

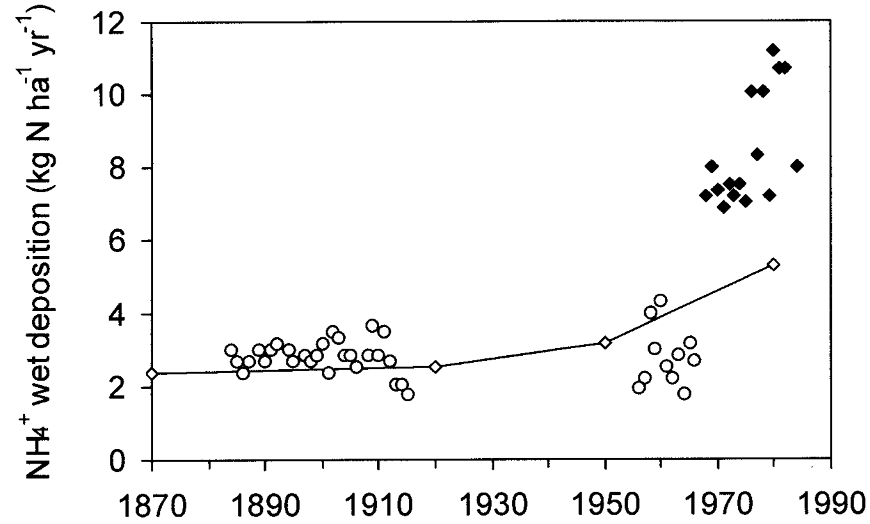

Figure 1. Long-term record of ammonium in precipitation at Rothamsted, England. Measurements from Goulding *et al.* (1986) are shown in filled symbols and those reported by Brimblecombe and Pitman (1980) in open symbols. The tie lines show the trend in modelled wet deposition of ammonium derived from Asman *et al.* (1988). After Sutton *et al.* (1993).

Such long-term datasets are rare, but exist in a few well-documented instances. The most famous long-term record of NH_x is for NH_4^+ in precipitation as recorded at Rothamsted, England since the 1850s. Combining published datasets, Sutton *et al.* (1993) compared the measurements with predictions of a European-scale atmospheric transport model of Asman *et al.* (1988) (Figure 1). Both measurements and model show a substantial increase, although the measurements increased more than the model. This was suggested to be due to effects of intensification, with larger emissions per animal in more recent years, which was not included in the model calculations. Although there appears to be some uncertainty in the data, with a questionable step change at around 1970, the broad consistency between model and measurements is strongly supportive of a long-term increase in NH_3 emissions.

A difficulty in very long-term measurements is the need to establish that any sampling bias is unchanged throughout the monitoring period. An approach that avoids this is the application of ice core sampling to examine temporal trends. By sampling with a consistent methodology through a single core, a robust estimate of the temporal trend in deposition to an ice pack may be established. Although the approach by definition measures total deposition, NH_x dry deposition to smooth frozen surfaces is expected to be small, so that the signal is dominated by wet deposition as NH_4^+ in snowfall. Work in Greenland has shown regular spikes in NH_4^+ deposition that couple to forest fire activity (Furher *et al.*, 1996). Although no overall increase in NH_4^+ deposition is seen for the last century, spring time

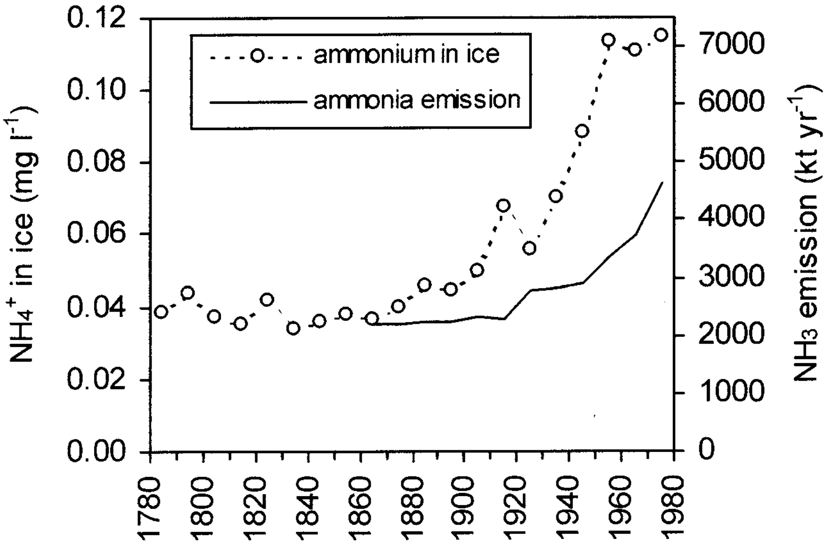

Figure 2. Record of 10 yr mean ammonium concentrations in ice over the period 1780–1980 from measurements Cole Gnifetti, Switzerland as compared with historical European NH_3 emissions estimated by Asman *et al.* (1988) (After Döscher *et al.*, 1996).

concentrations doubled over the last 40 yr, indicating a link to enhanced long-range transport of ammonium sulphate and nitrate. In a further example from an ice core study for a Swiss glacier, a dramatic increase in NH_4^+ was observed (Figure 2). Again the comparison with the modelled estimates of Asman *et al.* (1988) shows that the measurements increased more than the model, given that ice formation rates at this site may be assumed not to have changed substantially.

In both these historic examples, a substantial inter-annual and decadal variation can easily be seen. Hence the ability to see the trends is dependent on the availability of many decades of data for comparison.

3. Recent Case Studies: The Link between Agricultural Sector Activity and Atmospheric NH_x

As noted in the introduction, major changes in NH_3 emissions are believed to have occurred in eastern Europe, following the end of the communist system. From peak emissions in the mid-1980s, emissions across the whole of Europe are estimated to have declined by 19%. As shown in Figure 3, the changes in eastern Europe are by far the largest cause of this reduction; while the overall reduction for western European countries amounted to 3%, the overall reduction for 13 former communist states was 46%. Bearing in mind that the magnitude of this reduction is larger than most countries have agreed to under the Gothenburg Protocol, it ought to be possible to see the response in monitoring data from eastern Europe. Several

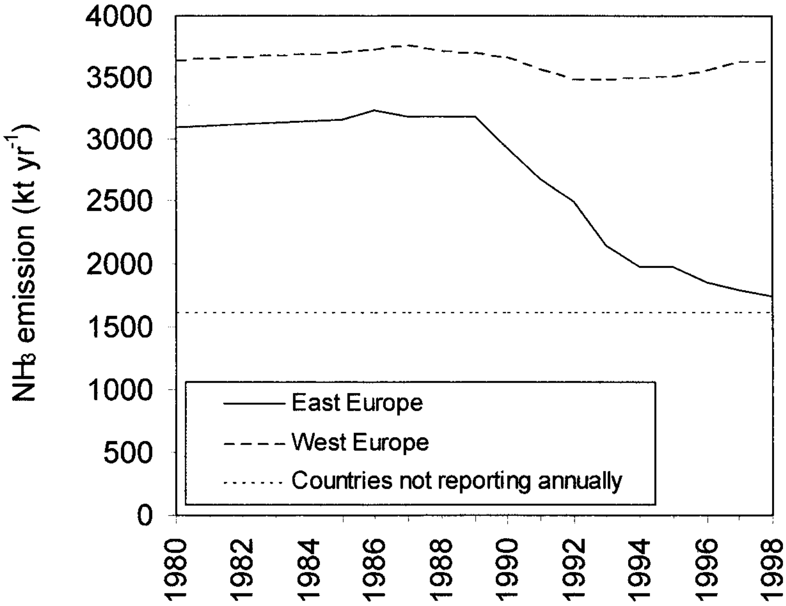

Figure 3. Changes in estimated European ammonia emissions for 1980–1998. Eastern countries include 13 former communist states, while western countries include the EU15 (excluding Luxembourg) plus Norway and Switzerland. 14 European countries including Turkey did not provide annual estimates. The estimates are from Vestreng and Støren (2000), with the separation between former East and West Germany from Strogies and Kallweit (1996).

examples are considered here including Hungary, the comparison of former East and West Germany, Slovakia and the former Soviet Union (FSU). The estimated changes in emissions for these and other example countries are shown in Figure 4.

3.1. HUNGARY

The analysis for Hungary of Horváth and Sutton (1998) considered data from the EMEP/GAW monitoring site K-puszta in the centre of the country. This site is unique in providing a long-term daily record of gaseous NH_3, aerosol NH_4^+ and wet deposition as NH_4^+. Despite an estimated reduction in national NH_3 emissions of 53%, no trend was seen in any of the components over the same period. The only change seen was a decrease in NH_4^+ in precipitation coupled to a decrease in sulphate aerosol, particularly at the beginning of the 1980s. This is relevant as it indicates, as will be seen in other examples below, an interaction with sulphur emissions and atmospheric chemistry. With decreasing SO_2 emissions, a reduced rate of ammonium sulphate aerosol formation is expected, resulting in less wet deposition of NH_4^+. A remaining question with this dataset is why aerosol NH_4^+ did not show a parallel trend. This may, for example, have been due to meteorological variations between wet and dry years masking any trend.

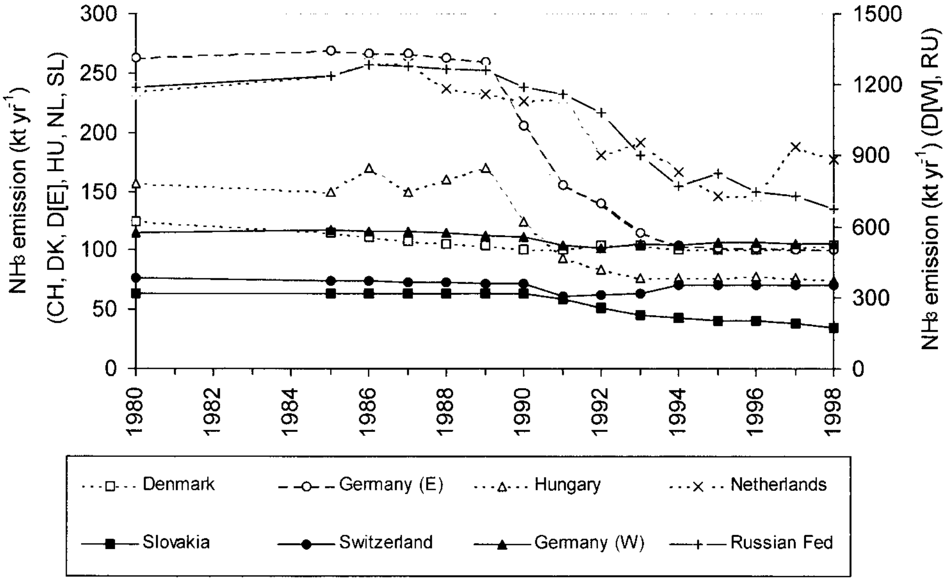

Figure 4. Changes in estimated ammonia emissions for example European countries considered in the document. For data sources see Figure 3.

While the earlier analysis considered NH_3 emissions only up to 1993, the more recent estimates shown in Figure 4 indicate that emissions have remained stable near 1993 levels. Data from the last 5 yr are now available to update the monitoring record at K-puszta, and these are shown in Figure 5. It is curious that both the NH_3 and NH_4^+ records continue to show no decrease from values in the mid-1980s. Ammonia shows spring and summer peaks, most probably linked to larger emissions in spring (Asman *et al.*, 1998), as well as a compensation point for exchange with vegetation, which is larger in warm conditions (Sutton *et al.*, 1995b). The seasonal trend is less pronounced for NH_4^+ aerosol, but the largest peaks occur from late autumn to early spring, probably associated with cooler, more humid conditions favouring aerosol formation. By contrast to the expected decrease, NH_3 during 1999 was in fact the highest on record at this site, and further analysis is needed to explore the reasons for this.

