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Monitoring Concentrations and Dry Deposition of Sulphur Dioxide in The United Kingdom

Final Contract Report to the Department of the Environment Transport & the Regions

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Contents

Summary

- Maps of mean annual SO_2 concentration in 1992-96 and mean annual dry deposition of SO_2 for 199294 are provided.

- Uncertainty in the estimates is discussed and standard error maps of SO_2 and non-marine sulphate in rain in 1995 are also provided.

- 'Continuous' dry deposition measurements at Sutton Bonington in the East Midlands indicate that ambient SO_2 concentrations in central UK are now small (~ 3 ppb SO_2) and have declined substantially since the mid 1980s.

- Comparison with the model used to calculate UK dry deposition shows that model over-estimates dry deposition by a factor of 2 or more.

- The measured median concentration of SO_2 at Auchencorth Moss in Central Scotland is very small (0.3 ppb) and concentrations seldom exceed 10 ppb.

- The measured SO_2 deposition rate for uplands is smaller than that used in the UK model.

- Validation of deposition model estimates against catchment budgets show good agreement between measured and modelled estimates of S deposition in both Leicestershire, where dry deposition dominates, and North Wales where wet deposition dominates.

- Rural SO_2 concentrations have declined over the last 20 years. However the decline in concentrations appears to exceed the decline in SO_2 emissions leading to an apparent non-linear relationship between dry deposition and total SO_2 emissions. The basis of non-linearity will be investigated in 1998/99.

- A deposition monitoring method of air pollution in Europe has been developed under the LIFE 1 programme. Dry deposition of 3 gases - SO_2, NO_x and NH_3 was measured continuously at 3 sites - Auchencorth Moss in Scotland, Speulder Forest in the Netherlands and Melpitz in southern Germany. The 3 sites follow a pollution gradient with the smallest fluxes occurring at Auchencorth Moss (except for sea salts) and the largest at Speulder forest, mainly due to the roughness of the forest and resulting high dry deposition velocities.

Introduction

The operation of UK rural monitoring network for SO_2 has continued for a further 3 years. NETCEN have continued to operate the UK network of rural SO_2 monitoring stations including 32 bubbler sites (providing daily SO_2 concentrations) and 5 continuous (hourly average) SO_2 monitoring stations using UVF methods. Monthly and annual SO_2 concentration fields have been provided by NETCEN for use in the ITE process-based SO_2 dry deposition model for the UK which provides inputs for the major land classes within each 20 x 20 km grid square of the UK.

The SO_2 dry deposition monitoring station for long-term validation of the dry deposition model over an agricultural landscape in Nottinghamshire has continued to provide continuous flux measurements. These data have been supplemented by data for an additional dry deposition monitoring station set up in 1994 under the EU LIFE programme at Auchencorth Moss near the ITE Edinburgh Research Station.

The estimates of dry (and wet) deposition from models can be validated using catchment hydrochemical budget for a range of UK sites. These include Beacon Hill in the East Midlands and Plynlimon and Llyn Brianne in central Wales.

Dry deposition inputs of SO_2 have been provided to the critical loads mapping centre and to the user community.

The reduction in SO_2 emissions over the past 10 years have been reflected in the SO_2 concentrations and deposition measurements. However the decline is non-linear, i.e. SO_2 concentrations and dry deposition in areas close to emission sources have shown a large decline in the rainfall concentration and wet deposition of S in areas remote from sources.

Objectives

- To provide monthly and annual SO_2 concentration and dry deposition estimates at 10 km by 10 km scale.

- To make continuous measurements of SO_2 dry deposition at a monitoring station in Nottinghamshire. As well as providing long term dry deposition measurements, this site will provide valuable information of the effects of the FGD equipment recently installed at Ratcliffe power station (6 km N of our monitoring station).

- To validate dry and wet deposition model estimates against catchment hydrochemical budgets for a range of UK sites. (Beacon Hill East Midlands, Plynlimon and Llyn Brianne in Central Wales.)

- To develop dry deposition estimates over large areas of Northern Europe based on monitoring networks in The Netherlands, Germany the UK, Denmark and Sweden in collaboration with the RIVM (Bilthoven) and IVL (Gothenburg).

- To compile and examine all existing rural UK SO_2 concentration measurements for the period 1970 to 1994 for evidence of changes in rural (and sub-urban) concentrations. In addition, these data, with existing rural and urban measurements and maps will be used to advise the Department of the Environment on the optimum density of monitoring stations.

Results

1. SO_2 concentrations and maps

Annual maps for SO_2 concentrations have been provided by NETCEN as part of this contract for the years 1994 and 1995. These have been used to model UK dry deposition at 20 km x 20 km resolution. The underlying methodology for concentrations and deposition is described in the recent CLAG fluxes subgroup report. The maps produced are averaged over the years 1992 to 1994 to reduce the influence of inter-year differences in meteorology and yet produce an up-to-date summary of the deposition climate.

The current network of approximately 40 sites with SO_2 monitors provides a concentration field using kriging methods. Such a network is not adequate to provide 10 km x 10 km resolution. Even with 20 km x 20 km resolution each monitor represents 600,000 ha of land area. The magnitude of variability in fine scale SO_2 concentration (e.g. 1 km x 1 km) is probably similar to those for SO_2 since both gases are deposited at terrestrial surfaces at smaller rates and both show pronounced chemical variations. Much of the systematic variability in SO_2 concentration could probably be simulated but the underpinning mechanistic studies and modelling have not been done.

Smith *et al.* (1995) estimated the fine scale variability in SO_2 deposition at some representative UK sites for illustrative purposes only. These estimates showed that in the polluted regions (e.g. E. Midlands) that local fine scale variability in deposition was mainly caused by variability in ambient SO_2 concentrations.

2. Deposition maps

Maps of mean annual concentration of SO_2 in 1992-95 and mean annual dry deposition of SO_2 for 1992-94 are shown in Figures 1 and 2. Possible uncertainty in the estimates of total deposition of sulphur have been studied.

The deposition models are linear with respect to the input concentrations of SO_4^{2-} or SO_2 and errors in these interpolated concentration fields are transmitted directly into the predicted deposition fields.

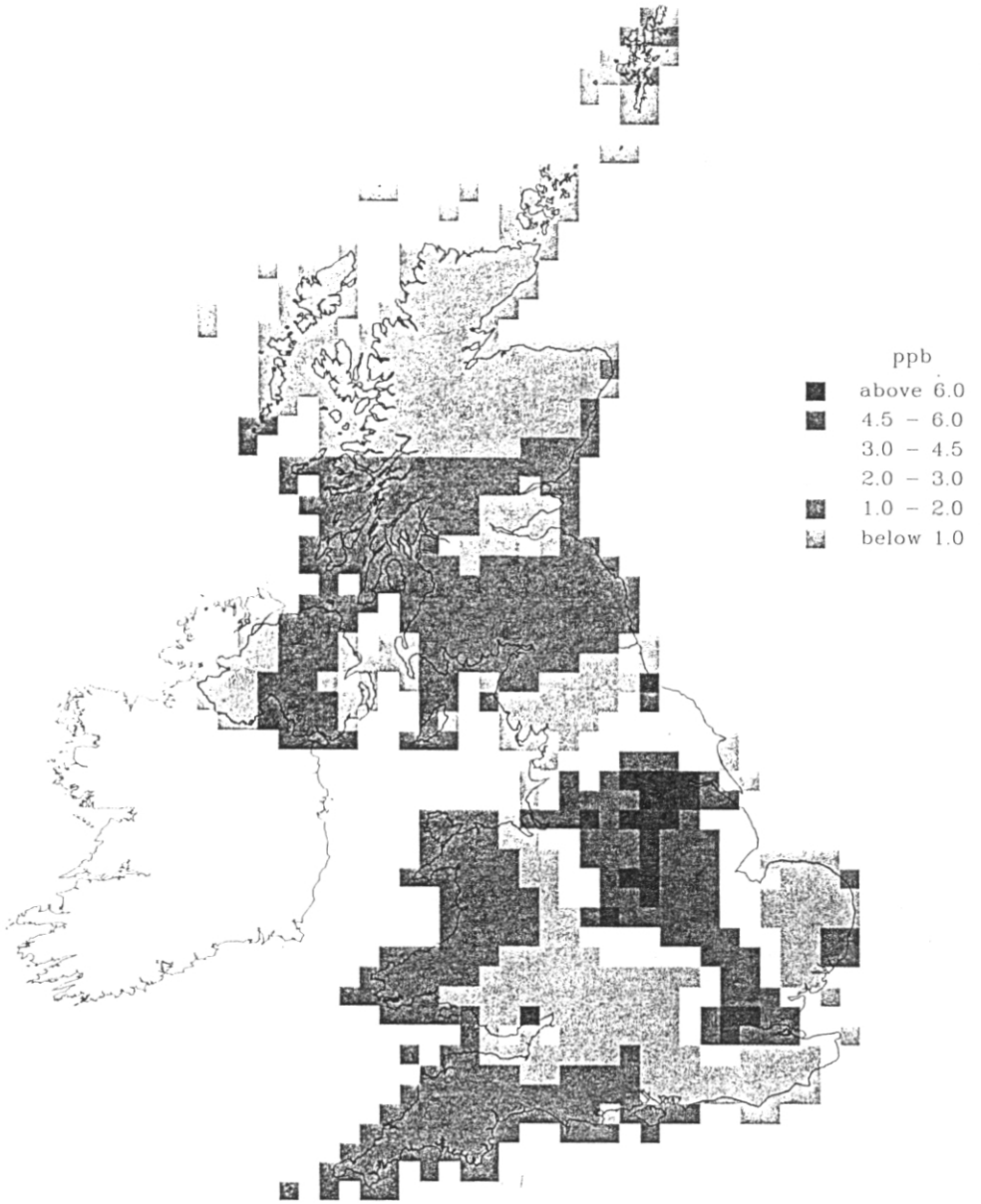

Figure 1: Mean annual concentration of SO_2 (ppb) for the UK during 1992-95.

