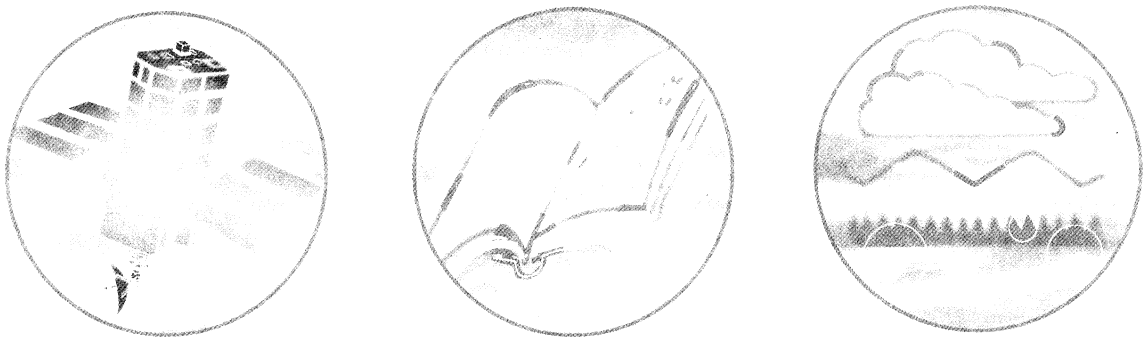
Linearity of Sulphur Chemistry in the HARM Atmospheric Transport and Deposition Model



Research and Development

Technical Report
P275

ENVIRONMENT AGENCY

Linearity of Sulphur Chemistry in the HARM Atmospheric Transport and Deposition Model

R&D Technical Report P275

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Statement of use
This report summarises the findings of research into the linearity of the sulphur chemistry in the Hull Acid Rain Model (HARM). The information in this document is for use by EA staff and others involved in the assessment of the release and deposition of acid species in the atmosphere.

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EXECUTIVE SUMMARY

1. Acid deposition from the atmosphere may result in damage to terrestrial and aquatic ecosystems. The principal anthropogenic emissions responsible for acid deposition are sulphur dioxide and nitrogen oxides. Ammonia, arising mainly from agriculture, also plays a part. Of these sulphur dioxide has traditionally had the major role.

2. In the UK, large point sources, principally oil and coal fired power stations, are responsible for more than 75% of sulphur dioxide emissions. These sources are regulated by the Agency as Part A processes under the terms of the Environmental Protection Act 1990.

3. The Hull Acid Rain Model (HARM) is used extensively to estimate the deposition of acidity. In particular it is used to assess the effects of different scenarios for combinations of emission limits for the major sources of sulphur dioxide.

4. It is common practice to assume that deposition is proportional to the emission of sulphur dioxide, that is to say that a reduction in emissions will result in a proportional reduction in deposition.

5. This assumption is based on the linear chemistry which is used in the HARM model but has not been tested in practice.

6. The purpose of the work described in this report is to test the assumption by using the model and scaling the deposition field by the magnitude of different emission and also by using the model with separate individual emissions. The two sets of results are compared to check whether they are the same.

7. It is shown that for both single sources and groups of sources together, the model does behave in a linear manner. This is a valuable feature of the modelling because the results can be manipulated to cover a range of scenarios without the need for multiple runs of the model.

8. This result is accurate only so far as the assumptions of linear chemistry are justified. It must be borne in mind that in practice there are situations when this may not be the case.

Keywords
Deposition, linearity, acid, sulphur.

Associated Reports

The following Technical Reports and Project Records have also been produced under this project:

Technical Reports:

P276	The implications of non-linear nitrogen chemistry in the HARM model for use by the Environment Agency.
P246	Review of dynamic models for catchment and surface water acidification.

Project Record:

P4/i019/01A	Working records of air dispersion modelling and environmental assessments for acid gas emissions from power stations and oil refineries.