Figure 6 updates the record for NH_4^+ in wet deposition from that provided by Horváth and Sutton (1998) and also shows gaseous SO_2 and aerosol SO_4^{2-} concentrations. This shows a general decrease of NH_4^+ concentration in rain, but not in NH_4^+ wet deposition, with this difference due to variations between wet and dry years. By contrast, there was a large decrease in SO_2 concentrations after 1989, with annual mean SO_2 and NH_3 concentrations seen to be at least weakly anti-correlated (Figure 7). This would support the hypothesis that interactions with SO_2 mask the expected decrease in NH_3 concentrations, since with less SO_2 in recent years, the rate of conversion from NH_3 to NH_4^+ would be slower.

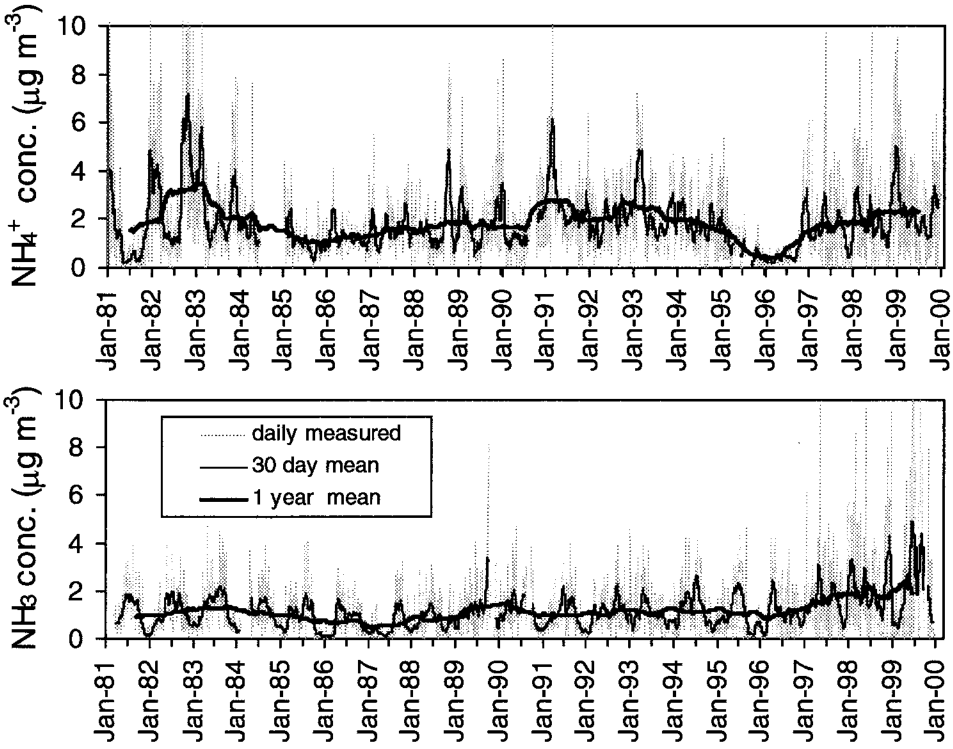

Figure 5. Long-term record of gaseous NH_3 and aerosol NH_4^+ at K-puszta in Hungary. Data for 1995–1999 extend the record reported by Horváth and Sutton (1998).

3.2. SLOVAKIA

Slovakia provides another example of an east European country where estimated NH_3 emissions decreased substantially (44% reduction, Figure 4). Precipitation chemistry is sampled as part of the EMEP network at Chopok and values for NH_4^+ concentrations in precipitation are shown in Figure 8. As with the precipitation NH_4^+ record at K-puszta, a slight decrease is observable, but at around 20% between 1990 and 1999, this is much less than the reduction in national emissions. In addition, it may be noted that the decline in NH_4^+ wet deposition is even smaller and less certain (data not shown).

3.3. FORMER EAST AND WEST GERMANY

One of the possible concerns with the data the Hungarian and Slovak sites is that much of the time winds arrive from western Europe, which would mask trends in NH_4^+ aerosol and wet deposition. In the case of K-puszta, such winds were calculated as occurring for 60% of the time (Labancz and Ferenczi, 1994). A further concern that was recognized by Horváth and Sutton (1998) was that averaging

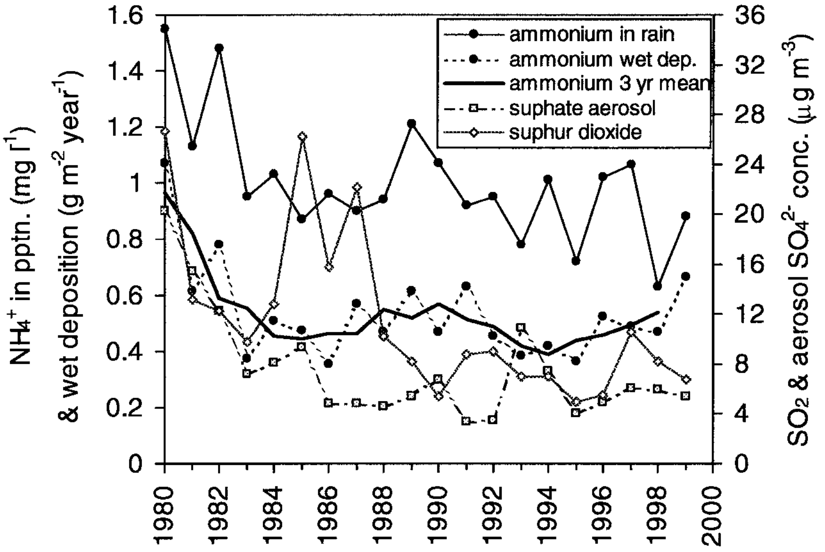

Figure 6. Inter-annual trends in NH_4^+ in rain and wet deposition at K-puszta in Hungary. The data are compared against trends in aerosol SO_4^{2-} and gaseous SO_2 concentrations. Data for 1995–1999 extend that reported by Horváth and Sutton (1998).

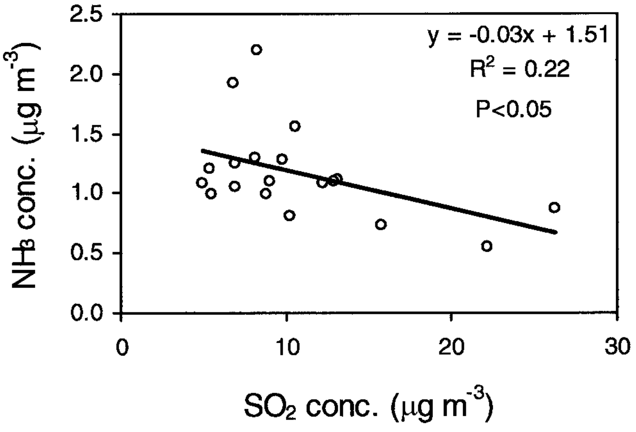

Figure 7. Annual mean NH_3 concentrations in relation to annual mean SO_2 concentrations for the years 1981–1999 from monitoring at K-puszta, Hungary.

out the effects of meteorological variations would require an assessment based on results from many sites. These points may be addressed by analyses comparing data from former East and West Germany.

At one site an analysis has been made to distinguish precipitation events depending on the air mass origins. Four-hourly measurements of wet deposition are made using wet-only collectors at the Seehausen measurement station, which is

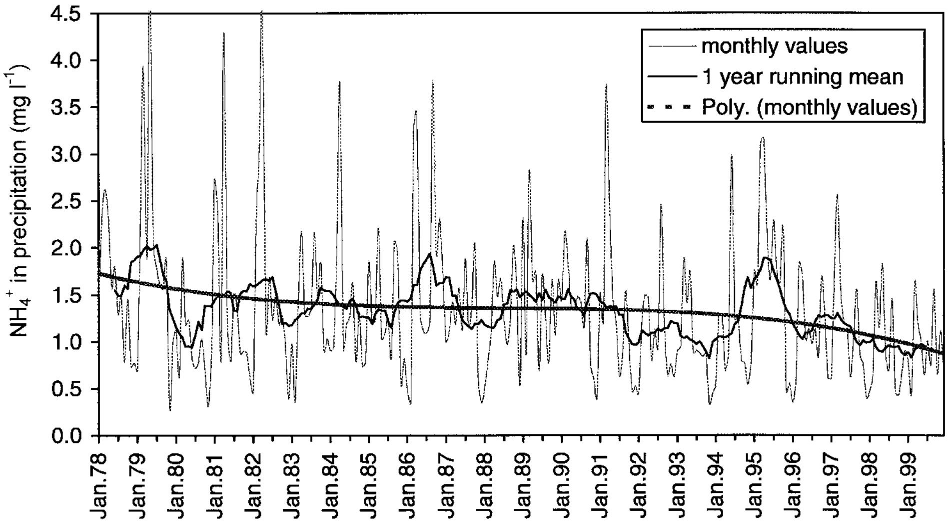

Figure 8. NH_4^+ in precipitation for 1978–1999 in the EMEP/GAW station at Chopok, Slovakia.

in a rural area near the border of former East and West Germany (Marquardt *et al.*, 1996; Brüggemann and Rolle, 1998; Acker *et al.*, 1998) (Figure 9). An entry sector analysis based on 48 hr back trajectories has been used by these authors to distinguish precipitation resulting from emissions in former East or West Germany. Given that only about 12% of the total precipitation comes from the east sector and 35% from the west sector, and that there are inter-annual differences in precipitation frequency for these sectors, the results are best viewed as precipitation NH_4^+ concentrations. However, it is relevant to compare the trend with wet deposition, since there are differences in precipitation amount between years. In both cases, the data are normalized by reference to the average for each sector, in order for any trend to be more clearly visualised (Figure 10). This analysis provides surprising results, as it shows NH_4^+ decreasing for both western and eastern sectors, which would not be expected on the basis of the national NH_3 emissions (Figure 4). The slight differences in reduction for west and east sectors between concentrations and deposition may be partly explained by the decrease in precipitation in the eastern sector for later years. However, this cannot explain why such a large reduction in western sectors should be expected, based on German NH_3 emissions. It may be noted that these uncertainties do not apply for NH_x alone, and that similar issues are raised in assessing the changes in wet deposition of SO_4^{2-} (Marquardt *et al.*, 1996; Acker *et al.*, 1998). However, the interactions with SO_2 may again provide a possible explanation to the NH_4^+ trends observed. In former East Germany SO_2 and NH_3 emissions declined in parallel, so that the ratio of national NH_3/SO_2 emissions (as mol/mol) remained at ~0.4 from 1983 to 1995, only increasing to >1 for 1998–1999. By contrast, in former West Germany legislation led to an increase in this

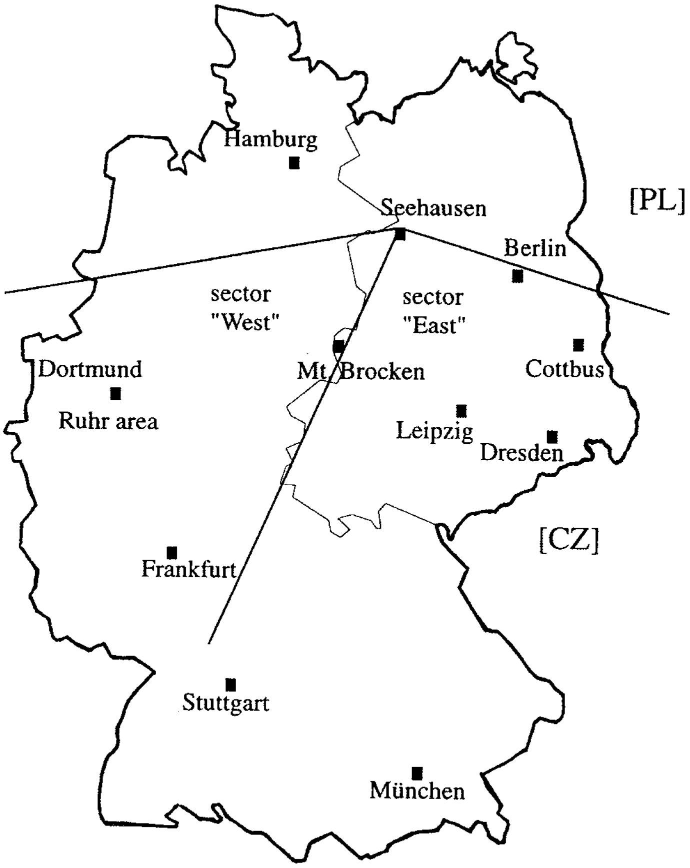

Figure 9. Map of Germany showing the Seehausen measurement station in relation to former east and west Germany (Acker *et al.*, 1999). The sectors ‘west’ and ‘east’ refer to the back trajectory analysis applied to distinguish precipitation events at Seehausen (Figure 8).

ratio from 0.8 in 1983 to >1 by 1987, with values relatively stable at >2 since 1989. Hence the larger decrease in precipitation NH_4^+ concentrations for the West sector in Figure 10 may in part be due to the larger decrease of SO_2 relative to NH_3 emissions, which would reduce the rate of NH_4^+ formation for the West sector.