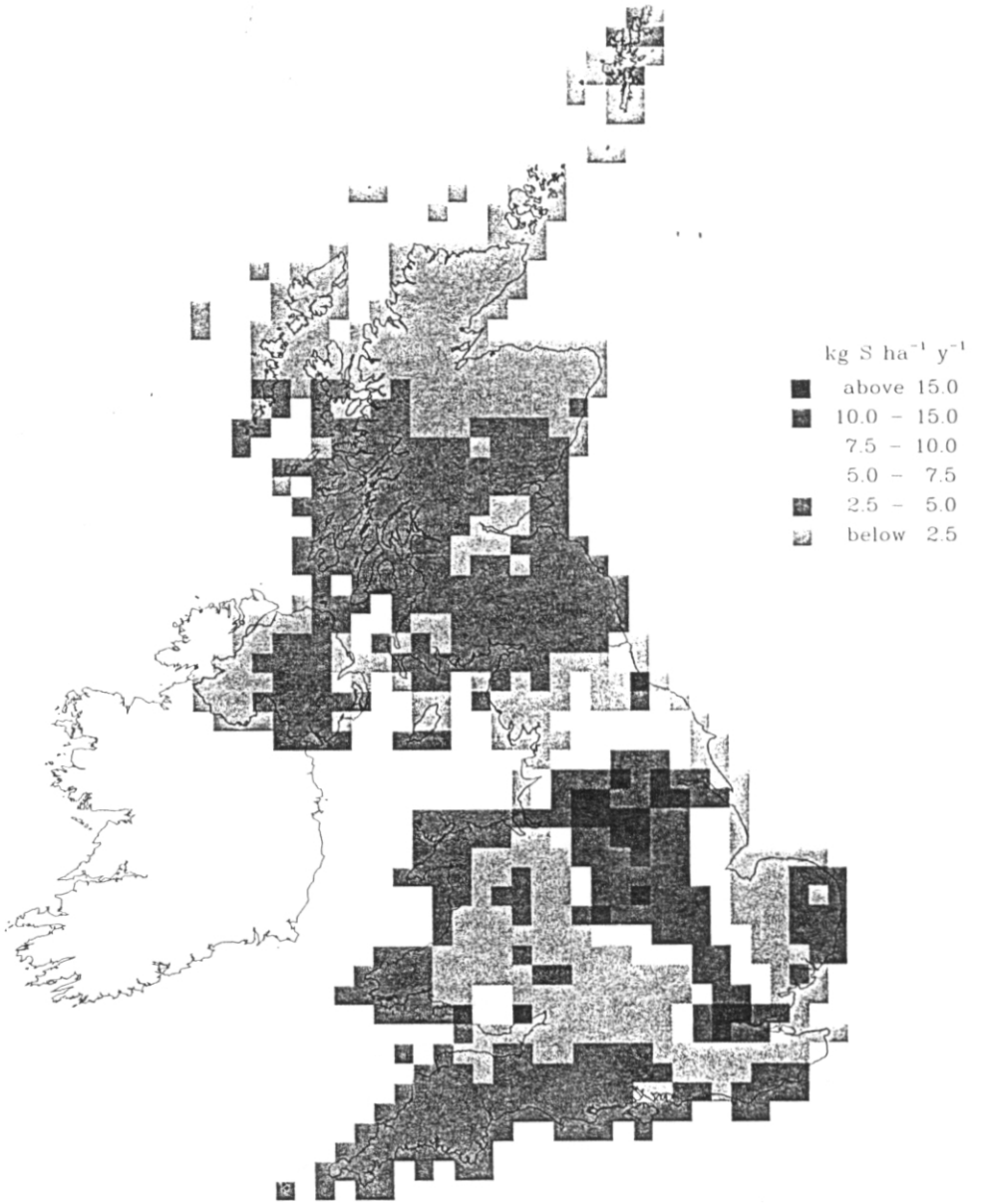

Figure 2: Mean annual dry deposition of SO_2 (kg S ha^{-1} yr^{-1}) for the UK during 1992-94

Figure 3: Kriged interpolation and standard error maps for 1995: (a) SO_2 concentration (rural sites only); (b) SO_2 standard error map; (c) SO_4^{2-} concentration in rainfall; (d) SO_4^{2-} error map.

The use of geostatistics in the interpolation and mapping of United Kingdom precipitation composition was described by Webster *et al.* (1991). The kriging method, being statistical in nature provides an estimate of the precision of the interpolated estimates. The precision, expressed as twice the standard error in the interpolated kriging estimates can be mapped in the same way as concentration. Figure 3 shows interpolated concentration and standard error maps for sulphur dioxide and for non-marine sulphate in rain for 1995.

However, the error estimates have to be interpreted with some caution. 'Simple' kriging has been used even though the marked trends in concentration in both sulphur dioxide and non-marine sulphate across the UK violates the assumption of constant expected value across the mapping domain. Tests of various methods for removing the trend showed that the concentration maps were not greatly affected by subtraction of the trend before kriging but that error maps were affected.

Using data from 1986 to 1988, Smith *et al.* (1995) performed a crude analysis of the likely levels of uncertainty in the predicted deposition of sulphur given the estimated uncertainty in the inputs. Following a similar analysis and assuming an error in the rainfall estimate to a 20 km square of ±10% with the kriging errors taken from the more recent maps, the uncertainty (i.e. an approximate 95% confidence interval assuming a normal distribution of residuals) in estimates of wet deposition is about ±20% rising to ±60% in high altitude, high rainfall areas. Using the kriging errors from the interpolated concentration maps of sulphur dioxide results in an uncertainty of ±30% of the annual mean concentration in central and eastern England rising to over 100% in the north-west of Scotland. Assuming an uncertainty in deposition velocity of ±20%, the overall uncertainty in dry deposition estimates would range from ±50% in central England to well over ±100% in many areas of Scotland. The major pathway for deposition varies in different areas of the UK with 70% as dry deposition in central England to 80% as wet deposition in north-west Scotland. Combining only the above two calculations to get an estimate of the uncertainty in total sulphur input to a 20 km square suggests a value of ±40% in central England increasing to ±70% on the west of Scotland and Wales.

The above estimates rely on an analysis of the likely causes of variation in the models combined with the error map from the kriging interpolation of the concentration field. For both wet and dry deposition, the kriging error is the dominant component of uncertainty and it increases substantially as the available information from the monitoring network reduces, as happens particularly in the north and west of Scotland. It should also be noted that the absolute values of deposition are smaller in remote areas and that large relative errors may still indicate quite small absolute errors. Until there is much more detailed monitoring of local variability, these estimates are indicative rather than accurate.

As discussed by Smith *et al.* (1995), the few catchment studies that are currently available suggest an uncertainty level of about ±30% in total sulphur input to areas of approximately the same size as a 20 km square. This value is rather less than the uncertainty level predicted above but indicates that uncertainties of about ±40% for most of the country rising to about ±60% towards the north and west are not unreasonable given current understanding of the processes. The error analysis which has been used here assumes symmetry.

3. Dry deposition measurements in the UK

Two sites have been developed to monitor SO_2 deposition in the UK. The first at Sutton Bonington has operated almost continuously since spring 1993. Ambient SO_2 concentrations, even in this polluted region of the UK are small, typically 3 ppb SO_2 and have declined substantially since the mid 1980s.

The data for Sutton Bonington are illustrated in Figure 4 which includes the monthly median concentrations for the 4 years as a time series. Also included on the figure is a sub-set of the data for the sector which winds from a northerly point (335° to 15°) in which the Ratcliffe Power Station stack is located at a distance of 5 km.

The concentrations with Ratcliffe Power Station upwind are larger, typically 5 to 12 ppb when averaged over the month.

