Trends in rural sulphur dioxide concentrations in the United Kingdom

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Abstract

Emissions of sulphur dioxide in the United Kingdom and the rest of Europe have declined markedly over the last two decades in line with a succession of international agreements. Over the area covered by the United Nations Economic Commission for Europe’s Environmental Monitoring and Evaluation Programme, total SO₂ emissions fell from 59 Tg in 1980 to 27 Tg in 1997. In the United Kingdom, emissions fell from 6.4 Tg in 1970 to 1.2 Tg in 1999. Measurements of rural SO₂ concentrations provide an opportunity to investigate the efficacy of these policy measures in improving air quality and reducing dry sulphur deposition. Concentrations have been mapped and show a dramatic decrease in major source areas, with smaller reductions in the north and west. Annual mean concentrations are now less than 4 ppb over most of the country. In the United Kingdom the decrease in emissions occurred in two broad phases. Until 1990 reductions were mostly in emissions from low-level domestic, commercial and industrial sources; thereafter the decrease was dominated by reduced high-level emissions from power stations. Measurements at a number of rural sites are consistent with these changes but lack of data on local emissions makes it difficult to interpret the profile of decrease at individual sites. As wet deposition has decreased less quickly than dry the ratio of wet to dry deposition has increased with the former now dominating over most of the country.
1 Introduction

Emissions of sulphur dioxide in the United Kingdom and the rest of Europe have declined markedly over the last two decades. Over the area covered by the United Nations Economic Commission for Europe’s Environmental Monitoring and Evaluation Programme, total sulphur dioxide emissions fell from 59 Tg in 1980 to 27 Tg in 1997 as reported by Tarrason and Schaug [1]. In the United Kingdom, Goodwin et al. [2] have estimated that emissions fell from 6.4 Tg in 1970 to 1.2 Tg in 1999. A national monitoring network allows the broad spatial pattern of rural SO₂ to be determined. Prior to the establishment of this network in 1991, gaseous and particulate sulphur species were less extensively monitored but some data are available to allow investigation of spatial patterns and temporal trends.

2 Measurements

A national rural sulphur dioxide monitoring network was established in 1992, based around a less extensive network that had been operating since 1987. Daily mean sulphur dioxide and particulate sulphate concentrations are measured at eight sites. Weekly means are measured at the other sites. The locations of sites referred to in this paper are shown in Figure 1. Other organisations in the UK have provided annual mean concentrations for recent years from rural automatic monitors for mapping. The Institute of Terrestrial Ecology operates a site in southern Scotland (Auchencorth Moss) and the Joint Environment Programme, an environmental association of power generation companies in the United Kingdom, operates twelve sites in England.

2.1 Methodology

Sulphur dioxide and particulate sulphate are measured using a low volume bubbler apparatus (AGL, Hitchin, UK). Air is drawn through a PVC feeder tube, a paper filter (Whatman 40), a 4 mm id glass tube and then a 0.5% hydrogen peroxide solution. The flow rate, and hence sensitivity, is limited by the loss of peroxide solution by evaporation, particularly in hot weather. Bubbler solutions are analysed by ion chromatography. Both the peroxide blank and the analytical detection limit are around 0.01 mg S l⁻¹. Hence, blank uncertainty limits the precision of the measurement of sulphur dioxide to around 0.2 ppb. A calibrated gas meter mounted on the vacuum side of the pump measures airflow and readings are not corrected for temperature or pressure. The bubbler system is checked for leaks annually and leakage is generally a few percent of the inlet flow. As a result of the uncorrected meter readings and the small leaks, measurements may be subject to a systematic under read of 5 to 10 % but are not corrected.

The full data set is available - http://www.aeat.co.uk/netcen/airqual/index.html.
2.2 Mapping

Annual mean concentrations of sulphur dioxide for 1987 to 1989 and 1997 to 1999 were mapped using a similar technique to that described for rainwater composition by Webster et al [3]. The resulting maps are shown in Figure 2. The spatial variance is larger than that in the concentrations of mapped ions in precipitation, especially in regions with significant low-level sources. Such low-level sources include the burning of coal in domestic grates and small oil-fired boilers. These maps show the general national-scale gradients but do not resolve local areas of elevated concentration. The maps show a decline in concentrations across the whole country with a clear decrease in the area experiencing concentrations greater than 4 ppb. The spatial patterns reflect the distribution of emissions and have remained broadly consistent over the ten-year period. Rural sulphur dioxide concentrations are largest in Central England due to a combination of major point sources and residual coal use in small boilers and domestic heating.
Maps utilising spatially disaggregated emissions to estimate urban concentrations have been published by Stedman [4] and Abbott & Vincent [5]. These, and measurements in South Wales, Central Scotland and eastern Northern Ireland, suggest that local peaks are no more than 2 ppb larger than the wider spatial pattern.