The issue of spatial variability in precipitation adding uncertainty to temporal trends has been addressed in Germany through the use of a dense precipitation chemistry monitoring network and mapping of the results (Gauger and Anshelm, 2000). On the basis of this analysis, estimates of average wet deposition to each of the German länder may be made. The averages for länder in former East and West Germany are contrasted in Figure 11. This supports a stronger decrease in wet deposition of NH_4^+ in the east länder than the west, but again the difference is much smaller than would might be expected based on the differences in estimated NH_3 emissions. For the period 1987–1995, an emission reduction of 8% in the west

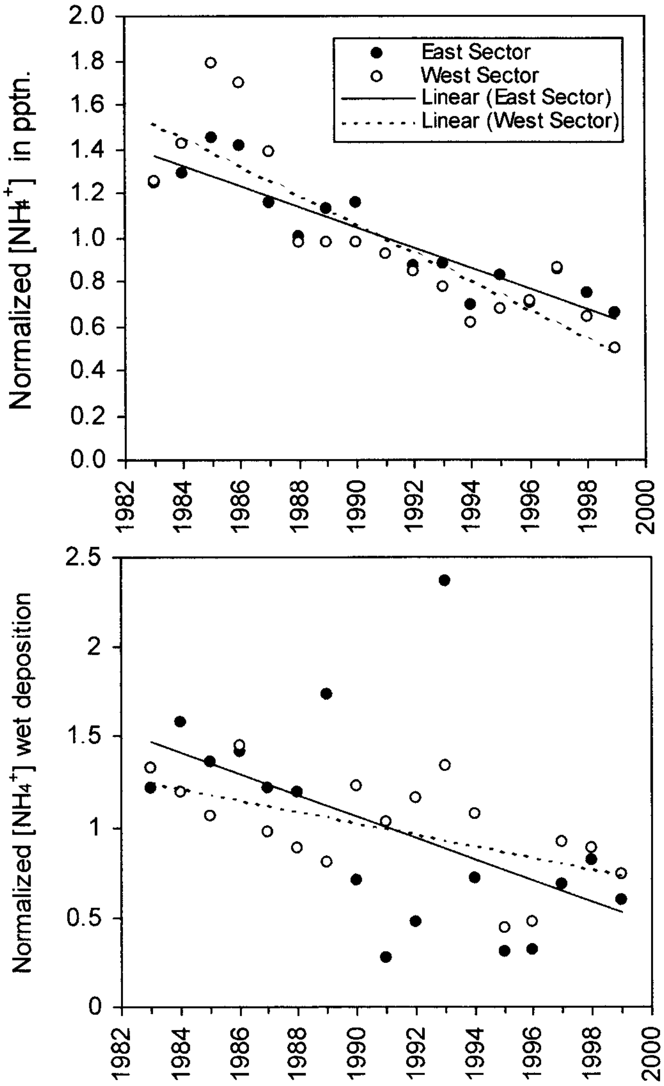

Figure 10. Comparison of NH_4^+ in precipitation for westerly and easterly trajectories arriving at Seehausen near the border of former East and West Germany (Figure 9). The decrease in NH_4^+ is expected for the east sector, but the decrease is less than expected based on emissions changes (Figure 4), while the decrease for the west sector is more than expected. Values are normalized to the mean for the period 1983–1999 (and therefore dimensionless). The values for the East Sector may also be considered as two populations, pre and post-1990, and a step change may be discerned in the NH_4^+ concentration.

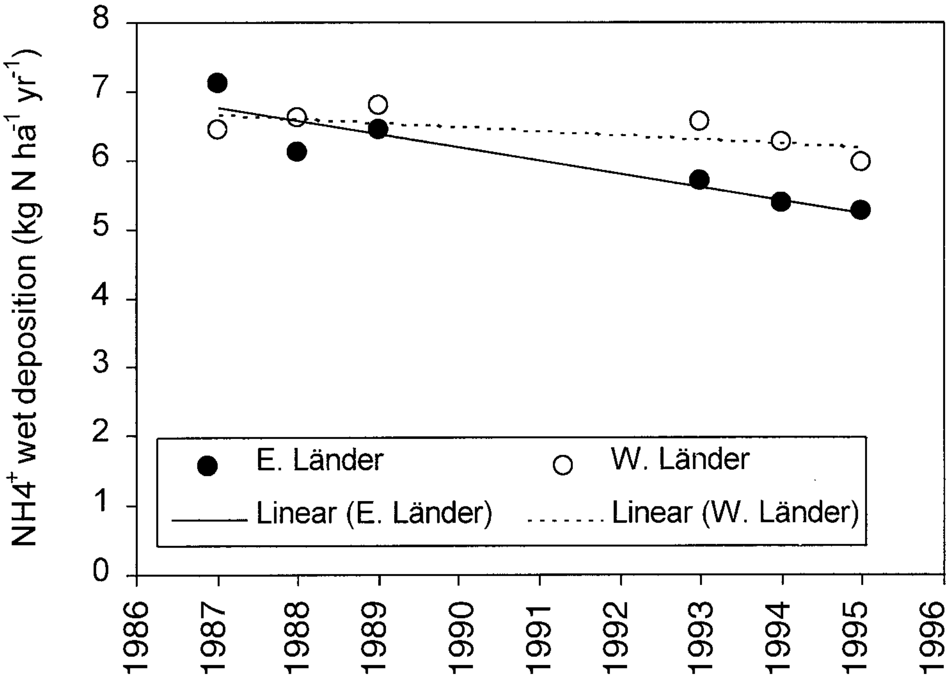

Figure 11. Comparison of NH_4^+ wet deposition for the Länder of former East Germany (former GDR and Berlin) and West Germany. Values are averages of mapped estimates for the 10 former western and 6 former eastern Länder (Gauger and Anshelm, 2000).

länder is matched by a 7% reduction in wet deposition of NH_4^+. By contrast, in the east länder a 62% reduction in emission is matched by only a 23% reduction in NH_4^+ wet deposition.

3.4. RUSSIA

As shown in Figure 4, a major reduction (46%) in NH_3 emissions in the former Soviet Union is estimated to have occurred between 1989 and 1998. A recent independent analysis by Ryaboshapko (2001) has confirmed the trend in the official estimates of Figure 4, in general providing approximately 40% larger emissions due to different emission factors and consideration of additional sources. With such a large territory, and with other neighbouring countries to the west also experiencing a reduction in NH_3 emissions, decreased levels of NH_x in monitoring data ought to have occurred. The most robust dataset for assessment of long-term trends in Russia is the precipitation chemistry record, and this has recently been assessed by Paramonov *et al.* (1999). Particular attention was given in this analysis to data quality control to establish the reliability of the results. The most relevant sites to consider are those in the European Territory of the FSU, since this is where NH_3 emission reductions may be most likely. Long-term data are available for

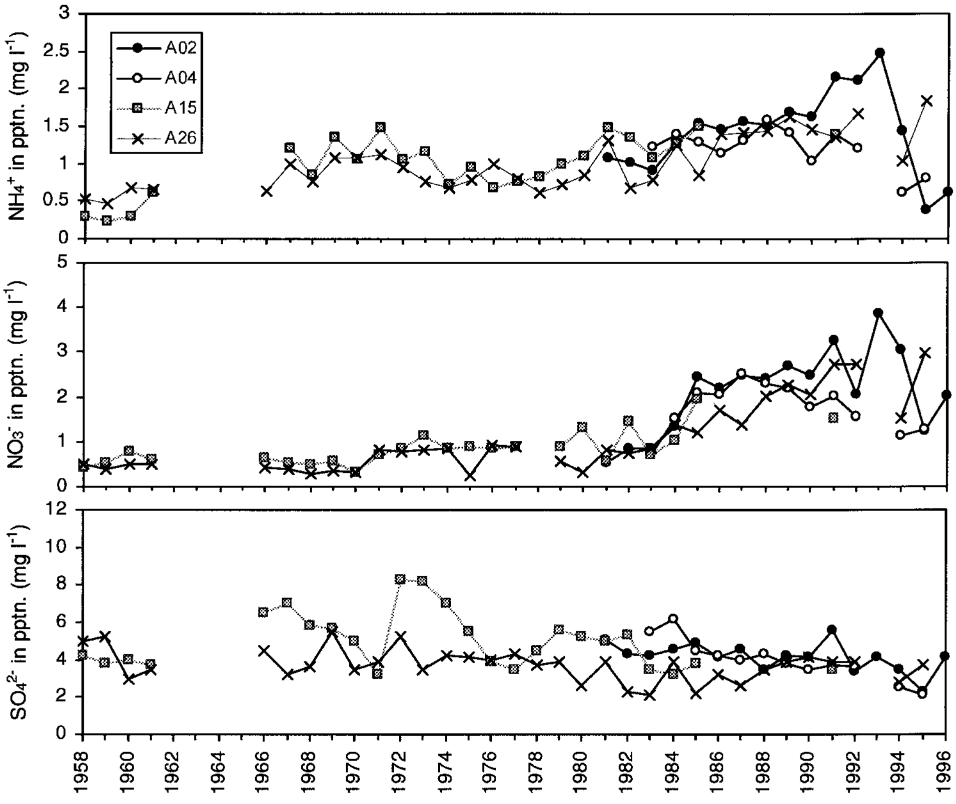

Figure 12. Record of NH_4^+, NO_3^- and SO_4^{2-} precipitation concentrations for sites in the European Territory of the Former Soviet Union. A02: Berezina Biosphere Reserve (BR); A04: Oka-Terrase BR; A15: Central Forest BR; A26: Syktyvkar-1.

four sites Berezina Biosphere Reserve (BR), Oka-Terrase BR, Central Forest BR and Syktyvkar-1, which are each sites in the WMO Global Atmospheric Watch (Figure 12).

Reductions in NH_4^+ concentrations in precipitation since 1993 may be seen for two of the sites (Berezina BR and Oka-Terrase BR), but there is much scatter in the signal, and there is no discernable reduction at Syktyvkar-1. This may be related to the distribution of NH_3 emissions, with Syktyvkar-1 being the most remote, northerly site (61.6°N, 50.5°E), well away from the main agricultural areas (Galperin and Sofiev, 1998). Taking an average of these 3 sites, precipitation NH_4^+ decreased by approximately 40% between 1989 and 1995, which is consistent with the national reduction in NH_3 emissions.

This consistency could be seen to suggest that with a sufficiently large territory surrounding the Russian monitoring sites, the precipitation NH_4^+ corresponds to NH_3 emissions. However, some caution is needed. Figure 12 also shows precipitation NO_3^- and SO_4^{2-} with these decreasing on average by 25–30% for the same period. Hence, part of the change may be a result of an altered atmospheric transport distance of NH_x linked to changing NO_x and SO_2 emissions.