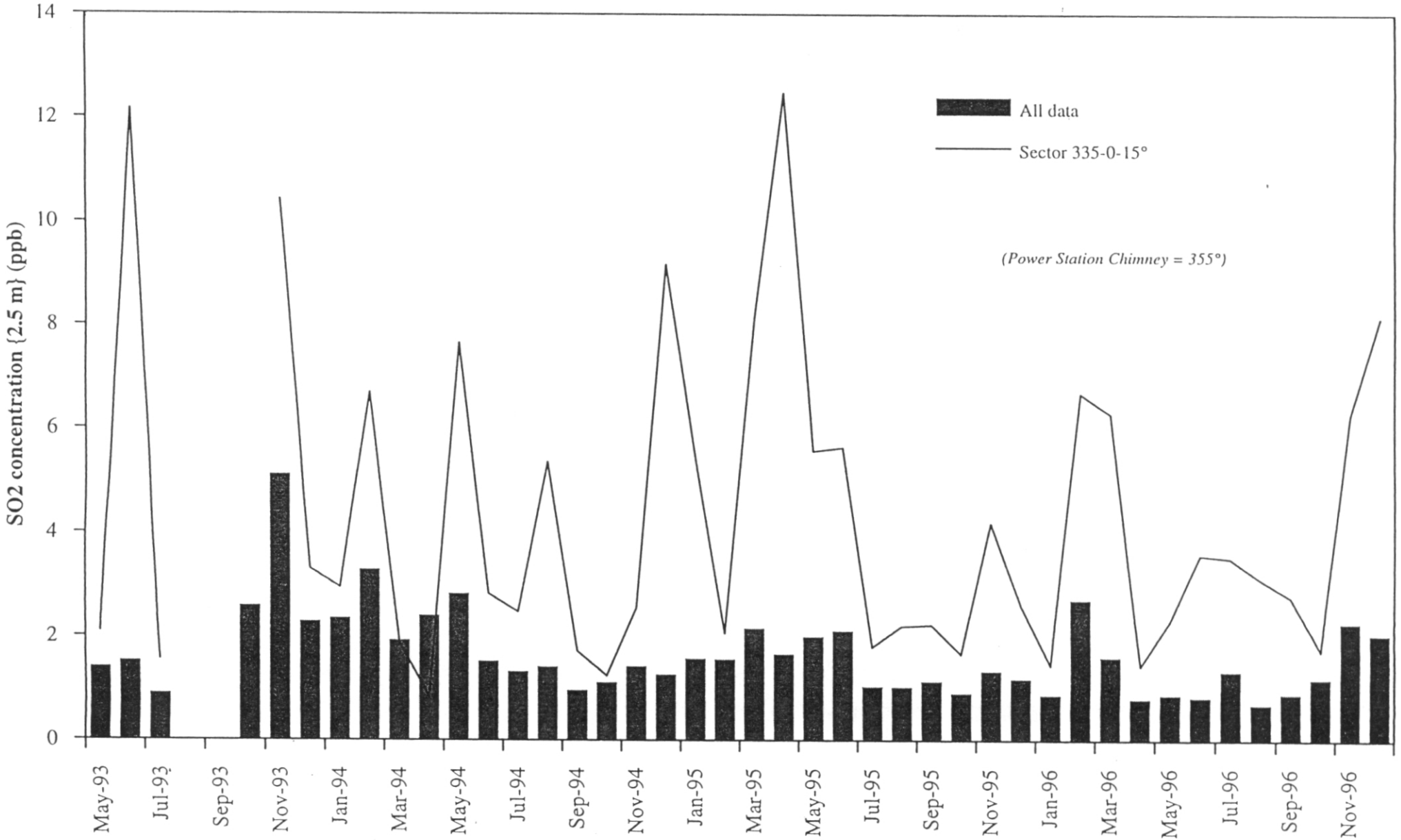

Figure 4: Monthly median SO_2 concentrations Sutton Bonington 1993-1996

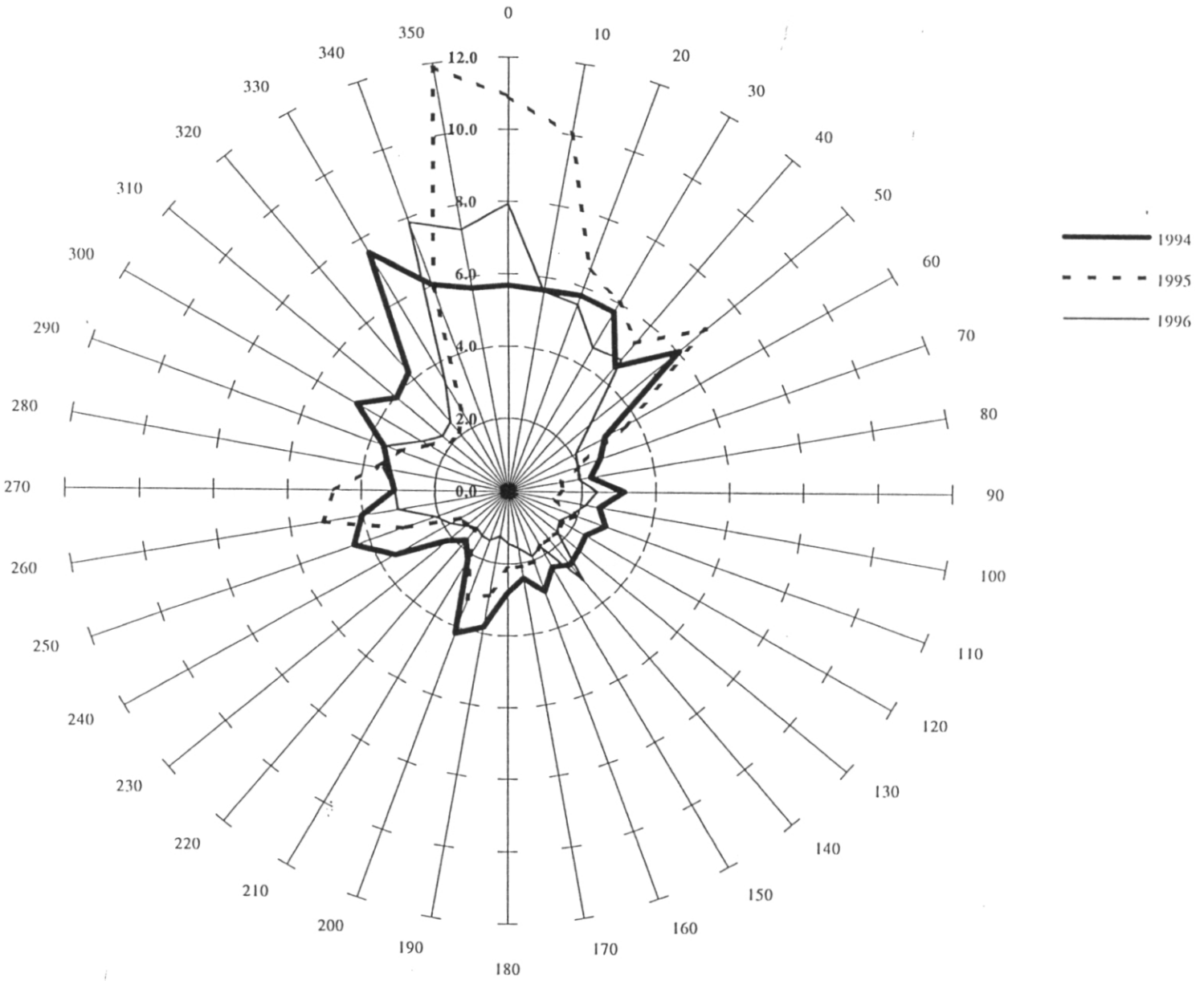

Figure 5: Wind direction dependence of SO_2 concentration (2.5 m) (ppb) at Autton Bonington 1993-96 (averages)

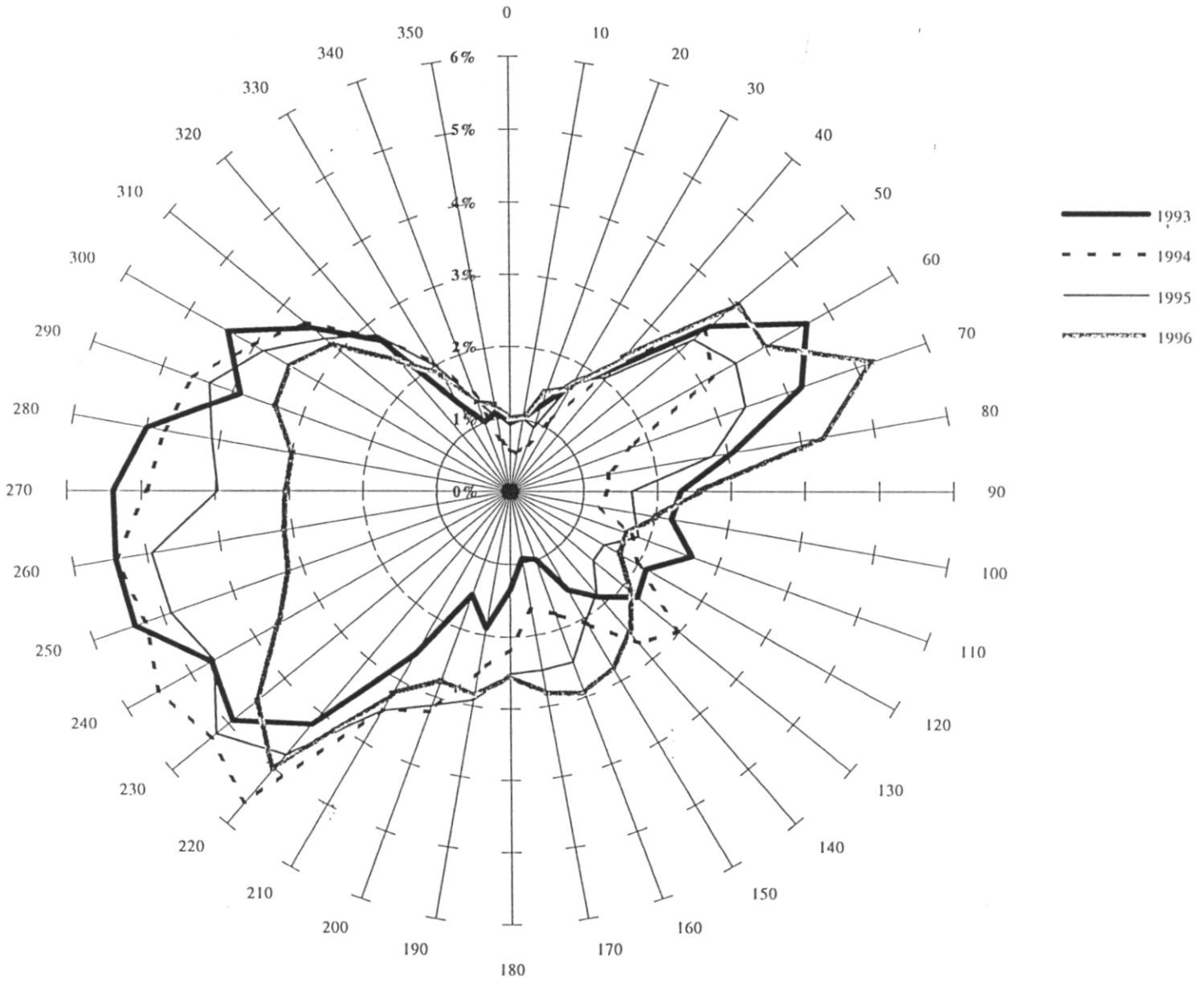

Figure 6: Frequency distributions of wind direction at Sutton Bonington 1993-96

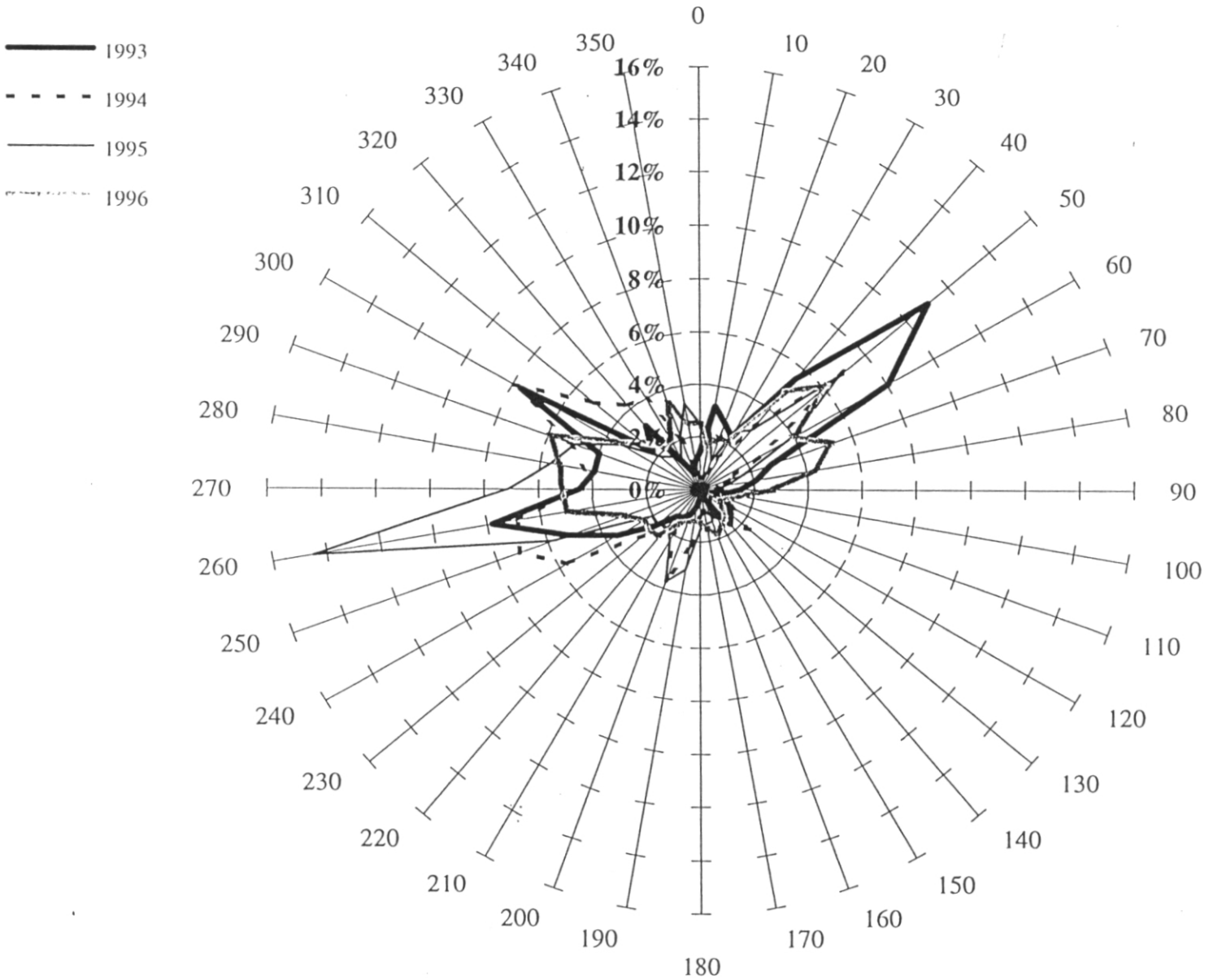

Figure 7: Contributions of the 36 (10°) wind direction sectors to the SO_2 advection fluxes over Sutton Bonington 1993-96

