Trends of nitrogen in air and precipitation in Europe, 1980-2002: Comparison of observations and model results

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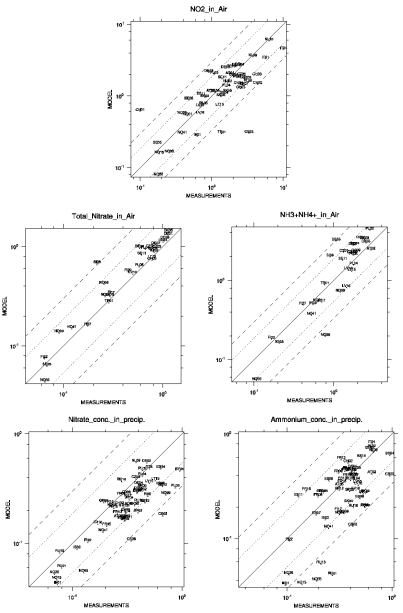

Figure 1. Scatter plots for modelled versus observed yearly averaged data at all EMEP sites that have measurements for N species in 2002.

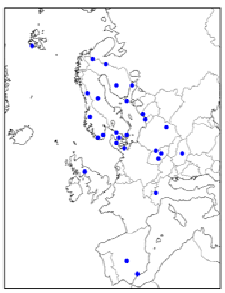

Figure 2. Location of EMEP stations that have long term measurements of nitrogen in air

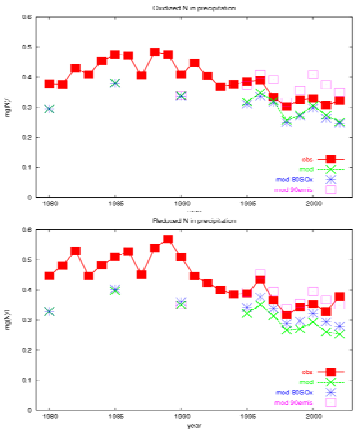

Figure 3. Model calculated and measured yearly averaged concentrations in precipitation of oxidized (upper) and reduced (lower) nitrogen in precipitation 1980-2002. The sites used in the average are the same as shown in figure 2, except a few sites were the long term time series contained unreasonable discontinuities. The modelled time series follow the observed ones closely, although concentrations of nitrogen in precipitation are somewhat underestimated. The model results indicate that reduction of SOx emissions have lead to a somewhat larger decrease in reduced nitrogen concentrations in precipitation than expected from the decrease in ammonia emissions. The effect on oxidized nitrogen in precipitation is small.

Introduction

A major aim of EMEP (Cooperative Programme on the Long Range Transmission of Air Pollutants in Europe, http://www.emep.int) is to support governments with scientific guidance on the causes of air pollution concentrations and depositions within Europe. The work within EMEP was initiated with a focus on reducing sulphur air pollution, but it was soon realized that nitrogen posed a threat as serious as that of sulphur. Recently, health impact aspects due to particulate matter have become more in focus, and ammonia emissions are the gaseous precursor of ammonium aerosols that can be transported over long distances and affect Particulate Matter (PM) levels over thousands of kilometers. The EMEP model (documented in Simpson et al. 2003 and Fagerli et al. 2004) results are an essential input to the RAINS integrated assessment model, and have been crucial to a number of UN-ECE Protocols and the European Union National Emission Ceilings Directive. It is essential that the model responds correctly to emission changes (e.g. in concentrations and depositions), and one way to test this is through it's ability to reproduce trends. In this study we have focused on the years 1990-2002 since almost no measurements of nitrogen compounds in air are available from the EMEP network before 1990. For nitrogen in precipitation we compare modelled and measured values back to 1980, at the same sites where measurements of nitrogen in air are available.

Emission changes

Sulphur emissions in Europe have been reduced by approximately 60% and 50% since 1980 and 1990, respectively (Vestreng et al. 2004), resulting in a large decrease in concentrations of sulphur in air. At the same time, NOx emissions have decreased by 25%, most of these reductions taking place in the 1990s. The total European reductions in ammonia emissions from 1980 to 2002 was similar to those of NOx. However, the reduction trend was somewhat different in different parts of Europe. In eastern European countries NH_3 emissions have been reduced by approximately 50% after 1990. In the rest of Europe, only small changes (~10%) have taken place. The large changes in emission regimes have led to changes in the atmospheric residence time of nitrogen as the reduced sulphur concentrations in air has been followed by less formation of ammonium sulphate and relatively more formation of ammonium nitrate. Unfortunately most of the measurement sites are located far from the source areas, reflecting only the averaged effect after long range transport. Moreover, very few eastern European countries have reported long term measurements to EMEP.