3 Trends

Measurements of rural SO₂ concentrations provide an opportunity to investigate trends and thereby the efficacy of policy measures both in improving air quality and reducing dry sulphur deposition.

3.1 Sulphur dioxide emissions

Total UK emissions of sulphur dioxide decreased from 6.4 Tg in 1970 to 1.66 Tg in 1997. Over the same period emissions from industry (excluding power generation) decreased from about 2.5 Tg to 0.45 Tg, while emissions from domestic and commercial sources decreased from about 0.84 Tg to 0.11 Tg. These changes are illustrated in Figure 3, which shows that the industry and combined domestic and commercial sources follow a similar trend. The rate of decrease was relatively constant over the period, although industrial emissions decreased particularly rapidly between 1979 and 1984. Initially emissions from power generation were relatively constant; only decreasing from 3 Tg to 2.5 Tg until about 1990. They have since decreased markedly as power generators replace coal-fired capacity with natural gas CCGT. The commissioning of flue gas desulphurisation on 6 GW of capacity and increased availability of nuclear
power have also contributed to the decline. By 1997, emissions from this sector had decreased to around 1 Tg.

Figure 3: UK total emissions of sulphur dioxide, 1970 to 1997 (ktone).

### 3.2 Trends in sulphur dioxide and particulate sulphate concentrations

Regression analyses were carried out for daily measurements of sulphur dioxide and particulate sulphate concentrations for the period 1986 to 1997 using annual mean concentrations derived from daily data. During this period national SO$_2$ emissions decreased from 3.7 Tg to 1.7 Tg. A summary of the regression output data is presented in Table 1. Lough Navar, Yarner Wood and High Muffles showed a statistically significant downward trend in sulphur dioxide concentrations. The detection limit for sulphur dioxide using the bubbler methodology is inadequate for the accurate determination of trends at remote locations such as Strathvaich Dam. For particulate sulphate, only at High Muffles, located closer to major sources, was a significant trend observed. This may be attributed to the decrease in the emission of sulphur dioxide and particulate sulphate from power stations in the region. For the other sites, a small insignificant trend in concentration was observed.

Monitoring of non-seasalt sulphate in rain, sulphur dioxide and particulate sulphate has been carried out at Eskdalemuir, a remote rural site in southern Scotland since the 1970’s. From about 1978, monitoring methodologies comparable to those currently employed allow trends to be assessed. Figure 4 shows annual mean concentrations for sulphur dioxide and particulate sulphate. Only sulphur dioxide concentration has a statistically significant trend. The measured concentration decreased sharply from 1981 to 1982. In view of this a potential site-specific effect was examined, namely a change in fuel for space heating at the Eskdalemuir Observatory which occurred during 1981. A
modelling study showed that this change was unlikely to have resulted in a detectable change in measured SO$_2$ concentrations at the sampling site.

Table 1: Regression of annual concentration against time for the period 1986 to 1997 (SO$_2$ in ppb and particulate sulphate in $\mu$g S m$^{-3}$).

<table>
<thead>
<tr>
<th>Site name and measurement period</th>
<th>Species</th>
<th>Period Mean</th>
<th>Regression Coefficient</th>
<th>Significance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eskdalemuir Measurement Period = 12 years</td>
<td>SO$_2$</td>
<td>1.04</td>
<td>-0.077</td>
<td>0.075</td>
</tr>
<tr>
<td></td>
<td>Particulate S</td>
<td>0.85</td>
<td>-0.005</td>
<td>0.725</td>
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<td>Lough Navar Measurement Period = 12 years</td>
<td>SO$_2$</td>
<td>0.62</td>
<td>-0.060</td>
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<tr>
<td></td>
<td>Particulate S</td>
<td>0.90</td>
<td>-0.043</td>
<td>0.141</td>
</tr>
<tr>
<td>Yarner Wood Measurement Period = 11 years</td>
<td>SO$_2$</td>
<td>1.37</td>
<td>-0.161</td>
<td>0.023</td>
</tr>
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<td></td>
<td>Particulate S</td>
<td>1.28</td>
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<td>0.051</td>
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<td>High Muffles Measurement Period = 11 years</td>
<td>SO$_2$</td>
<td>3.60</td>
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<td>0.0007</td>
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<td></td>
<td>Particulate S</td>
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<td>0.0016</td>
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<tr>
<td>Strathvaich Dam Measurement Period = 11 years</td>
<td>SO$_2$</td>
<td>0.50</td>
<td>0.003</td>
<td>0.8981</td>
</tr>
</tbody>
</table>
| | Particulate S | 0.66 | * | *

Note: The significant changes in concentration are marked in bold.