The large variability in concentrations with wind direction is illustrated in Figure 5 which shows that the largest concentrations occur with northerly winds and that the three years 1994, 1995 and 1996 are very similar. It is important however to show the frequency distribution of the wind direction (Figure 6), which shows the dominance of westerlies and that the northerly winds are uncommon (1% to 2% of the time only). Taking wind direction and SO_2 concentration together the advected flow of SO_2 over the monitoring station may be readily calculated. The analysis (Figure 7) shows that westerly and easterly winds dominate the SO_2 concentrations at the site with northerlies providing only 3% of the SO_2.

The concentrations and fluxes were monitored continuously and achieved 95% and 50% data capture respectively. While the monitoring equipment operates reliably and is frequently calibrated and subject to zero checks, the meteorological conditions and state of the field crops reduce the scope for satisfactory flux measurement. Nevertheless the 50% data capture covers almost the full range of atmospheric and surface conditions and allows testing and development of the model. A summary of the flux measurements is provided as Table 1 which shows an average deposition velocity of 5 mm s^{-1}, above 40% of V_{max}. The canopy resistance averages 126 s m^{-1}.

A time series of the data on fluxes shows over the 4-year period, quite small variations in deposition velocity relative to the shorter term variations (Figure 8).

The monthly averaged fluxes of SO_2 show a clear diurnal cycle with daytime maxima at typically 100 to 150 ng SO_2 m^{-2} s^{-1} and nocturnal values of 20 to 30 ng SO_2 m^{-2} s^{-1} (Figure 9).

The data may be compared directly with the model used to calculate UK dry deposition. The comparison between measurement and model of monthly fluxes at Sutton Bonington shows (Figure 10) that the model over-estimates dry deposition, sometimes by a factor of 2 or more. The cause is the assumption of leaf surface uptake rates, simulated using a fixed canopy resistance component for dry and wet surfaces. The model can be modified to fit the data quite readily but more analysis of the data to understand the precise cause is necessary.

Table 1: Summary table of measured SO_2 fluxes over agricultural land at Sutton Bonington for the Period Jan. 95 to Dec. 96.

Sutton Bonington	*N fluxes*	*% data capture*	*N concentr.*	*% data capture*	*RaH(1m) (s m-)*	*Rb SO2 (s m-1)*	*Flux SO2 (ng m-2 s-1)*	*SO2(1m) (ug m-3)*	*Vd mm s-1*	*Vmax mm s-1*	*Rc s m-1*
Jan-95	1130	76%	1478	99%	53.4	16.9	-41.37	3.73	11.1	14.2	20
Feb-95	1114	83%	1334	99%	48.0	16.4	-36.53	3.87	9.4	15.5	41
Mar-95	1240	83%	1488	100%	52.2	15.8	-31.39	5.50	5.7	14.7	107
Apr-95	683	47%	1155	80%	70.4	19.5	-32.21	3.84	8.4	11.1	29
May-95	586	39%	929	62%	60.7	22.9	-45.81	4.86	9.4	12.0	22
Jun-95	840	58%	1423	99%	47.4	25.2	-34.98	5.27	6.6	13.8	78
Jul-95	#N/A	#N/A	1484	100%	#N/A	#N/A	#N/A	#N/A	#N/A	#N/A	#N/A
Aug-95	#N/A	#N/A	1420	95%	#N/A	#N/A	#N/A	#N/A	#N/A	#N/A	#N/A
Sep-95	#N/A	#N/A	1105	77%	#N/A	#N/A	#N/A	#N/A	#N/A	#N/A	#N/A
Oct-95	1098	74%	1487	100%	63.9	24.3	-8.21	2.38	3.5	11.3	202
Nov-95	661	46%	1244	86%	72.3	21.7	-13.08	3.26	4.0	10.6	155
Dec-95	303	20%	1488	100%	59.7	25.7	-6.62	2.92	2.3	11.7	356
Jan-96	196	13%	1488	100%	29.6	20.3	-5.33	2.27	2.3	20.1	377
Feb-96	510	37%	965	69%	54.8	18.9	-26.73	6.64	4.0	13.6	175
Mar-96	218	15%	1488	100%	49.6	23.7	-15.44	4.04	3.8	13.6	188
Apr-96	712	49%	1392	97%	63.4	23.5	-2.56	2.09	1.2	11.5	728
May-96	689	46%	1488	100%	37.9	23.4	-6.86	2.19	3.1	16.3	258
Jun-96	959	67%	1426	99%	44.8	33.5	-5.85	2.11	2.8	12.8	283
Jul-96	1010	68%	1488	100%	41.6	29.2	-20.90	3.54	5.9	14.1	99
Aug-96	586	39%	1488	100%	46.9	30.1	-5.03	2.19	2.3	13.0	359
Sep-96	559	39%	1410	98%	53.8	23.6	-6.68	2.34	2.8	12.9	273
Oct-96	1051	71%	1482	100%	60.6	18.7	-17.10	2.87	6.0	12.6	88
Nov-96	1063	74%	1422	99%	59.1	19.1	-28.74	5.60	5.1	12.8	117
Dec-96	571	38%	1381	93%	58.3	26.4	-3.74	5.31	0.7	11.8	1334
Total	**15779**	**45%**	**32953**	**94%**	**59.1**	**19.7**	**-16.43**	**3.36**	**4.9**	**12.7**	**126**

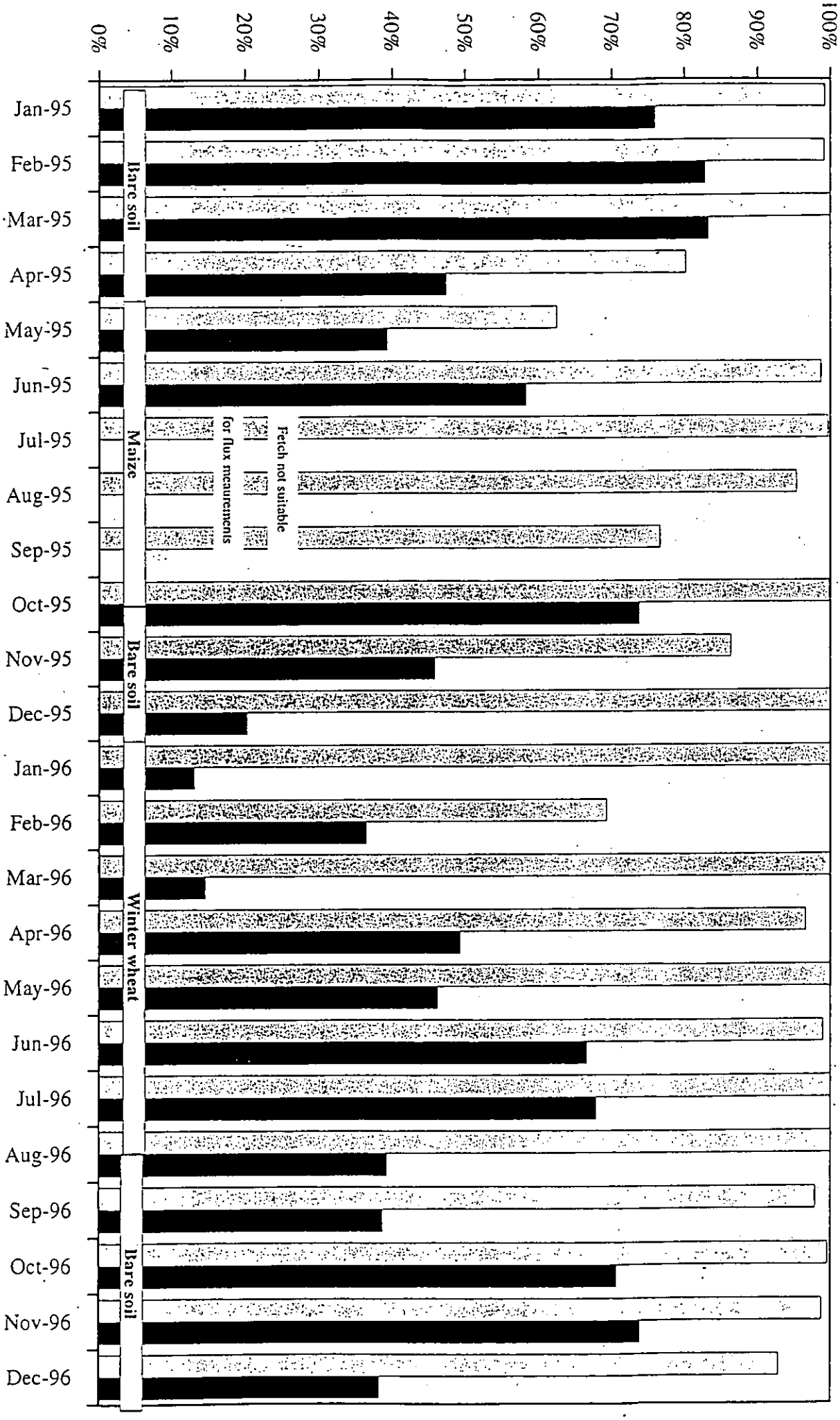

Figure 8: Sutton Bonington data coverage 1995-96.