1. BACKGROUND

Output from the Hull Acid Rain Model (HARM) has been used extensively by the Institute of Terrestrial Ecology (ITE) Monks Wood to determine the impacts of acid deposition on sensitive ecosystems in the UK from sources across Europe and the UK in terms of critical loads (CL) exceedance. As part of work for the Environment Agency, there has been particular emphasis on major point sources, especially coal and oil fired power stations, whose emissions to air have to be authorised by the Agency as Part A processes under the terms of the Environmental Protection Act 1990 (Murley, 1996). In the light of commitments under the European Community Large Combustion Plant Directive (1988) and the United Nations Economic Commission for Europe Oslo Protocol (1994), changes in emissions of SO_2 and the resulting total (wet + dry) S deposition, have been the primary interest. As a receptor orientated Lagrangian model HARM is ideally suited for source attribution (Hough and Eggleton, 1986) and footprints of both area and point sources can be produced. The different source types (group or individual) can be ranked either in terms of their total S deposition or in terms of their contribution to critical loads exceedance in a particular part of the country. By scaling deposition fields for individual sources, or groups of sources, the effectiveness of different emission control scenarios may be assessed (e.g. Metcalfe and Whyatt, 1994).

In order to assess the effects of changes in authorised emissions limits for power stations in England and Wales ITE have been scaling single source and multiple source sulphur deposition fields produced by HARM 7.2. The deposition fields have been scaled on the basis that the sulphur chemistry in HARM is linear, that is to say that a given increase or decrease in emissions will result in a proportional increase or decrease in deposition. The aim of this report is to test this assumption of linearity and investigate whether identical results are produced by scaling existing deposition fields and by running the model with a specific set of emissions.

2. SULPHUR CHEMISTRY IN HARM

The representation of sulphur in HARM has been described in some detail in Metcalfe *et al.* (1995), with the chemistry still largely as set out in Buckley-Golder and Derwent (1986). In the model, all sulphur is emitted as SO_2 and it is assumed that its conversion to SO_4 is driven by hydroxyl oxidation at a basic rate of 1% per hour. In order to provide a simplified representation of the seeder-feeder effect the standard oxidation rate is scaled by units of 250 mm rainfall. There is no oxidant limitation and the rate does not depend upon pH. Dry deposition of S is generated through land-use dependent deposition velocities for SO_2 which are independent of the concentrations of other pollutants in the atmosphere. Wet removal is through scavenging coefficients (based on constant drizzle) with rates again scaled by rainfall (in units of 500 mm) to represent orographic enhancement.

3. METHODOLOGY

Two versions of HARM 7.2 have been used. A multi-source version to produce deposition footprints for European sources and industrial and low level UK sources and a single source version to produce deposition footprints for individual power stations in England and Wales. The effects of a large source (power station) may, therefore, be viewed in isolation (e.g. deposition combined with critical load map to determine areas of exceedance from the power

station alone) or as a component of deposition from all sources (e.g. percentage contribution to critical loads exceedance).

The original deposition prints for individual power stations were based on 1991 emissions data supplied by HMIP. Each power station creates its own unique spatial pattern of deposition with higher levels of deposition close to source and lower levels of deposition with increasing distance from source. The unique pattern of each print is the product of spatial variations in wet and dry removal processes. These prints have been scaled upwards or downwards to reflect increases or decreases in emissions at a power station relative to some base year (e.g. Brown *et al.*, 1995). The spatial pattern remains basically the same, the amount of deposition per grid cell increasing or decreasing in direct relation to the change in emissions.

4. SINGLE SOURCE EVALUATION

In order to determine whether the assumption of linearity applies to the individual deposition prints we simply need to compare actual deposition prints for a particular source (generated by model runs employing specific emissions) with deposition prints produced from scaled model output.

For the purposes of this report a series of new *scenario specific* deposition footprints have been generated for Drax power station (North Yorkshire). These new footprints have been compared against prints produced through scaling the original Drax footprint supplied to ITE in 1994. The original deposition footprint for Drax power station is depicted in Figure 1a. The scaling factors required to convert the original print into a range of different scenarios are as follows:

Scenario	**Emission (k tonnes SO_2)**	**Scaling factor**
1991	236.5	1.0
1994	160.8	0.679
2010	24.7	0.104
Authorisation	324	1.369
National plan	270	1.141

In order to assess whether there are significant differences between modelled and scaled deposition footprints we need to compare a) the deposition budgets of the modelled and scaled fields and b) estimates of deposition from both the modelled and scaled fields at a range of sites across the UK (Figure 1b).