3.5. SWITZERLAND

Switzerland provides a contrast to the eastern European countries, since NH_3 emissions are estimated to have been largely stable over the past decade (Figure 4). Ammonium concentrations in precipitation have been reported for 1985 to 1998 for two sites Dübendorf and Payerne (BUWAL, 1999), and these are shown alongside the results for NO_3^- and SO_4^{2-} in Figure 13. Overall, a 20% decline in NH_4^+ concentrations may be seen for the period (based on linear regression). This was matched by a 27% reduction in NO_3^- and a 58% reduction in SO_4^{2-}. With such large changes in SO_4^{2-} and NO_3^- concentrations, it becomes difficult to see whether the decrease in NH_4^+ is a result of a reduced atmospheric lifetime of NH_x (leading to less transport with SO_4^{2-} and NO_3^-) or actually the result of Europe wide reductions in NH_3 emissions.

3.6. NORTH CAROLINA

While most interest in NH_x trends has focused in Europe, North America provides a dramatic example of increases in NH_3 emissions and atmospheric NH_4^+ levels. In the State of North Carolina, a rapidly expanding pig sector has given rise to concerns about N deposition to sensitive coastal ecosystems (Aneja *et al.*, 2000; Walker *et al.*, 2000). According to the U.S. EPA National Emissions Trends inventory, NH_3 emission in 1996 for coastal North Carolina was 117 ktons N, with 74 ktons of this arising from pigs. Overall, pigs are estimated to account for 77% of the total livestock NH_3 emission and 63% of the total emissions.

Figure 14 shows the measured record of NH_4^+ in precipitation and wet deposition at the NADP measurement site (NC35) in Sampson County, one of the most affected areas. A clear trend in both precipitation concentrations and wet deposition is seen for this site, which is not surprising given that pig emissions increased by a factor of 7 between 1985 and 1997. This would equate to at least a doubling of total NH_3 emissions, even if all other sources remained constant. This is consistent with the increase in measured NH_4^+, which roughly doubled over the period.

4. Recent Case Studies: The Link between NH_3 Emission Abatement and Atmospheric NH_x

4.1. THE NETHERLANDS

In The Netherlands, the lack of a detectable reduction in NH_3 concentrations following the implementation of abatement measures in 1993 has stimulated a great deal of activity to explain this feature (e.g. Erisman *et al.*, 1998, 2000; Boxman

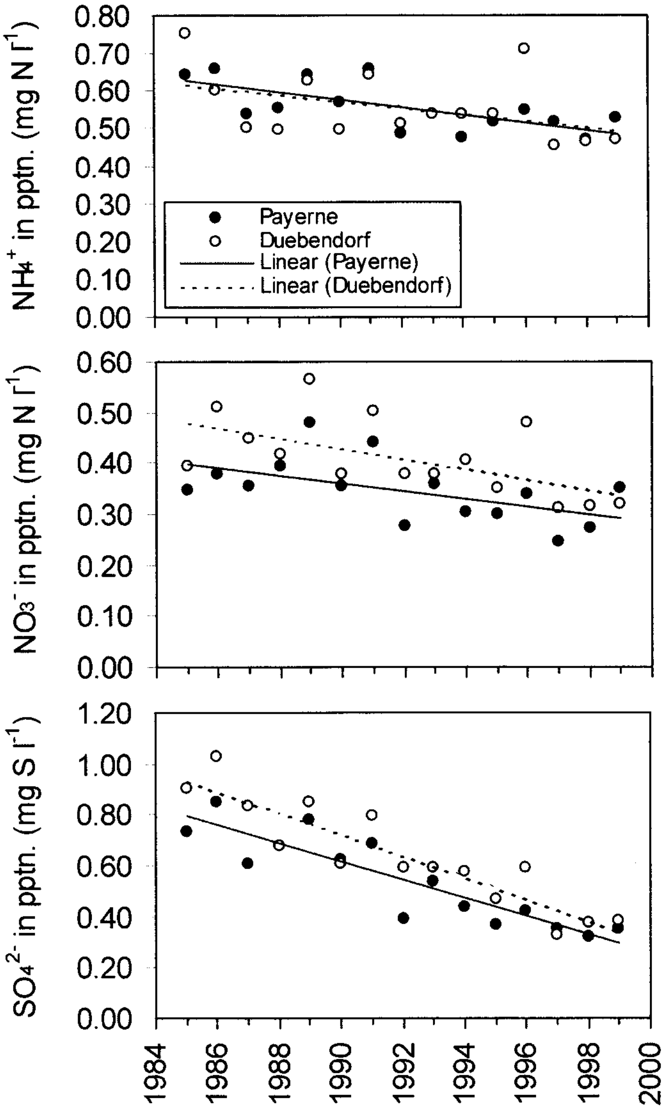

Figure 13. Record of ammonium, nitrate and sulphate concentrations in wet deposition at two Swiss sites. Trend lines are fitted using linear regression.

et al., 1998; Van Jaarsveld *et al.*, 2000). This missing signal has been referred to as the 'ammonia gap'. The example is tantalising since, although in terms of NH_3 abatement effort the estimated 35% decrease is large, this is a rather modest change compared with the major reductions estimated in other parts of Europe. As indicated by the scatter in datasets for other countries, it may be difficult to detect 35% change with 5–7 yr of monitoring for a small country such as The Netherlands,

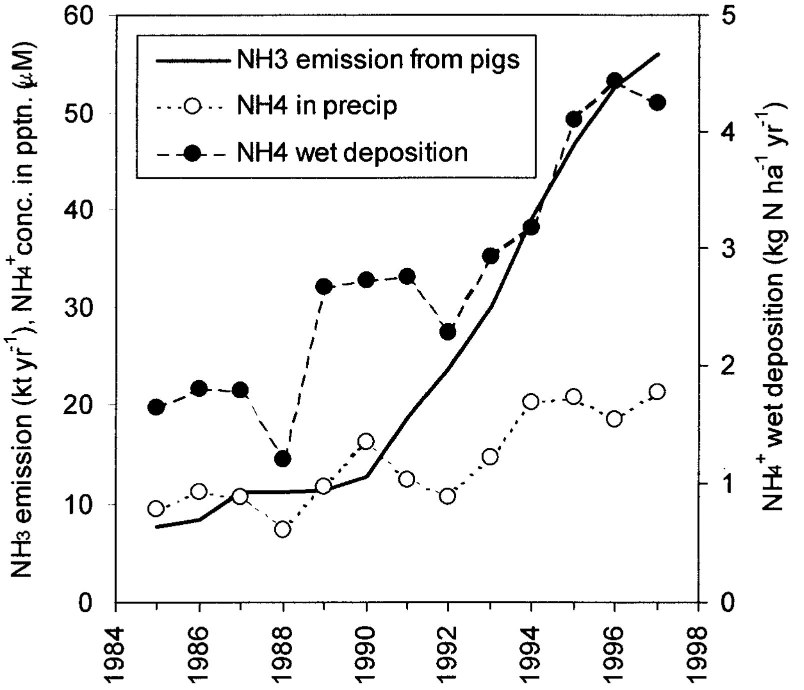

Figure 14. Comparison of NH_3 emissions in coastal North Carolina, U.S.A., against NH_4^+ precipitation concentrations and wet deposition at the NADP monitoring site NC35 in Sampson County (from Aneja *et al.*, 2000). Emissions are for the 6 counties of North Carolina surrounding NC35. This case study provides a good example of the limits to such comparisons between NH_3 emissions and wet deposition. Since both the emission and deposition clearly changed, the level of scatter in the measurement data can help indicate what would be the minimum detectable change or time-period to detect change. Walker *et al.* (2000) found that the trend for monthly volume weighted NH_4^+ concentrations at NC35 was not significant for the period 1983–1990, but was significant for 1990–1997. Hence, even for such large changes as observed here, a single site record of less than 5–7 yr becomes increasingly less able to detect changes. Conversely, even with 10 yr data at the one site, it would have been difficult to detect a change of less than 30–50%.

given the interactions with meteorology, other pollutant emissions/deposition and long-range transport.

The availability of monitoring data in The Netherlands is indicated in Figure 15, which shows normalized trends in NH_3, aerosol NH_4^+ and NH_4^+ wet deposition. The number of sites included in this comparison is shown at the top of the figure. The statistics for wet deposition are quite reasonable with 13–21 monitoring locations. Even so, with emission abatement only being applied since 1993, it is difficult to quantify precisely the reduction. As discussed above, NH_3 shows no decrease since 1993. Given the high spatial variability of NH_3 concentrations (e.g. Sutton *et al.*, 1994; Duyzer *et al.*, 2000) and few sites, it is not possible to generalize trends before 1992. For NH_4^+ aerosol, there appears to be a clear decrease in con-

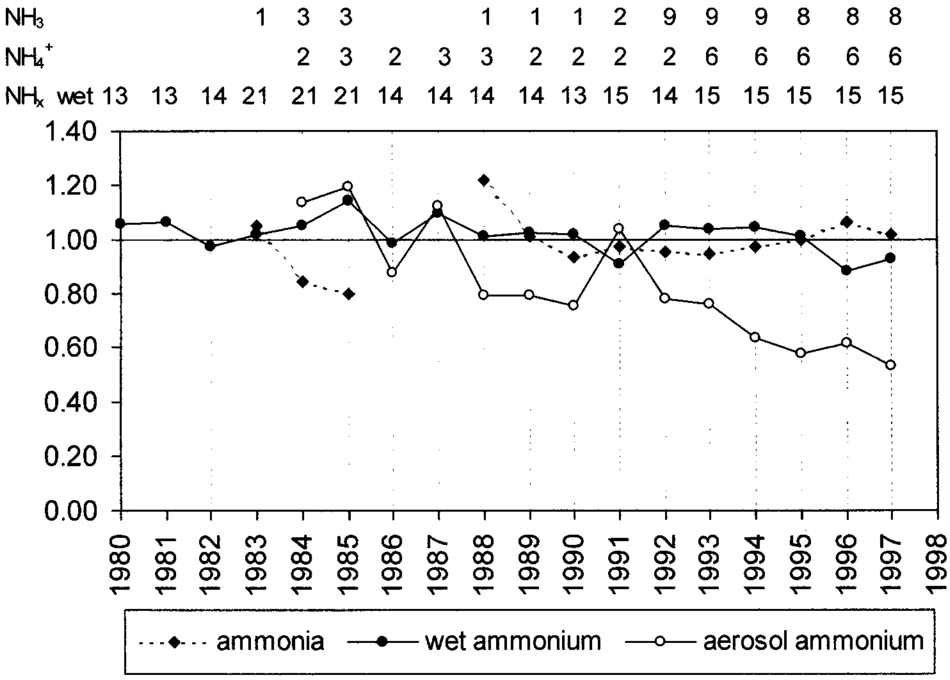

Figure 15. Averages of measured NH_3 concentrations, wet deposition of NH_x and NH_4^+ concentrations from monitoring in The Netherlands, normalized to highlight temporal trends. The number of sampling stations for different components and years is indicated at the top of the figure (from Van Jaarsveld *et al.*, 2000).

centrations of 50% between 1984 and 1997, or 30% between 1993 and 1997. As a reaction product in the atmosphere aerosol NH_4^+ concentrations are more spatially homogeneous, and even though most certainty should be attributed to the data for 1993–1997, the earlier data are expected still to be useful. This is supported by Figure 16, which shows the record of NH_4^+ at the different sites.

Application of the OPS model suggests that, had the expected 35% abatement really been achieved, then the NH_3 concentrations would have decreased more rapidly than the observations (Erisman *et al.*, 1998; Van Jaarsveld *et al.*, 2000). In Figure 17a, the NH_3 concentration record is compared with the OPS model estimates. The model estimates (assuming full achievement of the abatement target) are substantially less than the measurements. While the overall difference between the measurements and model may also be partly due to model uncertainties, it is notable that for the period 1993–1997, modelled NH_3 decreased by $\approx$5%, while measured NH_3 *increased* by $\approx$5%.