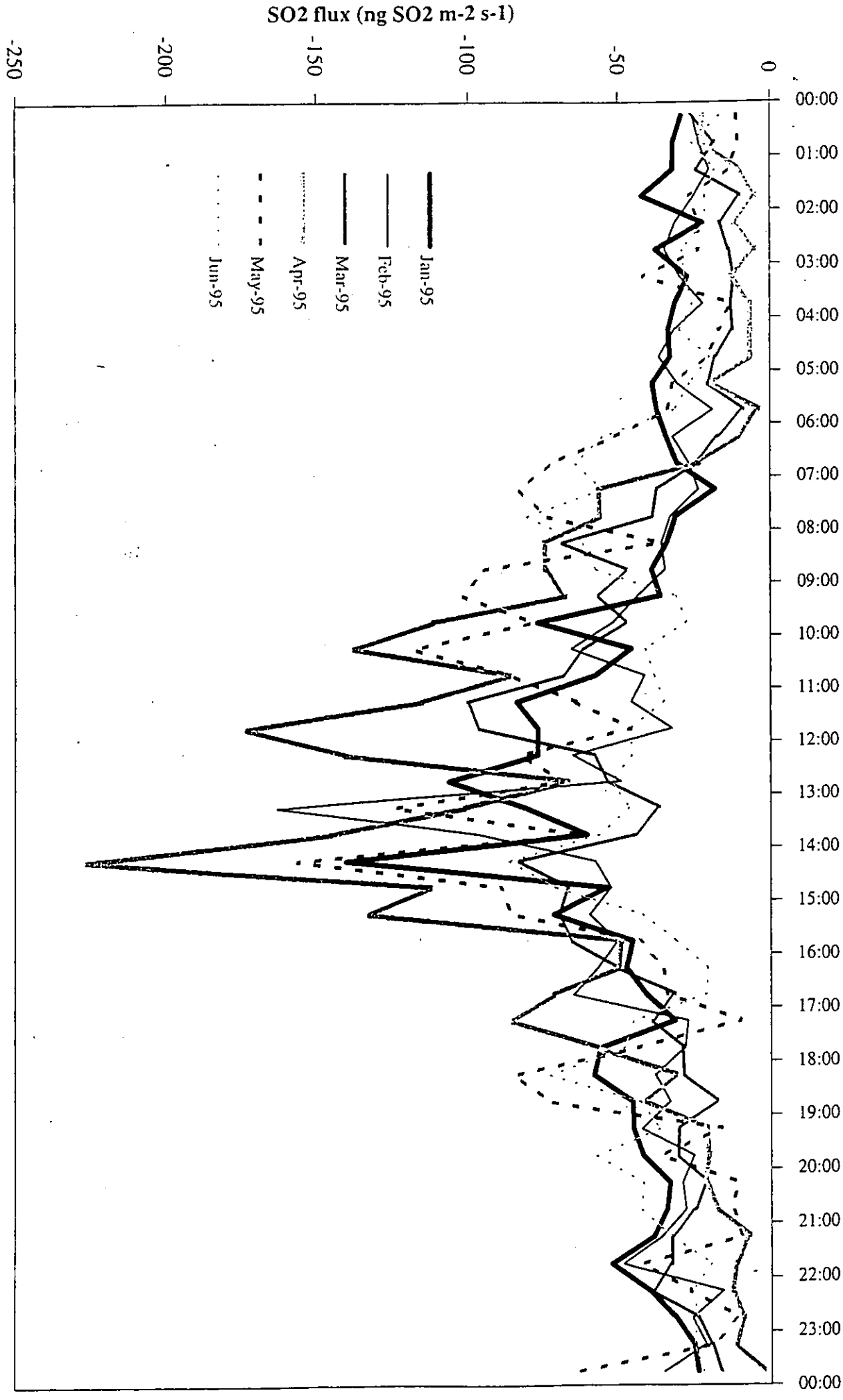

Figure 9: Monthly average diurnal cycles of measured SO_2 dry deposition fluxes at Sutton Bonington

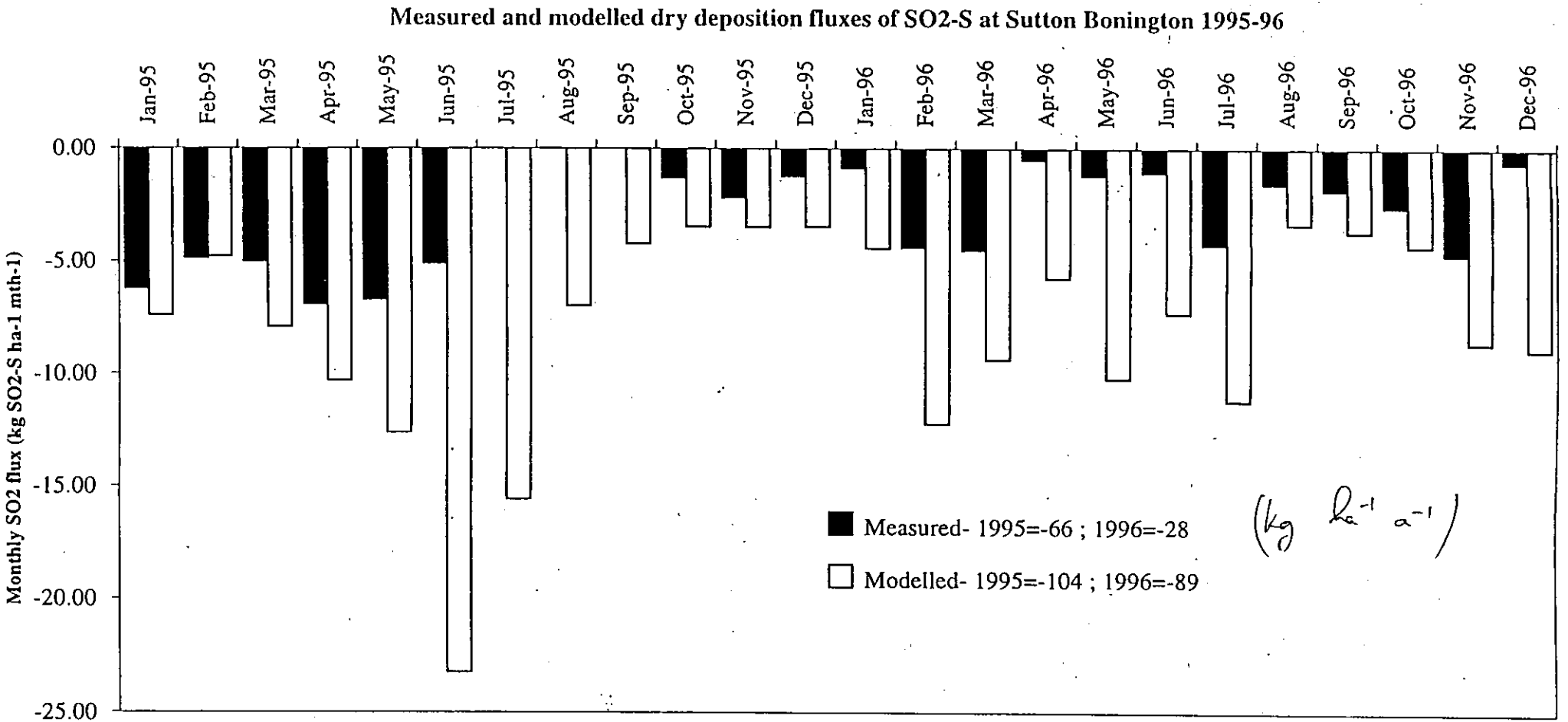

Figure 10: Measured and modelled dry deposition fluxes of SO_2-S at Sutton Bonington 1995-96

Auchencorth Moss

The site at an altitude of approximately 300 m contrasts with Sutton Bonington. The median concentration of SO_2 is 0.3 ppb and concentrations seldom exceed 10 ppb (0.2% time), (Figure 11 and Table 2). There are 3 distinct sources, the oil refinery at Grangemouth and Longannet Power Station some 60 km to the NW on a bearing of 310° (Figure 12) and the local urban areas of Edinburgh 25 km N and Penicuik to the east. The deposition velocity for SO_2 at Auchencorth moss is generally smaller than at Sutton Bonington and very much smaller than V_{max} (Figure 13). A summary of the data (Table 3) shows:

1. That despite being a windy upland site which is frequently wet with rain that deposition velocity is smaller than in the E. Midlands.
2. Mean deposition velocity is approx 3 mm s^{-1} (*cf* 5 mm s^{-1} at SB).
3. Aerodynamic resistances are smaller, as expected at the upland sites, 35 s m^{-1} v 50 s m^{-1} in agricultural lowlands of England.
4. Surface (or canopy) resistance at Auchencorth Moss is 300 to 600 s m^{-1} about a factor of 2 larger than at SB.
5. The measured deposition SO_2 rate for uplands smaller than that used in the UK model.

4. Validation of deposition model estimates against catchment hydrochemical budgets for a range of UK sites.

The combination of modelling, measurements and the number of components in the sulphur deposition budget makes estimates of uncertainty in the annual deposition a complex task. One of the most helpful validation checks on annual deposition is provided by hydrochemical budgets in catchments. Such measurements for conserved species (such as SO_4^{2-} and Cl^-) in 'water tight' catchments, in which there are no significant changes in internal storage, require only the outflow volume and composition to deduce inputs. The exercise is assisted at catchments with on-site wet deposition and cloud chemistry measurements, such as the Plylimnon catchments in central Wales and Beacon Hill in Leicestershire, to permit site specific modelling of deposition inputs.

Table 2: Concentration data Auchencorth Moss January-December 1995 (in ppbv)

	SO_2			O_3			NO			NO_2			NH_3		
N (1/2 hr)	17383	17382	17383	15794	15789	15790	17306	17300	17301	17066	17069	17058	11912	10462	12000
Average	1.00	0.92	0.86	26.38	25.34	24.58	0.35	0.34	0.35	3.11	3.04	2.98	1.59	1.39	1.14
Stdev	2.23	2.07	2.00	9.18	8.96	8.86	1.49	1.48	1.49	3.68	3.56	3.46	3.20	2.53	1.85
Median	0.34	0.32	0.30	26.81	25.78	25.06	0.09	0.09	0.09	1.83	1.81	1.79	0.73	0.68	0.57
Geomean	0.40	0.38	0.36	24.39	23.36	22.58	0.11	0.11	0.11	1.91	1.88	1.86	0.75	0.69	0.58
Geostdev	3.52	3.39	3.31	1.58	1.59	1.61	4.02	3.95	3.97	2.76	2.73	2.71	3.32	3.31	3.21