Area-weighted deposition budgets for scenarios generated by specific model runs and by deposition print scaling are as follows:

Clearly, the deposition budgets are very similar and small differences in the budgets may be attributed to rounding errors in the scaling calculation. Modelled and scaled estimates of deposition at the chosen sites (Figure 1b) are listed in Table 1. Differences between modelled and scaled deposition estimates at the sites are again very small and may also be attributed to rounding error. There is clearly no spatial variation in the difference between modelled and

Scenario	Deposition (tonnes S): Specific model run	Scaled deposition field
1991	17028	17028
1994	11579	11578
2010	1779	1778
Authorisation	23380	23378
National plan	19441	19440

scaled deposition at the representative sites (discrepancies are of the same magnitude irrespective of proximity to source) hence we can state with confidence that the assumption of linearity holds for the single source implementation of HARM and that scaled deposition prints are not significantly different to deposition prints produced from model runs employing specific emissions.

5. MULTIPLE SOURCE EVALUATION

The multiple source (standard) version of the model was tested in the same manner as the single source version of the model. A series of new *scenario-specific* deposition footprints were created for low level sources in the UK (Figure 2a) and for anthropogenic and natural sources across Europe (Figure 2b). These were then compared against scaled deposition footprints. The scenarios and scaling factors were as follows:

Scenario	Emission (k tonnes SO_2)	Scaling factor
UK low level 1991	773.2	1.0
UK low level 50% reduction	386.6	0.5
UK low level 75% reduction	193.3	0.25
European 1990	38088	1.0
European 50% reduction	19044	0.5
European 75% reduction	9522	0.25

Once again, the deposition budgets for the modelled and scaled scenarios showed a high level of agreement with minor discrepancies attributable to rounding error. The results of the site specific comparison for UK sources (Table 2) and European sources (Table 3) also suggest that differences between the modelled and scaled fields are minimal and well within the limits of rounding error. Hence we can also state with confidence that the assumption of linearity holds for the multi-source implementation of HARM and that scaled deposition prints are not significantly different from deposition prints produced from model runs employing specific emissions.

6. CONCLUSIONS

This report has demonstrated that the sulphur chemistry within the single source and the multiple source versions of HARM is linear. Sulphur deposition footprints supplied to ITE may simply be scaled upwards or downwards by an appropriate factor to reflect increased or

decreased emissions at a single source or range of sources. This is clearly a very valuable feature in terms of the ease with which model output can be manipulated, obviating the need for multiple model runs. It should be borne in mind, however, that although the S chemistry in HARM is linear, this is unlikely to be true in the real world; for example, liquid phase oxidation of SO_2 by H_2O_2 is highly non-linear. Changes in cloud water pH and oxidant availability will change the rates at which sulphate aerosols are formed affecting transport distance and the relative importance of wet and dry deposition. The amount of S dry deposited may also be affected by changes in NH_3 concentrations, although there is no direct evidence for this in the available NH_3 data (RGAR, 1997). A further consideration is that HARM is a coupled chemistry model, with sulphur tied in to the ammonia cycle through the production of ammonium sulphate aerosol, so simple assumptions of linearity would not apply in a multi-pollutant emissions reduction scenario.

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TABLE 1: SINGLE SOURCE VERSION (DRAX POWER STATION ONLY)