In Figure 17, the model estimates for NH_4^+ wet deposition are smaller than the measurements for the whole period, which must be due to model uncertainty. However, while the modelled estimates decreased by 20% between 1993 and 1997,

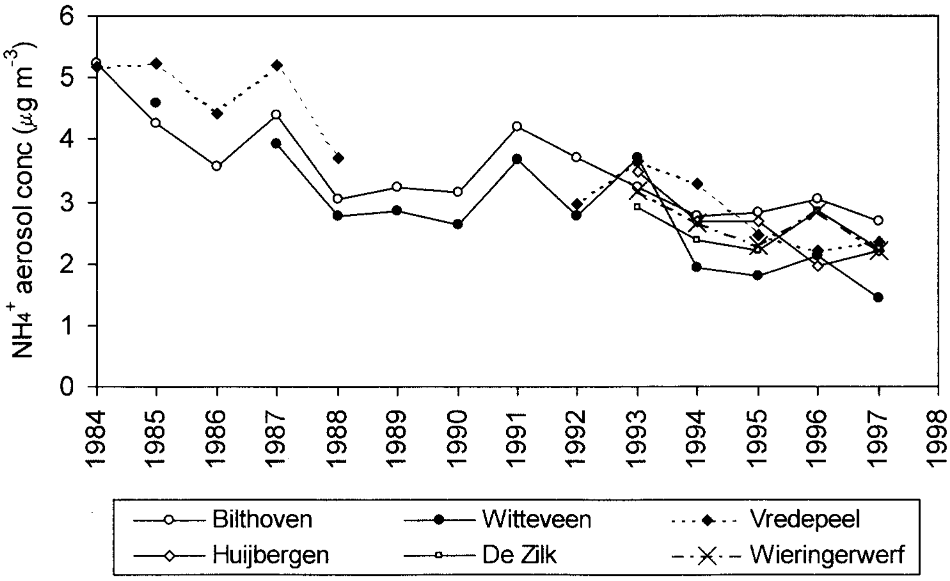

Figure 16. Record of NH_4^+ aerosol concentrations in The Netherlands (Erisman *et al.*, 1998; Van Jaarsveld *et al.*, 2000).

the measurements only decreased by 10%. By contrast, NH_4^+ aerosol decreased more in the measurements than in the model, by 29% compared with 14% for the same period. Although, there is less certainty in the earlier aerosol averages, this difference is consistent with the change over the period 1984–1998, with measured NH_4^+ aerosol decreasing by 56% and modelled by 44%.

Part of the reason for the differences between the model and the measurements may be explained by parallel changes in SO_2 and NO_x emissions over the same period. Van Jaarsveld *et al.* (2000) assessed this by comparing modelled NH_x results with those estimated if SO_2 and NO_x emissions had remained at 1984 levels. As shown in Figure 18, the effect in the model of less SO_2 and NO_x is to increase NH_3 concentrations in The Netherlands and decrease NH_4^+ wet deposition. The reason for this (as parametrized in the model), is that the net rate of ammonium sulphate and nitrate aerosol formation is slower, thereby giving a longer residence time of gaseous NH_3 in the atmosphere. By contrast, as The Netherlands is a source region, and wet deposition is dominated by scavenging of NH_4^+ aerosol, there is less NH_4^+ wet deposition in The Netherlands. This implies a shift from wet deposition to dry deposition, which may decrease the overall atmospheric residence time of NH_x, depending on the parametrization of dry deposition. On the basis of this effect, and less-than-average rain during the period, Van Jaarsveld *et al.* (2000) concluded that part of the ammonia gap could be explained. However, this did not explain the whole difference and it was concluded that only 45–70% of the foreseen reduction in emissions had been achieved (i.e. a national emission reduction of 16–25%).

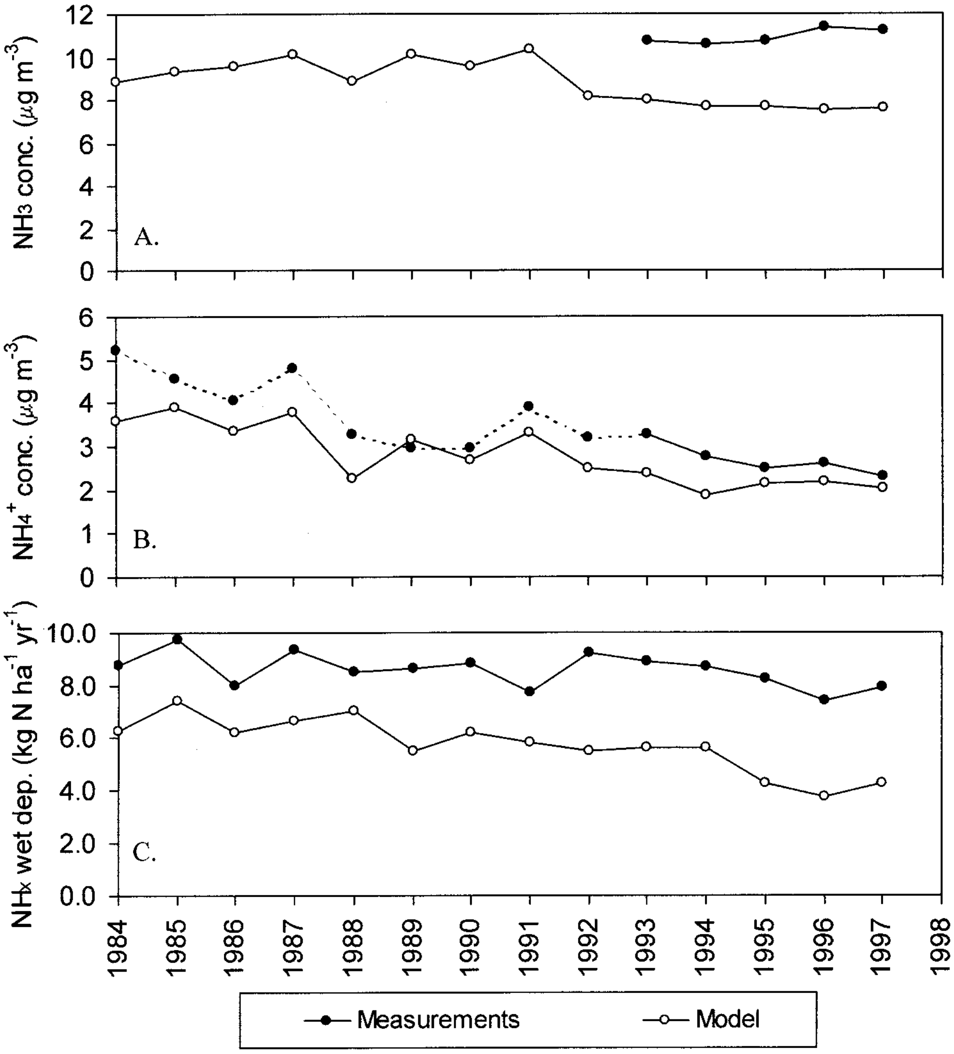

Figure 17. Comparison of modelled and measured NH_x for The Netherlands. Measurements are as summarized in Figure 15. Model results are from the OPS Model (after Van Jaarsveld *et al.*, 2000). In the case of NH_4^+, measured values for 1984–1992 show consistency even though there are few sites (Figure 16); these values are indicated by a dotted line. Because of the high spatial variability and few measurement sites, it is not possible to assess the trend in gaseous NH_3 before 1993.

Two further contributions to the debate on the ammonia gap should be mentioned, which both might appear to indicate that substantial reductions in NH_3 emission have occurred. In an analysis of NH_3 concentrations in a source area of The Netherlands (Gelderse Vallei), Duyzer *et al.* (2001) observed a 25% decrease in NH_3 concentrations between 1996 and 1998. While it could be speculated that this is a consequence of abatement, it should be noted that the abatement policy

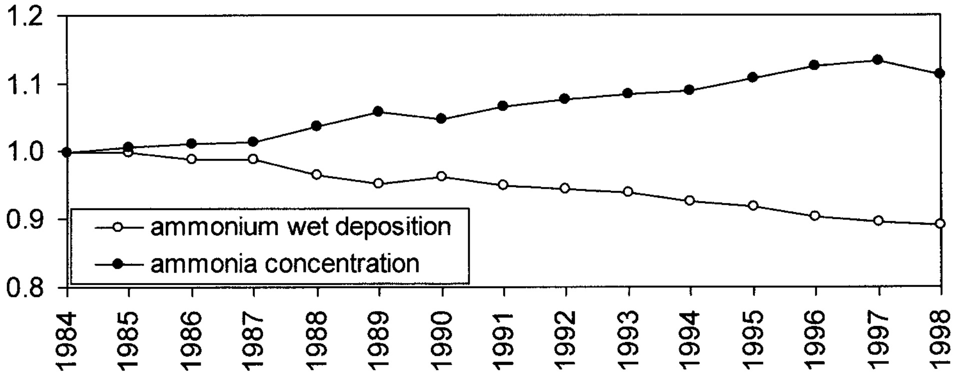

Figure 18. The effect of changing SO_2 and NO_x emissions on NH_3 concentrations and the contribution of NH_4^+ scavenging to wet deposition, as assessed using the OPS model (Van Jaarsveld *et al.*, 2000). The figure shows the ratio of modelled component calculated with normal SO_2 and NO_x emissions to that calculated with emissions of SO_2 and NO_x at 1984 levels. The model indicates that a decrease in SO_2 and NO_x leads to an increase in NH_3 and a decrease in the NH_4^+ scavenging contribution to wet deposition.

for The Netherlands did not change between these years. By contrast, Duyzer *et al.* explain the effect by inter-annual differences in meteorological conditions (Figure 19). As noted above, 1996 and 1997 were very dry years; by contrast, 1998 was very wet. If the OPS model is run for this area using 1996 emissions, much of the decrease in measured NH_3 concentrations can be explained by the increase in precipitation, leading to faster removal of NH_3 from the atmosphere. It is further possible that the unexplained residual is due to the links between meteorology and emissions. For example, manure spreading NH_3 emissions may be smaller in wet years due to the solubility of NH_3, as well as limitations to field access for manure spreading (Duyzer *et al.*, 2001).

In a second example, measurements of the NH_4^+ forest throughfall flux at IJsselstein showed a 45% reduction between 1993 and 1997 (Boxman *et al.*, 1998). Although throughfall is not the same as total deposition, due to canopy exchange processes, this suggests that NH_x deposition did indeed decrease over the period. However, as with the results at Gelderse Valley, it is difficult to use this to imply that NH_3 emissions also decreased, since the reduction parallels the decrease in precipitation over the period (Erisman *et al.*, 1998, 2000). Nevertheless, even accounting for the change in the reduction in precipitation, NH_4^+ is seen to decrease more than the reduction of the sum of the SO_4^{2-} and NO_3^- anions (Figure 20). This suggests that relatively less NH_3 is deposited to the canopy than SO_x and NO_y for the later years. This could reflect a reduced deposition velocity of NH_3, as reduced acid concentrations limit the potential for co-deposition (e.g. Adema *et al.*, 1986; Sutton *et al.*, 1994; Erisman *et al.*, 2000). Both a reduction in dry

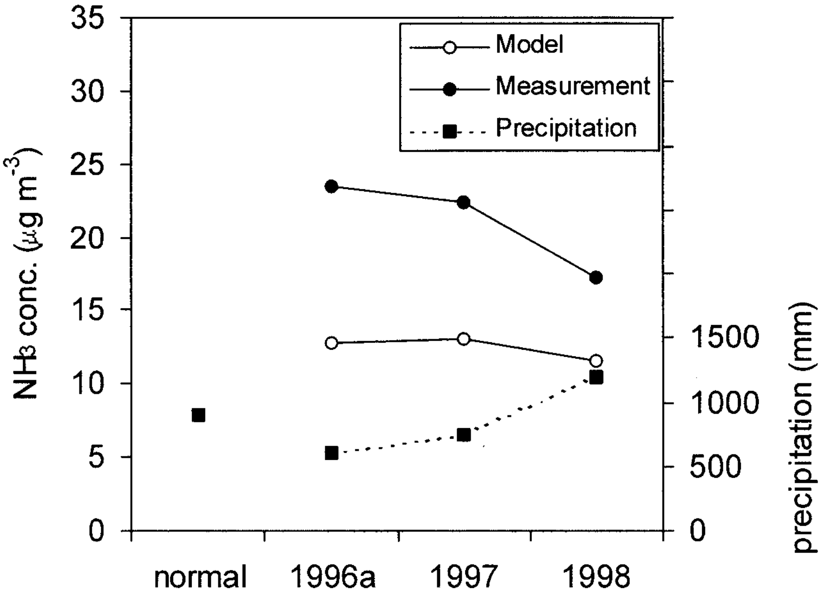

Figure 19. Interaction of meteorological conditions with NH_3 trends in a source area of The Netherlands (Gelderse Vallei) (from Duyzer *et al.*, 2000). Emissions in this area should be the same each year, apart from interactions with meteorological conditions. The model estimates are from the OPS model, run with emissions for 1996; the decrease in the modelled estimate for 1998 is due to this being a wetter year. If the model predictions are accurate, then NH_3 emissions are under estimated. The larger decrease in the model than the measurements for 1997–1998 may be attributed to wet conditions in 1998 limiting the opportunity for manure spreading. 1996a refers to the months of February to December.

deposition velocity and a decrease in precipitation would lead to an increase in NH_3 concentrations.