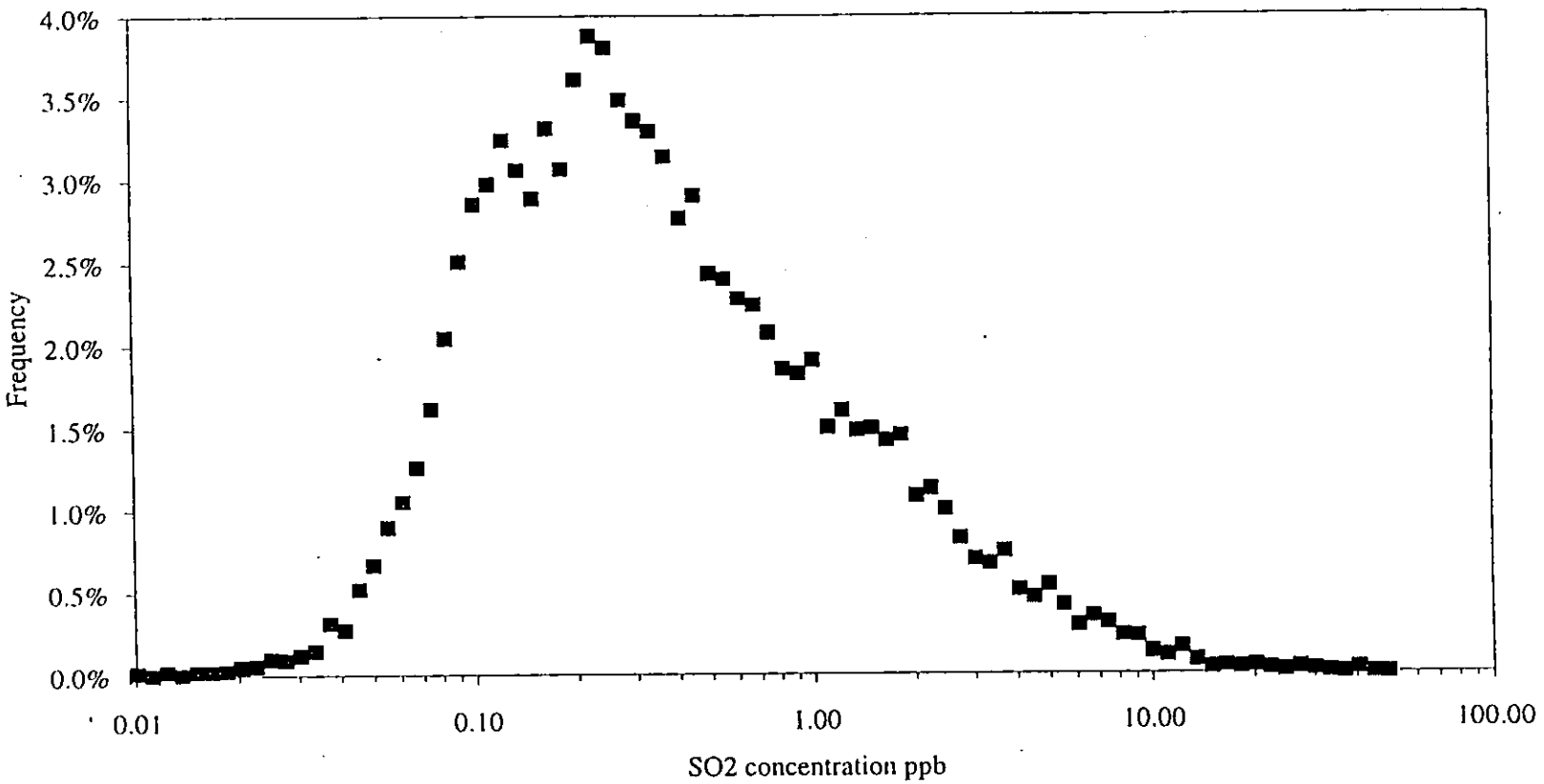

Figure 9: Frequency distribution of SO_2 concentration (1 m) Auchencorth Moss January-December 1995

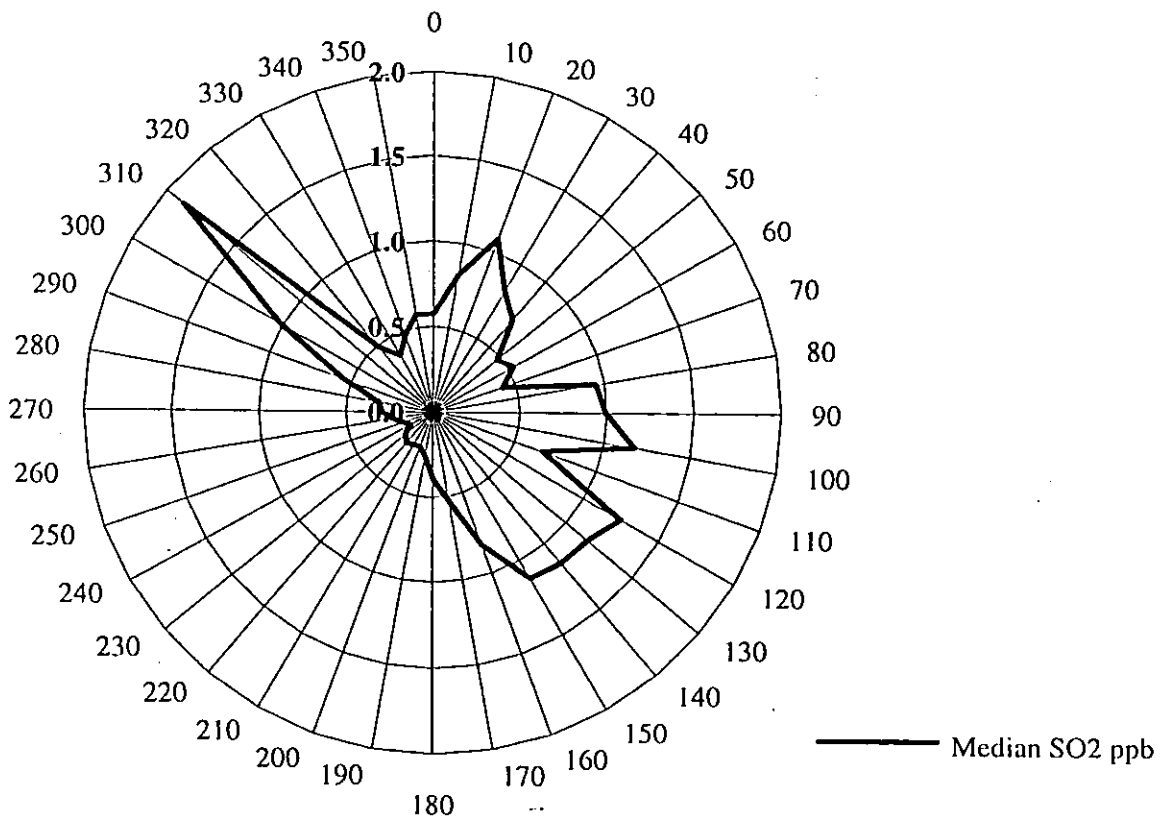

Figure 10: Sector dependence of SO_2 (1 m) Auchencorth Moss January-December 1995

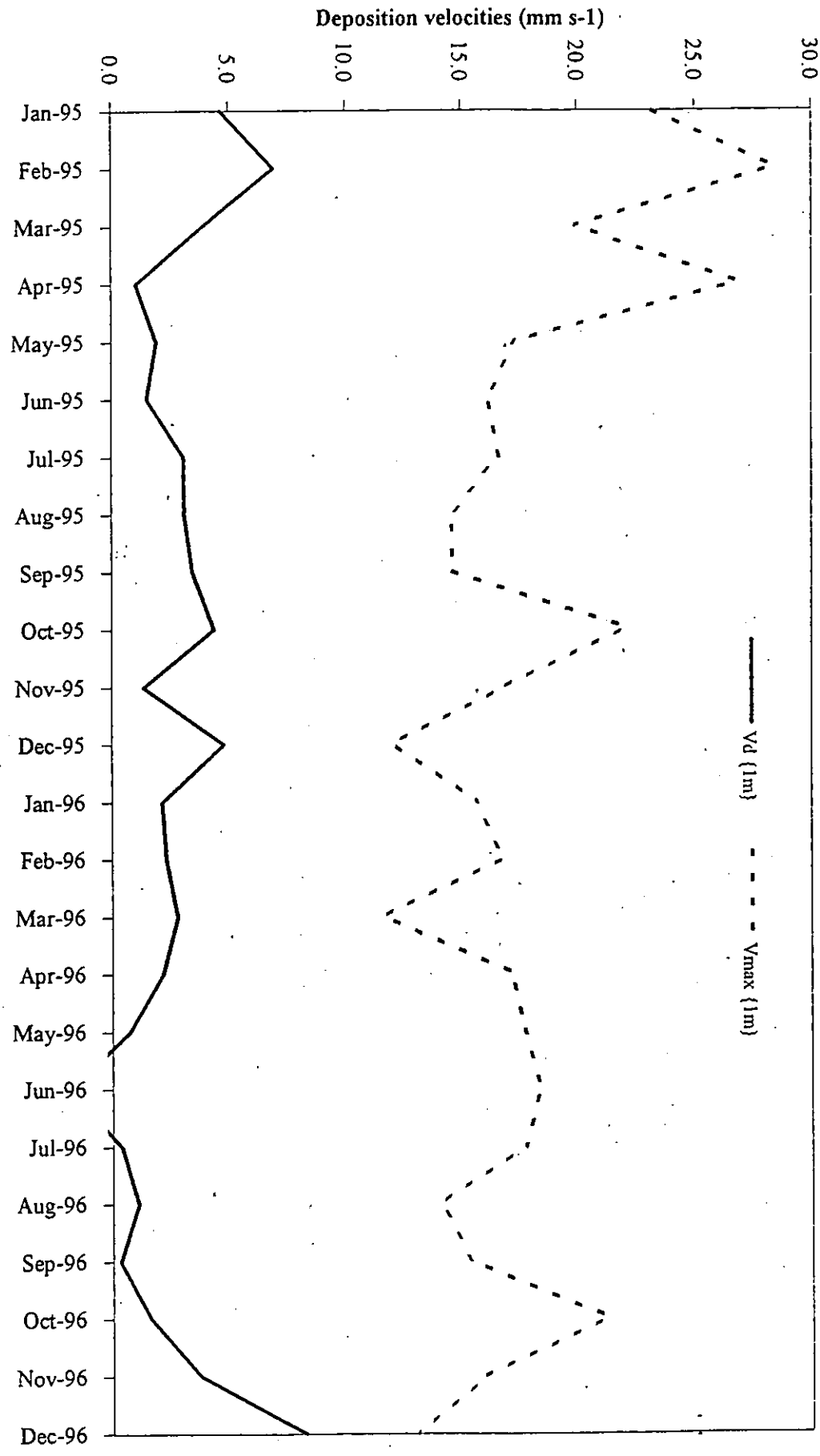

Figure 11: Monthly median deposition velocities for SO_2 at Auchencorth Moss 1995-96

Table 3: Summary table for SO_2 dry deposition at Autton Bonington and Auchencorth Moss 1995-96

Summary table for SO2 dry deposition at Sutton Bonington and Auchencorth Moss 1995-96								
		RaH(1m)	Rb SO2	Flux SO2	SO2(1m)	Vd	Vmax	Rc
		(s m-1)	*(s m-1)*	*(ng m-2 s-1)*	*(ug m-3)*	*mm s-1*	*mm s-1*	*s m-1*
Sutton	**1995**	56.7	20.7	-24.63	3.75	6.6	12.9	75
Bonington	**1996**	48.4	23.3	-10.01	3.00	3.3	13.9	228
Auchencorth	**1995**	36.9	18.4	-2.24	0.77	2.9	18.1	290
Moss	**1996**	40.9	19.9	-1.26	0.78	1.6	16.4	558

These two catchments are particularly helpful since in the Welsh uplands annual inputs are dominated by wet deposition, whereas in Leicestershire fry deposition is the substantially larger component, and those two catchments test the deposition inputs at opposing ends of the spectrum of partitioning of the inputs into wet and dry deposition. Table 4 summarizes the comparisons between measured sulphur outflow from Beacon Hill catchment for four years and the site specific inputs calculated using the same procedures used to national mapping. For the Welsh catchments (Table 5), the agreement between measured and modelled sulphur deposition is typically ± 10 to 15%, for catchments of 10-20 ha. Catchment measurement of S dry deposition shows good agreement with the modelled UK 40 km^2 mean S dry deposition. In the case of the other ions, the Na^+ budget shows good agreement for the Welsh catchments and this with the sulphur budgets in these uplands provides strong support for the simple model of wet deposition enhancement by seeder-feeder surveying.