Measurements

In 2002, many EMEP sites, covering large parts of Europe, measured concentrations of nitrogen in air and precipitation. The number of the stations which have long term measurements of total nitrate and sum ammonia+ammonium in air is a lot fewer. The location of these sites are shown in figure 2. Most of the sites are located in the north west of Europe, thus the conclusions drawn here on trends are only representative for this region. Trend analysis for EMEP sites with nitrogen measurements in air and precipitation from 1990 to 2002 were done using the Mann Kendall method. For most sites, the downward trend in NO_2 is statistically significant. Half of the sites show a negative trend for NHx in air, some sites even show positive trend due to local sources. For total nitrate, only 6 out of 26 stations show a statistically significant decrease. The downward trend for concentrations in precipitation is statistically significant at approximately half of the stations, both in reduced and oxidised nitrogen.

Calculations

We have performed 3 sets of calculations with the EMEP model:

1. Base case: 1980,1985,1990,1995-2002 using emissions (as reported to EMEP) and meteorology for the respective years
2. The effects of SOx emission reductions: same as in 1. but keeping SOx emissions at the 1980 level
3. Meteorological variability: 1990,1995-2002, keeping all emissions at the 1990 level

Conclusions

The EMEP model results for nitrogen in air and precipitation compare favourably with the observations as shown in figure 1. For most sites, nitrogen show a downward trend, although not always to an extent that is statistically significant. Model calculations demonstrate that the large reductions in SOx emissions since 1980 probably have influenced the trends of nitrogen species. Even with the limited number of long term measurements available for this study, it is clear from the results that it is important to understand the role of the interaction between nitrogen and sulphur in order to understand the trends of nitrogen.

Meteorological variability

The meteorological variability is large and in many cases of the same magnitude as the changes in concentrations over the time period studied here. This makes it difficult to extract trends in observations. However, using model calculations, the changes in concentrations due to emissions can be separated from the effect of meteorological variability. In figure 3-6 this can be done by in each point subtracting the green from the pink marks (The only differences in each point are the emissions) . This exercise shows that concentrations of all the nitrogen compounds have decreased for the sites shown here and most of the other sites.

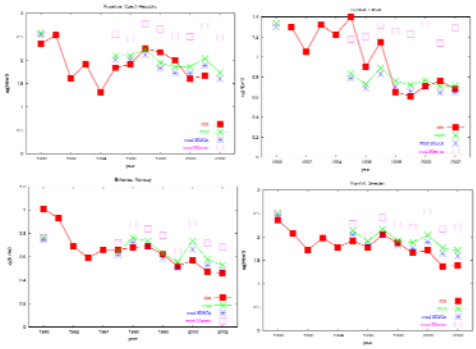

Figure 4. Comparison of observed and modelled NO_2 concentrations 1990-2002 at selected EMEP sites. The observations and the model agree that NO_2 concentrations have decreased substantially since 1990, largely following the emission reductions. The influence of SOx emission reductions has been small, increasing NO_2 concentrations somewhat through a lower night time conversion to HNO_3.

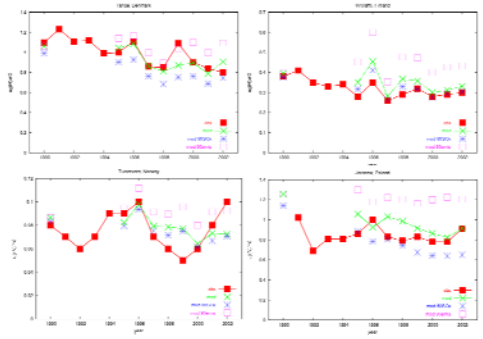

Figure 5. Comparison of observed and modelled total nitrate concentrations 1990-2002 at selected EMEP sites. The modelled time series of total nitrate in air are in most cases in reasonable agreement with the measurements, although nitrate is somewhat overestimated. The model results indicate that SOx emission reductions have influenced nitrate concentrations significantly. Nitrate in air has not decreased to the same extent as NOx emissions, probably due to relatively higher formation of ammonium nitrate at the cost of ammonium sulphate.

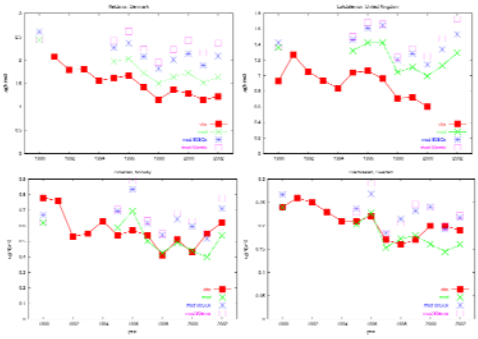

Figure 6. Comparison of observed and modelled NH_3+NH_4^+ concentrations 1990-2002 at selected EMEP sites. The observed and modelled NH_3+NH_4^+ time series correlate well. SOx emission reductions have decreased the level of NHx more than what should be expected from the NH_3 emission reductions, probably due to a lower degree of NH_4^+ aerosol formation.

References

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