4.2. DENMARK

Extensive monitoring of NH_x is made in Denmark and the data permit the assessment of trends between 1989 and 1999 (Figures 21 and 22). Substantial decreases may be seen in NH_4^+ aerosols, while there are also some significant decreases for NH_4^+ wet deposition and NH_3 concentrations. The overall changes are summarized in Table I, and show that, on average, NH_3 decreased by 6%, NH_4^+ aerosol decreased by 34% and NH_4^+ wet deposition decreased by 20%. The relative balance of these changes is similar to The Netherlands, with the largest change for aerosol NH_4^+ and the smallest for NH_3. If these changes for NH_4^+ are compared with the general trend for The Netherlands over 10 yr, they are seen to be rather similar, with $\approx$30% decrease and $\approx$15% decrease for aerosol NH_4^+ and NH_4^+ wet deposition in The Netherlands, respectively.

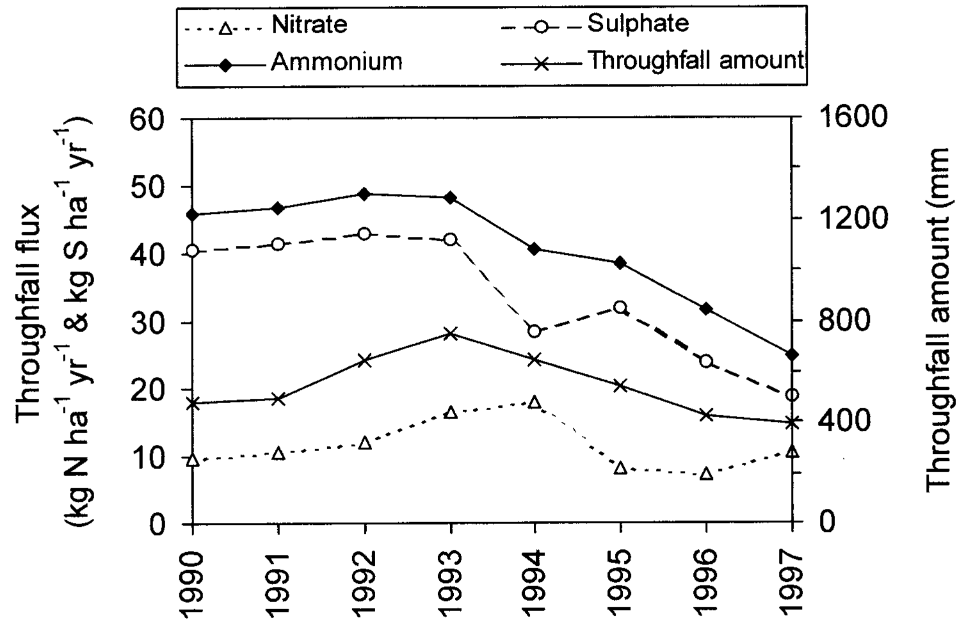

Figure 20. Record of ammonium, nitrate and sulphate flux in forest throughfall compared with amount of throughfall at IJsselstein in The Netherlands (from Boxman *et al.*, 1998; Erisman *et al.*, 2000).

TABLE I

Relative trends in NH_x concentrations and deposition in Denmark 1989–1999 (% change of the values for 1999 compared with 1989)

Component	NH_3 in air	NH_4^+ in air	NH_x wet deposition[a]
Ulborg	*14*	**–32**	*–22*
Tange/Sepstrup Sande[b]	**–26**	**–33**	*–12*
Lindet	c	**–35**	*–21*
Anholt	*–10*	**–35**	*–19*
Frederiksborg	*–13*	**–38**	**–27**
Keldsnor	*5*	**–32**	*–17*[d]

Bold values are significant at a level >99%; *Italic* values are not significant to this level. Negative values indicate reductions.

[a] The amount of precipitation has also increased during this period, but not significantly.

[b] NH_3 and NH_4^+ in air taken from Tange. NH_x wet deposition taken from Sepstrup Sande. The distance between these stations is about 30 km.

[c] Not taken into account because sampling height was changed.

[d] Less certain precipitation data at this site makes this value doubtful.

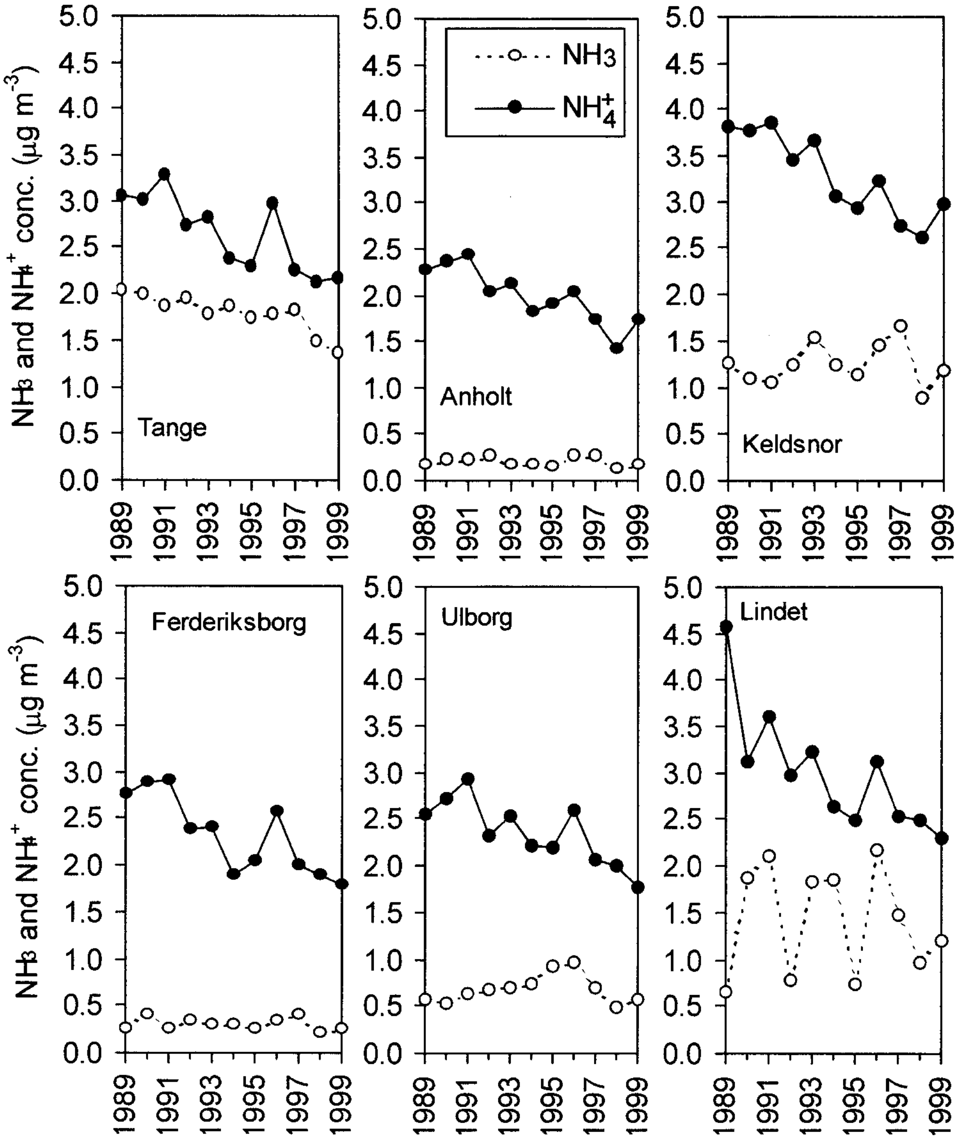

Figure 21. Record of gaseous NH_3 and aerosol NH_4^+ concentrations at selected monitoring sites in Denmark.

What is currently uncertain in Denmark is the extent to which NH_3 emissions have reduced. The values shown in Figure 4, are from officially submitted figures (Vestreng and Støren, 2000), but these may be inaccurate. For example, more recent values published by Andersen *et al.* (2001) indicate that NH_3 emissions have reduced by 27% over the period 1989–1999.

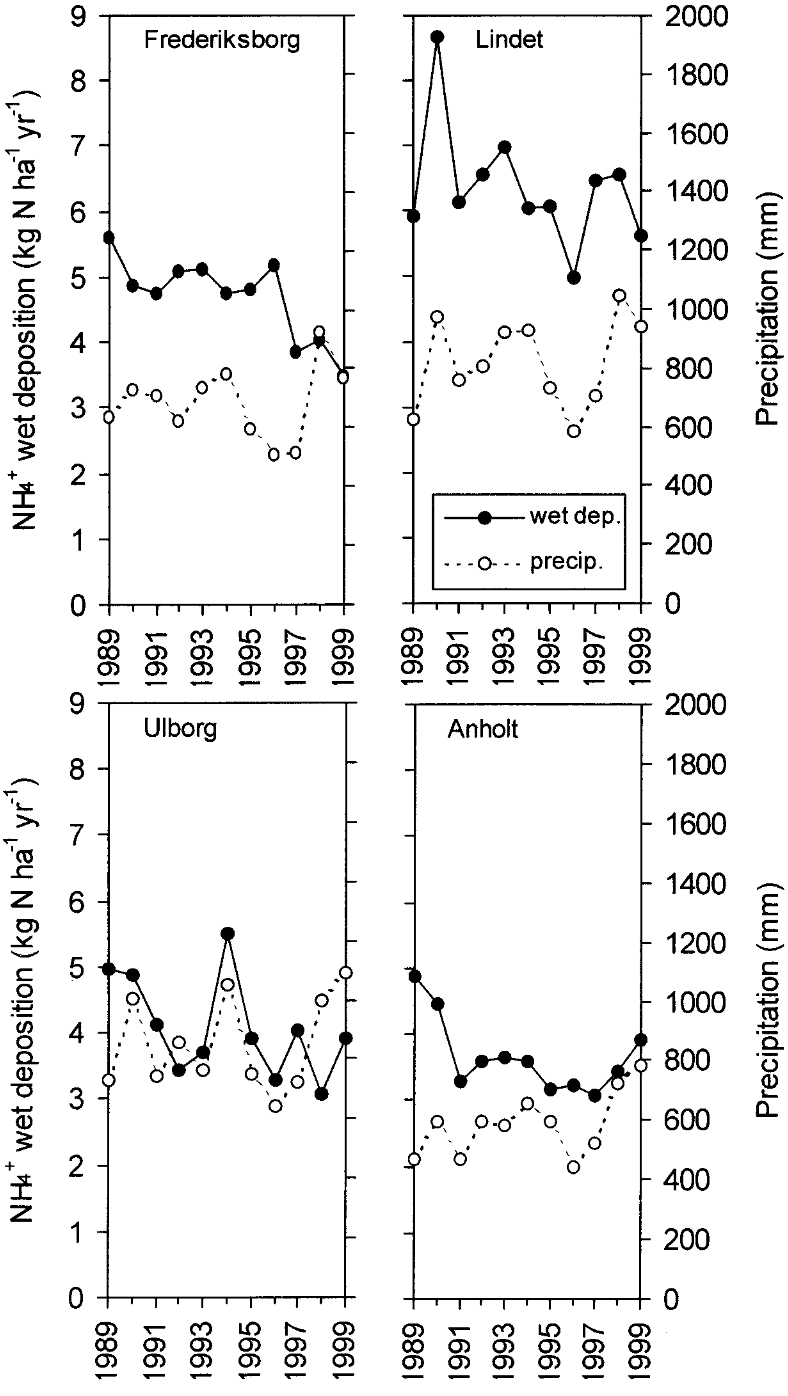

Figure 22. Record of NH_4^+ in wet deposition at selected monitoring sites in Denmark. The decreases are paralleled by reductions in sulphate and nitrate deposition (data not shown).