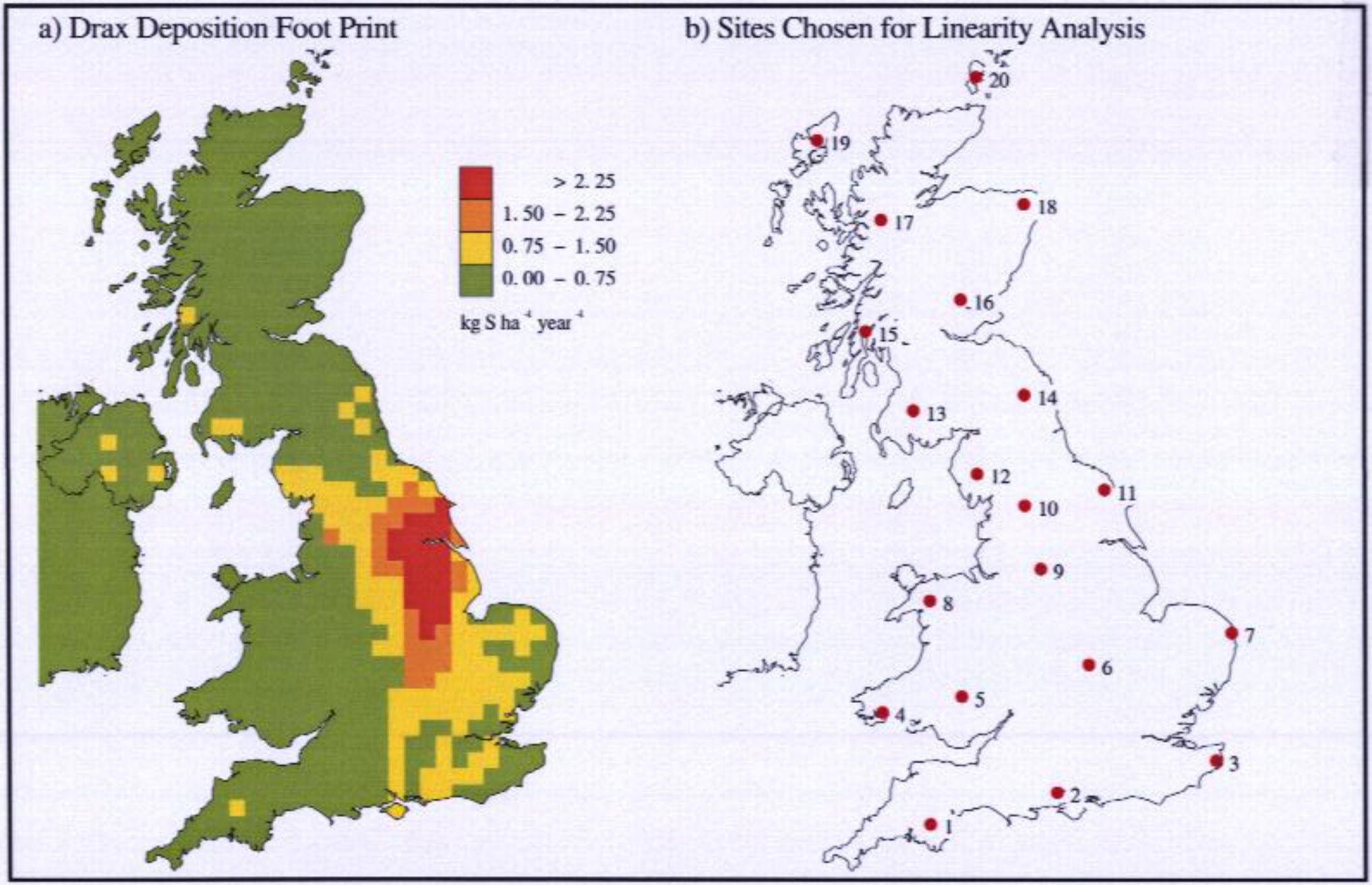
	Model 1991*	Model (1994)	Scaled (1994)	Model (2010)	Scaled (2010)	Model (Auth)	Scaled (Auth)	Model (Plan)	Scaled (Plan)
01 DARTMOOR NATIONAL PARK	**0.225**	0.153	0.153	0.024	0.023	0.310	0.308	0.258	0.257
02 NEW FOREST	**0.539**	0.367	0.366	0.056	0.056	0.740	0.738	0.616	0.615
03 Kent Downs	**0.131**	0.089	0.089	0.014	0.014	0.179	0.179	0.149	0.150
04 PEMBROKESHIRE COAST NATIONAL PARK	**0.277**	0.188	0.188	0.029	0.029	0.379	0.379	0.316	0.316
05 BRECON BEACONS NATIONAL PARK	**0.676**	0.460	0.460	0.071	0.071	0.925	0.926	0.771	0.772
06 Northampton	**2.163**	1.471	1.471	0.226	0.226	2.964	2.963	2.471	2.469
07 NORFOLK BROADS NATIONAL PARK	**0.476**	0.324	0.324	0.050	0.050	0.653	0.652	0.544	0.543
08 SNOWDONIA NATIONAL PARK	**0.124**	0.084	0.084	0.013	0.013	0.170	0.170	0.141	0.142
09 PEAK DISTRICT NATIONAL PARK	**1.154**	0.784	0.785	0.121	0.121	1.581	1.581	1.318	1.317
10 YORKSHIRE DALES NATIONAL PARK	**0.946**	0.643	0.643	0.099	0.099	1.295	1.296	1.080	1.080
11 NORTH YORK MOORS NATIONAL PARK	**1.059**	0.720	0.720	0.111	0.111	1.450	1.451	1.209	1.209
12 LAKE DISTRICT NATIONAL PARK	**1.229**	0.836	0.836	0.129	0.128	1.684	1.684	1.404	1.403
13 Galloway	**0.636**	0.432	0.432	0.066	0.066	0.870	0.871	0.726	0.726
14 NORTHUMBERLAND NATIONAL PARK	**0.578**	0.393	0.393	0.061	0.060	0.792	0.792	0.660	0.660
15 Kyles of Bute	**0.082**	0.056	0.056	0.009	0.009	0.113	0.112	0.094	0.094
16 Perth	**0.403**	0.274	0.274	0.042	0.042	0.551	0.552	0.460	0.460
17 Glen Affric	**0.335**	0.228	0.228	0.035	0.035	0.459	0.459	0.383	0.382
18 Fyvie	**0.286**	0.195	0.194	0.030	0.030	0.392	0.392	0.327	0.327
19 Isle of Lewis	**0.155**	0.105	0.105	0.017	0.016	0.213	0.212	0.177	0.177
20 The Orkneys	**0.181**	0.123	0.123	0.018	0.019	0.248	0.248	0.206	0.207