Figure 4: Annual mean concentrations of sulphur dioxide (ppb) and particulate sulphate ($\mu$g S m$^{-3}$) at Eskdalemuir.
Data on sulphur dioxide concentrations have also been reported for Ratcliffe, Marshfield and Husborne Crawley by Downing & Campbell [161]. These sites are situated in rural areas but are within approximately 3 km of major roads or small villages and in the case of Husborne Crawley close to an area of brick production. At each site, annual mean concentrations have decreased significantly; concentrations greater than 25 ppb were measured in the early 1970's, by 1997 measured concentrations were 2.8 ppb, 2.1 ppb and 1.3 ppb, at Ratcliffe, Husborne Crawley and Marshfield, respectively.

Detailed emission inventories are not available for the period in question and modelling has not been undertaken. However, the large decreases at sites such as Ratcliffe follow a similar pattern to the decrease in emissions from the domestic use of coal (Figure 5). The Trent Valley was formerly an area of large-scale coal production, with mined coal provided free to miners. Both the emissions of sulphur from domestic coal consumption and measured concentrations at Ratcliffe have decreased by an order of magnitude. Emission of sulphur dioxide from high-level sources (power stations) in the East Midlands remained more or less constant until the start of the 1990's. Hence, it can be inferred that the power stations in this region played a secondary role in the large decrease in sulphur dioxide concentrations before then.

![Figure 5: Sulphur dioxide concentrations measured at Ratcliffe (ppb) and emissions of sulphur dioxide from the domestic consumption of coal in the United Kingdom from 1970 to 1997 (ktonnes year⁻¹).](image)
A similar decrease is observed throughout the 1980's for many areas of the country. For some locations, such Marshfield and Husborne Crawley (these sites are nearly 200km apart) the reduction in concentration followed a similar pattern from 1979 onwards (Figure 6). The profile of the trend is different to that observed at Ratcliffe, but the same order of magnitude decrease was observed. The observed decreases may be expected to arise from the general reduction in emission from low-level sources.

Figure 6: Sulphur dioxide concentrations measured at Husborne Crawley and Marshfield, 1970 to 1997 (ppb).

Figure 7 shows how sulphur dioxide and non-sea salt sulphate concentrations have decreased from about 1989 onwards at a sampling site situated within the major source region in the East Midlands (Bottesford, Leicestershire). The decrease in sulphur dioxide concentrations appears to follow the rate of decrease in emission from local power stations. Here local is defined as power stations within 100 km of the sampling site. The sulphur dioxide emitted from these local stations remained a constant proportion of the national total (typically, 41 to 43 %) until 1995. Thereafter the fraction decreased to about 35 %. A modelling study by Abbott and Vincent [S] estimated that emissions from low-level area sources within 15 km of the site at Bottesford contributed less than 1 ppb of the concentration measured in 1996. Hence, over the last decade SO₂ concentrations at Bottesford would be expected to have reduced broadly in line with national high-level emissions.
Figure 7: Changes in nss sulphate in precipitation and SO₂ concentrations at Bottesford, with SO₂ emissions from power stations within a 100 km radius of the site.

Taken overall, these results suggest that for the period from 1970 to about 1990 annual mean concentrations of SO₂ in rural areas responded mainly to reductions in low-level emission sources. From 1990 onwards, the significant reductions in sulphur dioxide emitted from power stations became the major influence on concentrations in the source region. Further afield, and particularly in remote areas, the detection of a decrease in sulphur dioxide concentration is limited by the sensitivity of the bubbler methodology.

Conclusions

Evidence from a number of rural sites shows an order of magnitude reduction in SO₂ concentrations since 1970. At sites in the East Midlands this appears to have been largely influenced by reductions in low-level emissions over the period from 1970 to 1990 with power station emissions only contributing to the decrease from 1990 onwards. However, lack of data on emissions for earlier years makes it difficult to interpret the profile of decrease at individual sites. Dry deposition budgets prior to the establishment of a rural monitoring network are, therefore, subject to a great deal of uncertainty. As SO₂ concentrations have, in general, decreased more quickly than wet deposition the ratio of wet to dry deposition over the country has changed with wet deposition now dominating in all but some parts of the south and east.
Acknowledgements

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References