As with the example of former East and West Germany, it is of interest to establish the extent to which the trends are due to domestic or foreign changes in NH_3 emissions. To address this, a model analysis using the TREND model (Asman and Van Jaarsveld, 1992) was made for several of the Danish monitoring stations. The modelling applied the emission factors of Asman (1992a, b) for Europe and Denmark, respectively. Danish emissions were modelled on a 5 km × 5 km grid and foreign emissions on a 75 km × 75 km grid. These calculations give an indication of the relative contribution of Denmark to the concentrations and wet deposition, using the stations of Anholt, Tange and Keldsnor as examples. Anholt is a small island 40 km from the nearest land in the Kattegat Sea, between Denmark, Sweden and Norway; Tange is an inland site in mid Jutland (56.35°N, 9.60°E), while Sepstrup Sande/Keldsnor is a coastal site at the south tip of the island of Langeland (54.73°N, 10.73°E). Table II shows the results of this analysis.

It is clear from the analysis that the overwhelming part of the NH_3 concentration at all stations originates from Danish sources. This reflects the nature of NH_3 as a primary pollutant affected strongly by nearby ground level sources. By contrast, there are remarkable differences between the contribution of Danish sources to the NH_4^+ aerosol concentration at the three stations. At Tange the Danish sources dominate (64% from Danish sources) and at Keldsnor the foreign sources dominate (only 21% from Danish sources). Parallel differences are observed for the NH_x wet deposition. These differences give the possibility in principle to see whether an observed trend in the monitoring results is caused by a trend in Danish or a trend in foreign emissions (or both).

Applying these findings to the trends reported in Table I shows that the measured decrease in NH_3 concentrations at Tange are due almost entirely to changes in Danish NH_3 emissions. Similarly, although foreign sources contribute to 36% of aerosol NH_4^+ and wet deposition at Tange, most of the change appears likely to be due to Danish changes in emissions. By contrast, much of the decrease in NH_4^+ aerosol and wet deposition at Anholt and Kelsnor, are almost certainly due to changes in foreign NH_3 emissions or long-range transport.

5. Discussion

The series of case studies presented here illustrate the complexities of the atmospheric behaviour of NH_x and how this impacts on the link between emissions, concentrations and deposition. It can be seen from the most extreme cases, such as the changes over the last two centuries (Figures 1 and 2) and at Sampson County in North Carolina (Figure 14), that there is clearly a correlation between estimated changes in NH_3 emissions and wet deposition of NH_x. However, it is equally apparent that both NH_3 emissions and deposition couple strongly to meteorological conditions, so that there is substantial inter-annual variability, which makes detection of trends from periods of a few years rather difficult. On to this may be

TABLE II

Model estimates of the contribution of domestic and foreign sources to NH_x at three Danish measurent stations. Values were calculated with the TREND model (see text)

	Sources		
	From DK	From all	% from DK
Station Anholt			
NH_3 conc. (μg NH_3 m^{-3})	0.113	0.130	87
NH_4 conc. (μg NH_4 m^{-3})	0.673	1.602	42
Contr. NH_3 to NH_x wet dep. (mol ha^{-1} yr^{-1})	19.60	20.92	94
Contr. NH_4^+ to NH_x wet dep. (mol ha^{-1} yr^{-1})	74.22	184.77	40
NH_x wet dep. (mol ha^{-1} yr^{-1})	93.82	205.69	46
Station Tange			
NH_3 conc. (μg NH_3 m^{-3})	3.246	3.251	100
NH_4^+ conc. (μg NH_4^+ m^{-3})	1.633	2.568	64
Contr. NH_3 to NH_x wet dep. (mol ha^{-1} yr^{-1})	126.58	127.60	99
Contr. NH_4^+ to NH_x wet dep. (mol ha^{-1} yr^{-1})	85.52	201.51	42
NH_x wet dep. (mol ha^{-1} yr^{-1})	212.10	329.11	64
Station Keldsnor			
NH_3 conc. (μg NH_3 m^{-3})	0.448	0.656	68
NH_4 conc. (μg NH_4 m^{-3})	0.625	2.920	21
Contr. NH_3 to NH_x wet dep. (mol ha^{-1} yr^{-1})	11.70	54.31	22
Contr. NH_4^+ to NH_x wet dep. (mol ha^{-1} yr^{-1})	31.40	284.09	11
NH_x wet dep. (mol ha^{-1} yr^{-1})	43.10	338.40	13

added the effect of interactions with other atmospheric pollutants, such as SO_2 and NO_x, which may affect the partitioning between NH_3 and NH_4^+ aerosol, as well as affect the lifetime of NH_x in the atmosphere. Parallel changes in SO_2 and NO_x emissions therefore complicate the interpretation of NH_x monitoring data. There are also other difficulties in interpreting NH_3 and NH_4^+, such as spatial variability and scale of the region influence, as well as non-linear effects introduced by atmospheric chemistry or bi-directional NH_3 exchange. These different issues are addressed below.

5.1. THE EFFECT OF INTERACTIONS BETWEEN NH_x AND OTHER POLLUTANTS

The interaction of NH_x with SO_2 and NO_x presents particular difficulties in interpreting trends in monitored NH_x. In particular, precipitation NH_4^+ concentrations and wet deposition, which are often the only datasets with long-term records, are very closely coupled to oxidized N and S chemistry. This is clearly seen from the recent case studies of The Netherlands, Hungary, Russia and Switzerland, but has also been mentioned in interpreting the historical data on NH_4^+ in ice cores (Döscher *et al.*, 1996). Currently, atmospheric transport models tend to include parametrizations of part of the problem, namely the interaction with wet deposition, but do not treat the interaction with dry deposition (e.g. Asman and Van Jaarsveld, 1992; Bartniki, 2000; Van Jaarsveld *et al.*, 2000; Singles *et al.*, 1998; Sorteberg and Hov, 1996). These two interactions of NH_3 with SO_2 and NO_x chemistry are both important since they may have opposite effects.

Reaction of NH_3 with SO_x and NO_y in the atmosphere produces NH_4^+ aerosol that is inefficiently removed by dry deposition. This results in a longer atmospheric residence time of NH_4^+ aerosol, with most removed by wet deposition. As a consequence, increased amounts of SO_2 and NO_x emission would tend to increase the atmospheric residence time of NH_x, leading to greater long-range transport. Hence controls on SO_2 emission may decrease NH_4^+ wet deposition in regions remote from sources, but increase it in NH_3 source regions (Möller and Schieferdecker, 1989). It is important here to distinguish between the contribution of NH_4^+ aerosol scavenging to wet deposition (Figure 16), which would decrease, while scavenging of NH_3 would increase. This provides one reason why the wet deposition did not decrease as much as expected in The Netherlands, while more than expected decreases are seen in other areas, such as Switzerland and West Germany.

Similar chemical interactions that govern the atmospheric transformation of NH_3 also affect dry deposition, with the only difference being that the aerosols and water droplets are attached to ground surfaces, which add other ions to the chemical interaction (Sutton *et al.*, 1995a; Erisman and Draaijers, 1995; Flechard *et al.*, 1999). The presence of SO_2 may increase the cuticular uptake for NH_3, since its solubility in water films is pH-dependent (Adema *et al.*, 1986). Leaf surface interactions with other acid gases such as HNO_3 and HCl may also be important. However, because the NH_4^+ salts with these gases (NH_4NO_3 and NH_4Cl) have significant vapour pressure, they may re-dissociate back to the precursor gases, limiting accumulation of salts on leaf surfaces (Nemitz *et al.*, 2000b). By contrast, ammonium sulphates have negligible vapour pressure and can easily accumulate on leaf surfaces. As a result of these interactions, a decrease in supply of SO_2 may result in reduced rates of NH_3 deposition. This may be parametrized through increased cuticular resistances (R_w) or as a reduced dynamic uptake by leaf cuticles (Sutton *et al.*, 1995b, 1998; Flechard *et al.*, 1999; Erisman *et al.*, 2001).

The trend in throughfall to forest at IJsselstein in The Netherlands is useful to consider these interactions (Figure 20). Although NH_3 concentrations in The Netherlands did not decrease, the throughfall flux of NH_4^+, NO_3^- and SO_4^{2-} decreased substantially (even considering differences in precipitation between years). The reduced SO_2 concentrations may have led to larger values of R_w for NH_3, which reduced NH_3 dry deposition. This effect is therefore opposite to the wet deposition interaction. Increased SO_2 leads to more dry deposition of NH_3, which tends to reduce the atmospheric residence time of NH_x. This effect would be largest in source areas since here NH_3 concentrations and dry deposition are largest. Hence in The Netherlands example, a decrease in SO_2 levels would both tend to reduce the rate of NH_3 deposition, and tend to increase NH_3 concentrations and NH_3 export.

5.2. CONTROLS ON THE TEMPORAL VARIABILITY OF NH_x COMPONENTS

While the chemical and biophysical controls on NH_3 and NH_4^+ in the atmosphere are relatively well understood, it is accepted that it is rather difficult to quantify these effects. This is because of the complexity of the interactions, for example with wetness, temperature, leaf chemistry, meteorological conditions and agricultural management practices. Figures 5 and 8 demonstrate the substantial temporal variations in NH_3 and NH_4^+ concentrations and NH_4^+ wet deposition. While values for Figure 5 are shown on a daily level, there are also substantial fluctuations over minute and hourly scales (e.g. Burkhardt *et al.*, 1998). It is important to understand these if a reliable interpretation of monitoring data should be made.

With regard to long-term trends, inter-annual weather differences present the most obvious difficulty, as illustrated in many of the examples. These show that unambiguous trends should not be expected at single sites for periods of less than 5–7 yr, even for major changes in emission (Figure 14). Even where multi-site monitoring can provide more representative values (e.g. Figure 19), the effect of common meteorological differences between sites cannot be avoided. The only way to do this would be to select sites far enough apart to have different annual meteorological patterns. Unfortunately, such sites would also be so far apart as probably to have very different emission trends.

