

## Deposition trends in the UK between 1986 and 1999

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Sulphur dioxide emissions in Europe have seen an approximate 50% reduction over the last decade. By 1997, the total sulphur deposition to Great Britain (England, Wales and Scotland) had fallen by 52%, with dry deposition decreasing by 62% and wet deposition by 42%. There has been a clear non-linearity between the deposition and the emission trends over the period.

The trend in concentration reduction is not uniform across the country. Concentrations of sulphate in rainfall decline at some sites but not apparently at others, and there is considerable variability between years. Site groupings were created from the concentration data using a similarity matrix. A dendrogram and a principal component plot with a minimum spanning tree showed groupings which were geographically consistent. An assessment of the strengths of the group linkages allowed an initial grouping into 11 categories to be combined to give four groups in the final stage. The groups allowed an assessment of regional trends with greatest decline in rainfall concentrations ( $3.25 \mu\text{eq l}^{-1} \text{y}^{-1}$ ) in the traditional source emission regions of the UK but negligible, if any, declines in the west and north of the country. Recent data have confirmed the usefulness of the group structure with some of the more remote sites now showing small declines by 1999.

Although the sites with no declining trend were also sites with relatively low concentration, these are also in the areas with substantial critical load exceedances in Britain. These areas have high deposition because of the orographic enhancement of both rainfall amount and rain ion concentrations. Even with relatively small concentrations in rainfall over low ground, the ion concentration in hill cloud droplets can be quite large and, when they are washed out by rain from higher altitude clouds, lead to substantially greater deposition in hill areas than predicted without the orographic effects. The consequence of the lack of decline in sulphate in the west of Britain is that the recovery of fragile ecosystems following emission controls will be much slower than anticipated.

One of the various hypotheses suggested for the non-linearity between emissions and deposition of sulphur was the potential relationship between canopy resistance and the relative  $\text{SO}_2$  and  $\text{NH}_3$  concentration ratio. Deposition fluxes are measured routinely at Auchencorth Moss, near Edinburgh, and at Sutton Bonnington, in the east Midlands of England. The LIFE programme also provided data for an extended period at Melpitz in Germany and Speulde in the Netherlands. These showed that a reduction in the  $\text{NH}_4/\text{SO}_4$  aerosol concentration ratio from 0.8 to 0.4 was associated with an increase in  $R_c$  from  $40 \text{ s m}^{-1}$  to  $180 \text{ s m}^{-1}$ . Similar evidence comes from Sutton Bonnington where an average  $R_c$  in May to July for 1974-1975 was  $130 \text{ s m}^{-1}$  while, following a substantial decline in  $\text{SO}_2$  concentrations, the same measurement procedures in 1995-1997 gave a value of  $80 \text{ s m}^{-1}$ . An important consequence for the trend analyses required for the assessment

report is that deposition and transport models may need to have a variable canopy resistance over the period 1980-2000.

Recent data on atmospheric nitrogen concentrations show a similar pattern to sulphur and confirm that the non-linearity issue is not a purely sulphur phenomenon. Emissions of nitrogen oxides in the UK have approximately halved (850 kt NO<sub>x</sub>-N in 1989 to 470 kt NO<sub>x</sub>-N in 1999) over the decade. Nitrogen dioxide concentrations in air have fallen in line with emissions at all monitoring sites, but mean concentrations over all sites of nitrate in rainfall have declined slightly (0.3 µeq l<sup>-1</sup> y<sup>-1</sup>) but not statistically significantly. However using the site groupings derived for sulphate, the nitrate concentrations have fallen significantly in the high sulphate, source areas by 0.8 µeq l<sup>-1</sup> y<sup>-1</sup> to around 35 µeq l<sup>-1</sup> y<sup>-1</sup> since the peak emissions in 1989. Ammonia emissions are assumed to have been approximately constant over the period at 280 kt NH<sub>3</sub>-N. There is an apparent decline in ammonium in rainfall over all sites of 0.4 µeq l<sup>-1</sup> y<sup>-1</sup> which is not statistically significant but there is also evidence of substantial inter-year variability which would obscure trends even in site groups. There are two sites with significant declines in concentration (Stoke Ferry with 1.3 µeq l<sup>-1</sup> y<sup>-1</sup> and Barcombe Mills with 1.5 µeq l<sup>-1</sup> y<sup>-1</sup>) and the data suggest an unconfirmed fall in ammonium concentration in the more highly polluted regions.

In summary, the improvements in air quality and in deposition in the UK have been concentrated in the source regions. The remoter areas have seen less improvement, particularly with wet deposition, and it appears that the recovery of acid sensitive sites will take much longer than originally predicted. There is also evidence that deposition velocities of sulphur dioxide and ammonia have changed over the period 1980 to 2000 and therefore the transport and deposition models may require adjustments to include these changes for satisfactory long-term trend analysis.