Table 4: Sulphur deposition budgets for Beacon Hill Catchment 1984-88 based on UK deposition maps, modelled by Fowler and Smith (*Pers. Comm.*), and of measurements made on site (Black and Greenwood, *Pers. Comm.*)

	UK deposition maps 40 km^2 average S deposition, kg S ha^{-1} y^{-1}	Catchment model 0.622 km^2 S deposition kg S ha^{-1} y^{-1}	Catchment measurement 0.662 km S deposition kg S ha^{-1} y^{-1}
Wet	10.5	10.5	13.2
Cloud	0.2		
Dry	19.5	26.4	17.9*
Total	**30.2**	**36.9**	**31.2**

*Estimated from output-input 1984-88.

5. Trends in rural UK SO_2 concentrations 1979-1995

Sulphur dioxide has been measured at a number of rural sites since the 1960s as part of the National Survey of Smoke and Sulphur Dioxide. The more extensive Rural Sulphur Dioxide Monitoring Network was established in 1991 and hence few runs of measurements obtained by

Table 5: Total atmospheric inputs and streamflow solute outputs (kg ha^{-1} yr^{-1}) averaged over 2 years for the experimental catchments at Plynlimnon. (From Reynolds *et al.*, 1997).

Catchment	Total input	Output	Mass balance
Cyff			
N	22.6	3.9	+18.7
SO_4-S	21.9	33.3	-11.4
Cl	120.4	126.7	-5.3
Gwy			
N	23.6	3.5	+20.1
SO_4-S	22.7	24.8	-2.1
Cl	124.0	127.1	-3.1
Hafren			
N	33.0	6.1	+26.9
SO_4-S	29.0	29.1	-0.1
Cl	150.5	147.0	+3.5
C16			
N	24.9	-	-
SO_4-S	17.6	19.3	-1.7
Cl	76.1	78.4	-2.3
L12			
N	47.2	-	-
SO_4-S	36.0	30.4	+5.6
Cl	119.9	115.6	+4.3

consistent methods exist. However, from 1978, measurements consistent with those used in the current rural sulphur dioxide monitoring network commenced at Eskdalemuir in south-west Scotland. Annual mean concentrations over the period 1979 to 1995 are shown in Figure 13. There was a marked reduction in concentration in the mid-1980s. Annual mean concentrations after 1994 were around a quarter of the late 1970s values.

The marked decline in rural SO_2 concentrations between 1980 and 1993 is evident throughout the UK (RGAR, 1997). At many sites annual mean concentrations declined to values between 2 and 5 ppb from values between 10 and 20 ppb. Even in the East Midlands, the region currently showing the largest concentrations, the decline was approximately 70% between 1980 and 1995.

The critical level for SO_2 effects on crops is an annual mean of 11.25 ppb (CLAG, 1994). Although this level was widely exceeded in the 1970s, it is no longer exceeded in rural areas.

The downward trends in rural SO_2 concentrations appear to be greater than those in SO_2 emissions, leading to an apparent non-linear relationship between dry (and total) deposition and total SO_2 emissions. Explanations for any non-linearity must lie in either non-linear responses of removal processes to decreasing SO_2 concentrations or to changing patterns of ground-level concentrations in response to changing patterns in emissions. Bearing in mind that there have been changes in the relative contributions of different source types to SO_2 emissions over the 1980 to 1994 period, particularly in the domestic sector with the demise of coal burning, the changing pattern of SO_2 emissions is an obvious place to begin in the search for an explanation.

Low-level sources contribute much more significantly to ground-level concentrations compared with elevated sources. Hence, if either the emissions from local low-level sources decrease faster than total UK emissions, or the impact of industrial sources decreases as small plant switch to other fuels faster than large plant, concentrations at individual monitoring sites may decrease faster than total emissions. This is shown by measurements at the University of Nottingham agricultural research centre in Nottinghamshire. Figure 14 shows two pollution roses, for 1984 and 1994. As local dispersed sources declined, the pollution rose became dominated by relatively infrequent incursions of sulphur dioxide from Ratcliffe power station a few kilometres to the north. Flue gas desulphurisation equipment has since been fitted to this plant.

While it is likely that some of the observed decrease in rural SO_2 concentrations is due to the curtailment in domestic coal burning in rural communities since 1980, the magnitude of the decline is not consistent with the decrease in emissions reported in national inventories for the domestic sector. Indeed, there is no sector in the SO_2 inventory which can match the observed decline in rural concentrations over this period.

There are other possibilities which could help to explain the decrease in rural SO_2. There could have been an increase in the efficiency of dry deposition removal of SO_2 with declining SO_2 concentrations and/or the conversion efficiency of SO_2 to sulphate aerosol could have increased.

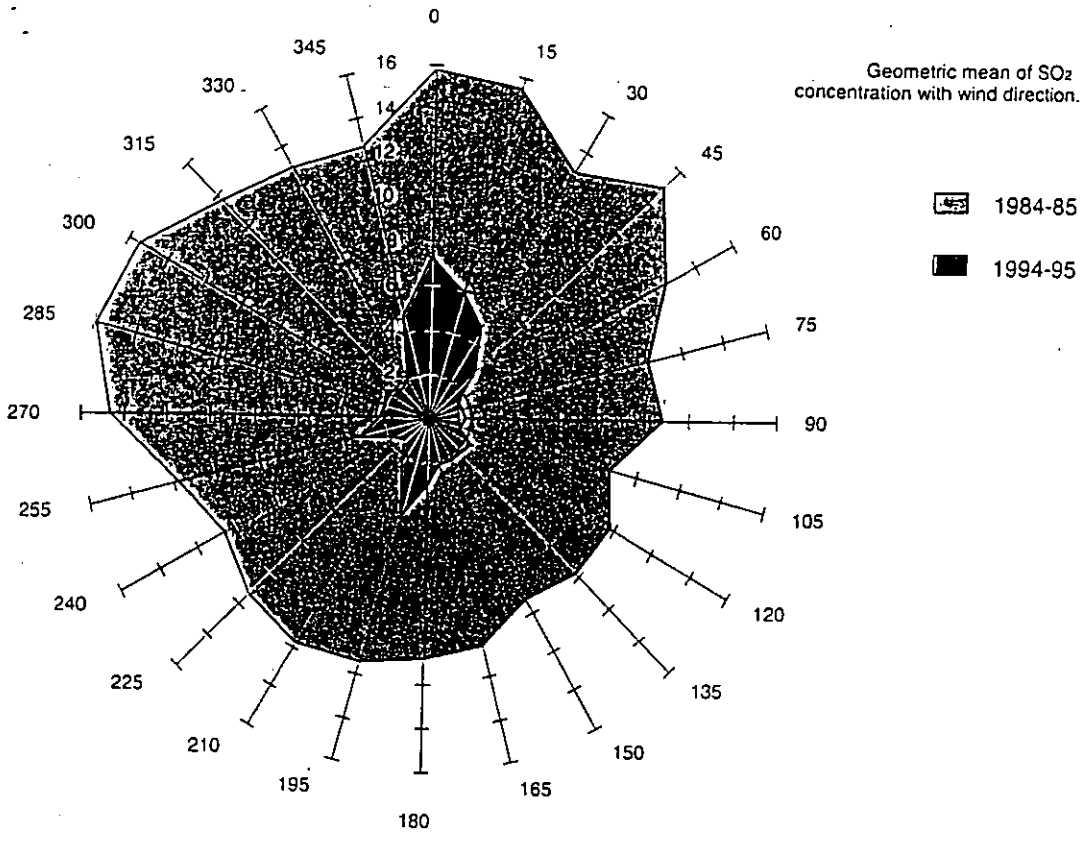

Figure 14: Pollution roses for sulphur dioxide at Sutton Bonington, 1984 and 1994 (ppb).

6. Development of dry deposition estimates in Northern Europe

Several national and a few European monitoring networks (e.g. EMEP) exist, where routinely ambient concentrations and wet deposition measurements are made of a range of components. Dry deposition is more difficult to measure and is not included in these networks. To date, measurements of dry deposition have been made in intensive field campaigns. Monitoring of dry deposition for some gases has only recently been reported (see Erisman and Draaijers, 1995, for an overview). The purposes of a deposition monitoring network should be:

- long-term measurements of dry and wet deposition of air pollution over several types of receptors under different pollution climates;
- identification of sites and ecosystems at risk;
- testing of existing long-range transport models;
- improvements of parameterisations currently used in long range transport;
- support of European policy on pollution control;
- trend analysis and monitoring of effectiveness of emission reduction plans, i.e. evaluation of critical load exceedances.

In January 1993 within the framework of the LIFE programme a project was financed whose aim was to develop a deposition monitoring method for air pollution of Europe (Erisman *et al.*, 1996). The project was co-financed by the Dutch, German and British governments and by the participating institutes. The project was executed by ITE (Institute of Terrestrial Ecology, UK), IFT (Institut für Tropostfahrenforschung, Germany), TNO (Institute of Environmental Sciences), ECN (Netherlands Energy Research Foundation), KEMA (Laboratory for Environmental; Research) and RIVM (National Institute of Public Health and the Environment), the Netherlands. The objective of the project was to develop a deposition monitoring method for air pollution of Europe to be used to extend existing European monitoring networks of air concentrations to provide deposition inputs on an ecosystem scale.