Short-term interactions with meteorological conditions may provide a number of important non-linear effects. An example of this is the link between NH_3 concentrations and windspeed, with larger NH_3 concentrations occurring in source areas during low windspeed conditions (Figure 23). The reason for this is that dispersion away from a source is less effective in low wind conditions, an effect that is exacerbated by stable stratification. As a result of this, in source regions NH_3 concentrations are larger at night than in the day (e.g. Burkhardt *et al.*, 1998) with the result that nocturnal conditions have a larger influence on the average NH_3 concentration in long-term monitoring (Van Jaarsveld *et al.*, 2000). This point is relevant to assessing the effectiveness of NH_3 emission control when using the NH_3 concentration record. In The Netherlands, much of the expected abatement is due to

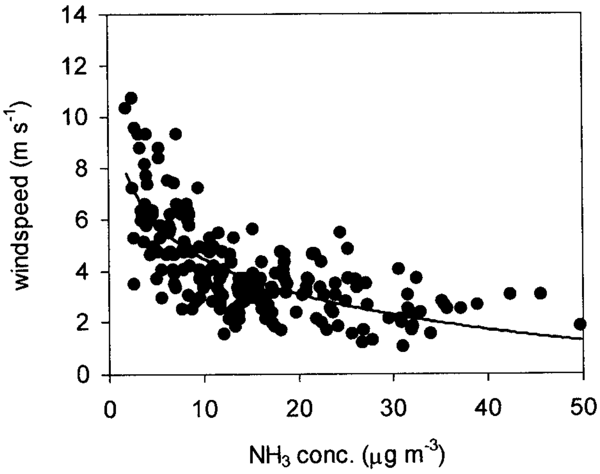

Figure 23. Relationship of NH_3 concentrations to windspeed at Vredepeel, a high NH_3 emission area of The Netherlands (Van Jaarsveld *et al.*, 2000). In source regions NH_3 concentrations are smaller at high windspeeds, as emissions are better mixed with the atmosphere.

control of emissions from manure spreading, while housing emissions are largely unchanged. Emissions from manure spreading tend to be largest in the day and small at night, while emissions from ventilated animal houses continue similarly in the day and night. As a result, the monitoring record reflects the emissions from animal housing more than manure spreading, and it becomes harder to detect the influence of manure-spreading-based NH_3 abatement measures in the monitoring record.

Further temporal interactions and non-linear effects are introduced by the existence of the compensation point for NH_3, which allows bi-directional fluxes of NH_3 to occur (Farquhar *et al.*, 1980; Sutton *et al.*, 1995b; Schjoerring *et al.*, 1998). The tendency is for NH_3 emission during the day (when the canopy is warm) and for deposition at night (when the canopy is cool and wet) (Schjoerring, 1995; Sutton *et al.*, 1998; Flechard *et al.*, 1999). This modifies the diurnal pattern of NH_3 concentrations depending on the region and the nature of sources, and similarly imposes an annual cycle on NH_3 concentrations. In regions with few livestock, the compensation point may provide a major control to ground level NH_3 concentrations (Langford and Fehsenfeld, 1993; Sutton *et al.*, 2000). This is relevant when interpreting the long-term NH_3 record at K-pustza in Hungary. It is possible that NH_3 concentrations at this site are largely controlled by a compensation point, rather than more distant livestock sources of NH_3. Further analysis of the local sources of NH_3 at this site is warranted, particularly given the unexplained increase in NH_3 during 1999.

5.3. SPATIAL VARIABILITY OF NH_x COMPONENTS

As a secondary product, NH_4^+ aerosol tends to have a spatially smooth distribution (Figure 16) making it well suited for monitoring at a limited number of sites. By contrast, the existence of NH_3 sources in the rural environment leads to substantial spatial variability in NH_3 concentrations. While one option is to sample in remote locations well away from sources, as at K-puszta, when sampling in NH_3 emission areas it is essential to be aware of the local influences on NH_3 concentrations. For this reason, generalizing NH_3 concentration trends requires assessment from a large number of sites (Lövblad and Sutton, 1997). In the U.K. a network of active and passive sampling has been established with around 70 sites, while in The Netherlands, high-spatial-resolution sampling is specifically targeted to example source areas. Such multiple-site sampling can be used to assess trends more reliably, as well as spatial patterns. As an example, Figure 24 shows the record of NH_3 concentrations contrasting two source areas in England. While there is a high degree of scatter between the individual sites, when averaged it becomes clear that the concentrations in the two regions are correlated even though they are approximately 300 km apart. This demonstrates the common seasonal and inter-annual influences on the NH_3 records resulting from meteorological and agricultural management interactions.

The spatial variability of NH_3 concentrations in a source area are illustrated by Figure 25, which shows results from monitoring in the Peel area of The Netherlands. Even when not sampling within 150 m of farmsteads, the concentrations vary by a factor of 5 across this area of 10 km × 10 km. Duyzer *et al.* (2000) estimate that to average out such effects requires sampling at approximately 8 locations for each 5 km grid square.

6. Conclusions: Improving the Link between NH_3 Emission Abatement and NH_x Monitoring

From the above it can be seen that there are a several difficulties and uncertainties in assessing the effectiveness of NH_3 abatement from monitoring networks. To do this requires sound monitoring methods implemented at sufficient sites and over a sufficiently long period, even assuming that there are no other changes in pollutants which may interact with NH_3. For NH_4^+ aerosol and NH_4^+ in rain, a modest number of sites can be used to indicate trends, whereas for NH_3 in source areas a high density of sites is essential. In contrast to the need for many NH_3 sampling locations, is the requirement for high temporal resolution NH_3 concentration data at selected sites. This may be used for example to address the effectiveness of different emission controls by contrasting trends in day and night-time NH_3 concentrations. For example, the expected trend in NH_3 in The Netherlands due to improved manure spreading technology may be more easily detectable by analysis

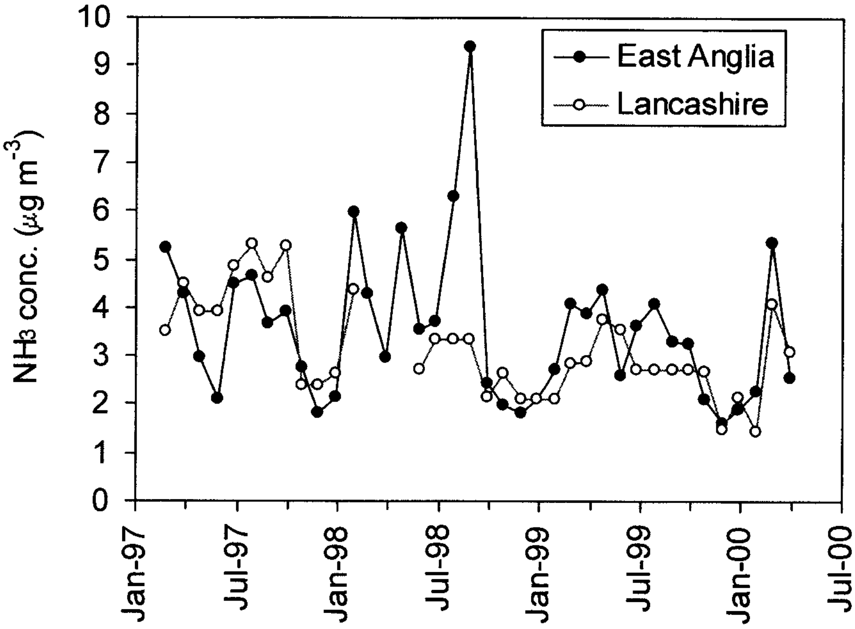

Figure 24. Record of NH_3 for example source regions of the U.K. Values for East Anglia are the means of 9 sites, while those for Lancashire are the mean of two sites. NH_3 emissions in East Anglia might have been expected to have declined from 1999 as large quantities of poultry waste are now incinerated for electricity generation rather than spread on land. The correlation between concentrations in these regions 300 km apart demonstrates the influence of weather conditions and agricultural management.

of day-time NH_3 concentrations. It is necessary to review recent recommendations for NH_3 monitoring strategies (e.g. Lövblad and Sutton, 1997; Sutton *et al.*, 2001) and provide clear guidance on the sampling frequency, density and robust methods for separating NH_3 and NH_4^+. Having confirmed the necessary criteria for monitoring, it is relevant to assess what the limitations are which may prevent countries from adopting the current recommendations (Sutton, 2001).

Quantifying the interactions of NH_x necessary to interpret long-term trends also requires improved mechanistic understanding and modelling. There are major uncertainties in dry deposition which still need to be built into atmospheric transport models. Firstly, better generalization is required on the bi-directional controls on NH_3 exchange, while secondly the chemical interactions that are recognized for atmospheric chemistry also need to be treated in relation to dry deposition. These interactions are well recognized, but still very uncertain and require further process studies to allow generalizations to be developed. Work is also required to advance the regional-temporal modelling of NH_3 emissions in relation to environmental conditions.

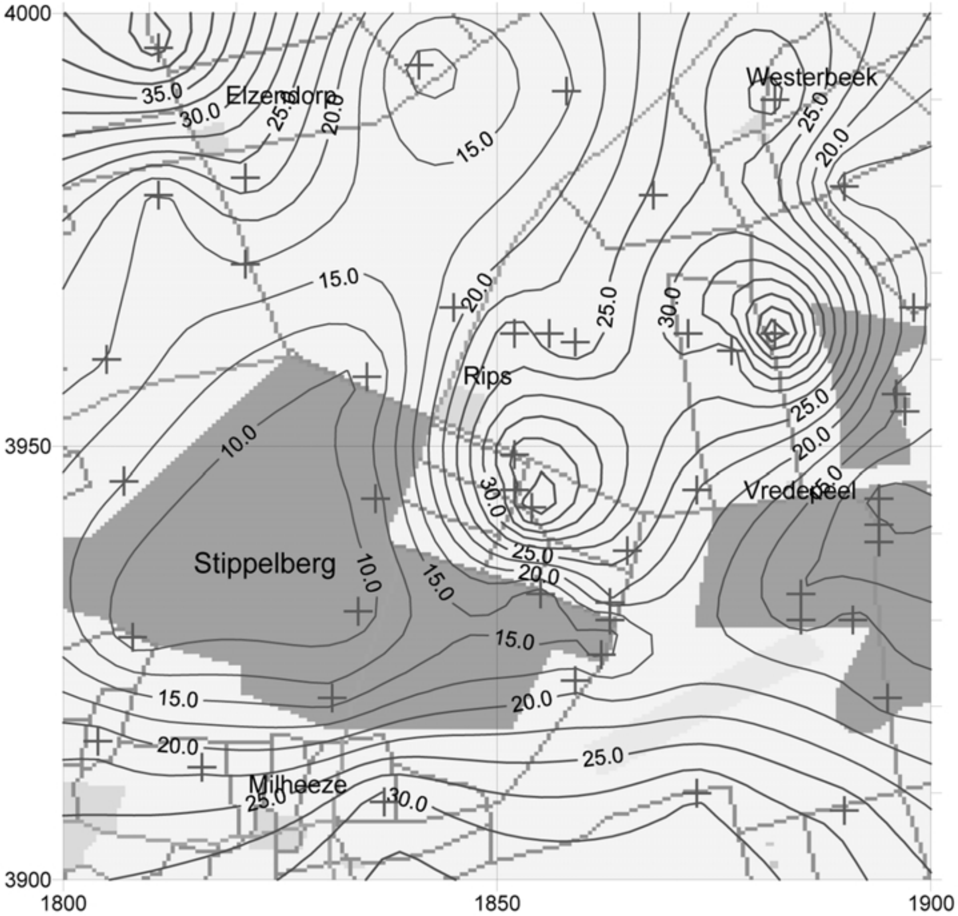

Figure 25. Ammonia concentration in the Peel area of The Netherlands. + indicates measurement site. Values are in μg m^{-3}. The map covers an area of 10 km × 10 km. Sampling was avoided within 150 m of livestock buildings and manure stores.

Finally, it is important to retain caution in attributing changes in atmospheric NH$_x$ to changes in NH$_3$ emission. This applies both to increases and decreases in measured NH$_x$, and whether the change links to altered source sector activity (e.g. changed animal numbers and fertilizer usage) or to the application of technical abatement measures. The examples illustrated here show how it is sometimes difficult to detect NH$_3$ emission changes even where these certainly occurred. In assessing the success of any abatement policy based on technical measures a combination of appropriate modelling and sufficient measurements should be able to determine whether the measures are broadly effective. However, where there is a gap between the monitoring response expected and that observed, this may be as much due to limitations in atmospheric process quantification and monitoring, as to ineffectiveness of the abatement techniques.

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