The locations of the three sites is shown in Figure 15. Some site specific characteristics are listed in Table 6. The sites were selected for their different land use type, surface characteristics and environmental climates. The northern Scottish site can be classified as humid background site. Speulder forest is characterised as a rough surface in a humid sea climate with moderate pollution

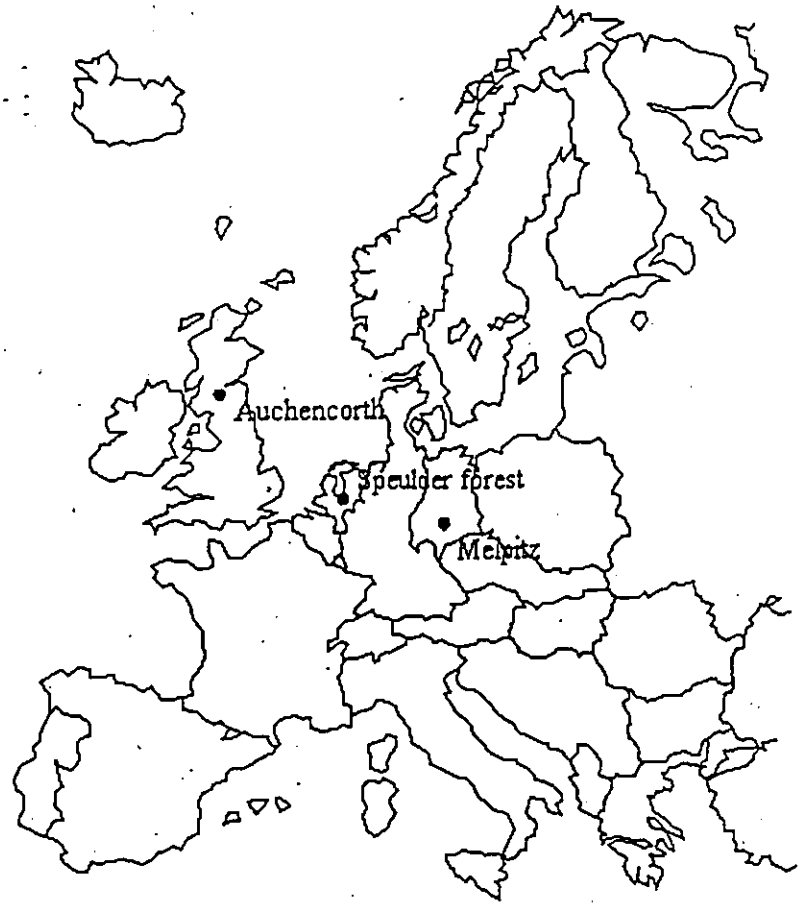

Figure 15: Location of the three monitoring sites.

levels, except for NH_3 concentrations which are high. Melpitz can be considered as a site with a land climate, with relatively high sulphur and particle loads. After installation, the monitoring equipment was almost immediately operational and performed well during the whole year, except for some minor problems. Data coverage for all three sites is quite high (80-90%).

Table 6: Site characteristics

	Auchencorth Moss Southern Scotland	Melpitz South Germany	Speulder Forest The Netherlands
Type of vegetation	peat bog/heath	grassland	Douglas fir
Vegetation height (m)	0.10	0.15	20-22
Roughness length (m)	0.01	0.04	1.20-3
Displacement height (m)	-	0.05	140
SO_2 measuring heights	3.0, 1.2, 0.5	5.3, 3.6, 2.5, 1.0	36, 32, 28, 24
NO_x measuring heights	3.0, 1.2, 0.5	5.3, 3.6, 2.5, 1.0	36, 32, 28, 24
NH_3 measuring heights	3.4, 1.4, 0.4	4.53, 1.80, 0.76	34, 28, 24
Particles	1.8	1.84	26
Acid gases	1.8	1.84	26
Wet-only collectors	1.5	1.5	1.5 (3 km from the forest)

Dry deposition of three gases (SO_2, NO_x and NH_3) was measured semi-continuously using the aerodynamic gradient technique. SO_2 concentration gradients were measured at 4 heights above the surface (three at Auchencorth Moss) using an UV pulsed-fluorescence monitor, a second monitor was used to measure continuously the concentration at the highest level; a change in SO_2 concentration during a measuring cycle could thus be detected and corrected for (Erisman *et al.*, 1993). The same measuring method was used for NO_x; concentrations were measured with a chemiluminescent monitor (Duyzer *et al.*, 1991). For NH_3 gradients were measured using three continuous-flow denuders (Wyers *et al.*, 1993). Meteorological measurements were made with a sonic anemometer and the Bowen ratio method, and a method to estimate annual average dry deposition fluxes was developed by Erisman *et al.* (1993, 1997).

The quality of the annual deposition estimate depends on the site characteristics and the quality of the measurements (selection) and of the surface resistance parameterisation. The surface resistance parameterisation is based on literature values and experimental data obtained at several experiments in Europe and was tested using the current (selected) data. These parameterisations are used together with concentration and meteorological measurements to infer dry deposition of other gases and particles.

The differences in pollution level is clearly identified in the different fluxes measured at the sites (Tables 7-9). The date show that generally the fluxes at Auchencorth Moss are lowest, except for components of sea salt origin (Na^+, Mg^{2+}, Cl^-). For these components, Melptiz has lower fluxes than Auchencorth Moss, as the result of the difference in distance to the sea. For these components wet deposition is higher than dry deposition. Speulder forest has the largest fluxes for all components, mainly as the result of the roughness of the forest and the resulting high dry deposition velocities. Heavy metal inputs are much lower than gases and base cations, because concentrations and also dry deposition velocities are much lower. It is remarkable that heavy metal fluxes are much higher in Auchencorth than in Melpitz. Heavy metal inputs mainly result from wet deposition and it must be emphasised that uncertainties associated with heavy metal inputs are large for all sites. At Auchencorth Moss, the background site, wet deposition is clearly the dominant input. At the higher pollution sites, dry deposition becomes more important, and generally dominates total deposition at the forest (high roughness) site.

The results of the LIFE experiment show that the methods implemented to monitor the deposition can be successfully applied. Routine application is possible and annual fluxes derived from the measurements are within reasonable accuracy. In addition to annual fluxes, improved surface exchange parameterisations can be derived from the gradient measurements. This is necessary because current parameterisations are inadequate to estimate dry deposition for the range of conditions and ecosystems of Europe. Both a comparison of hourly modelled and measured dry deposition values and a comparison between annual modelled and measured values revealed

Table 7: Average dry, wet and total deposition fluxes to Auchencorth Moss for the period 1.2.95 to 1.12.95 (mol ha^{-1} a^{-1}).

Component	Dry deposition	Wet deposition	Total deposition
SO_4	11	128	
SO_2	71		
SO_x	82		210
NH_4	24	173	
NH_3	177		
NH_x	201		374
NO_x	41		
NO_3	14	133	
HNO_3	18		
HNO_2	6		
NO_y	79		212
Cl	48	395	
HCl	17		
Cl_x	65		460
Na	53	378	431
Ca	3	36	39
K	2	25	27
Mg	6	50	56
Zn	0.13	0.907	1.037
Pb	0.03	0.065	0.095
Cd	0	0.003	0.003
Cu	0.02	0.144	0.164
Ni	0.01	0.006	0.016
Cr	0	0.016	0.016

Table 8: Average dry, wet and total deposition fluxes to the Melpitz site for the period 1.2.95 to 1.12.95 (mol ha^{-1} a^{-1}).

Component	Dry deposition	Wet deposition	Total deposition
SO_4	35	209	
SO_2	751		
SO_x	786		995
NH_4	47	336	
NH_3	518		
NH_x	565		901
NO_x	12		
NO_3	20	233	
HNO_3	55		
HNO_2	20		
NO_y	95		340
Cl	12	75	
HCl	31		
Cl_x	43		118
Na	21	70	91
Ca	32	49	81
K	5	24	29
Mg	3	14	17
Zn	0.35	4	4.35
Pb	0.03	0.08	0.11
Cd	0.001	0.004	0.005
Cu	0.047	0.05	0.097
Ni	0.005	0.03	0.035
Cr	0.006	0.01	0.016

Table 9: Average dry, wet and total deposition fluxes to the Speulder forest for the period 1.2.95 to 1.12.95 mol ha^{-1} a^{-1}).

Component	Dry deposition	Wet deposition	Total deposition
SO_4	106	291	397
SO_2	345		345
SO_x	452		742
NH_4	337	806	1143
NH_3	1277		1277
NH_x	1615		2420
NO_x	203		203
NO_3	267	374	641
HNO_3	97		97
HNO_2	69		69
NO_y	636		1010
Cl	214	855	1069
HCl	69		69
Cl_x	284		1139
Na	317	646	963
Ca	37	46	83
K	25	29	54
Mg	40	78	118
Zn	5	4	9
Pb	0.88	0.11	0.99
Cd	0.03	0.04	0.07
Cu	0.66	0.19	0.85
Ni	0.25	0.06	0.31
Cr	0.11	0.07	0.18

large differences. Models are used for policy development and abatement strategy using the critical load concept. it is necessary to improve models and reduce uncertainty in exceedance estimates. In this way optimal emission reduction measures can be determined, protecting ecosystem 'vitality' against lowest costs.

The monitoring data obtained at the three sites are not representative for the total range of ecosystems, climates and conditions in Europe. For a thorough evaluation of models and of policy development, more measuring sites of deposition are needed in Europe. For development and testing of deposition models, more 'intensive' sites, like the 3 LIFE sites are needed in different ecosystems in different conditions are needed. As the measurements show, it is not so much the ecosystem itself that regulates the deposition, as the roughness characteristics and the surface conditions (wetness, snow cover, etc.). It is therefore not necessary to cover most of the ecosystems, but only a few in strong varying conditions (dry weather, cold, etc, etc.), and check whether the assumptions on the roughness characteristics are right. About 10 sites in different climatic regions of Europe, including the three described here, need to be equipped with monitoring stations such as those developed in LIFE. For validation of the spatial representatively of model results, about 100 sites in Europe are needed, in addition to current concentration monitoring networks. For this, the development of low-cost deposition monitoring methods is necessary. These 100 low-cost sites, together with the maximum of 10 intensive sites should provide the minimum of measurements for model development and evaluation, and for policy evaluation in Europe, based on measurements. It is therefore strongly recommended to extend the current EMEP concentration and wet deposition network with dry deposition facilities. The EMEP monitoring data, together with the EMEP and EDACS model, then provides good quality information for policy development for reducing excessive inputs of pollutants to ecosystems in Europe.

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