TABLE 2: MULTI-SOURCE VERSION (UK SOURCES ONLY)

		Model 1991*	Model (50%)	Scaled (50%)	Model (25%)	Scaled (25%)
01	DARTMOOR NATIONAL PARK	**1.944**	0.972	0.972	0.486	0.486
02	NEW FOREST	**2.542**	1.271	1.271	0.636	0.635
03	Kent Downs	**2.763**	1.381	1.381	0.691	0.691
04	PEMBROKESHIRE COAST NATIONAL PARK	**1.782**	0.891	0.891	0.446	0.446
05	BRECON BEACONS NATIONAL PARK	**2.527**	1.264	1.264	0.631	0.632
06	Northampton	**3.049**	1.525	1.524	0.763	0.762
07	NORFOLK BROADS NATIONAL PARK	**1.936**	0.968	0.968	0.484	0.484
08	SNOWDONIA NATIONAL PARK	**3.807**	1.904	1.903	0.952	0.952
09	PEAK DISTRICT NATIONAL PARK	**4.249**	2.124	2.124	1.062	1.062
10	YORKSHIRE DALES NATIONAL PARK	**3.223**	1.611	1.611	0.806	0.806
11	NORTH YORK MOORS NATIONAL PARK	**3.569**	1.785	1.784	0.892	0.892
12	LAKE DISTRICT NATIONAL PARK	**4.029**	2.014	2.014	1.007	1.007
13	Galloway	**2.426**	1.213	1.213	0.606	0.607
14	NORTHUMBERLAND NATIONAL PARK	**2.058**	1.029	1.029	0.515	0.514
15	Kyles of Bute	**1.338**	0.669	0.669	0.335	0.335
16	Perth	**1.493**	0.747	0.747	0.374	0.373
17	Glen Affric	**1.498**	0.748	0.749	0.375	0.374
18	Fyvie	**1.736**	0.868	0.868	0.434	0.434
19	Isle of Lewis	**0.569**	0.284	0.284	0.142	0.142
20	The Orkneys	**0.719**	0.360	0.359	0.179	0.180

TABLE 3: MULTI-SOURCE VERSION (EUROPEAN EMISSIONS ONLY)

		Model 1991*	Model (50%)	Scaled (50%)	Model (25%)	Scaled (25%)
01	DARTMOOR NATIONAL PARK	**8.436**	4.218	4.218	2.109	2.109
02	NEW FOREST	**6.695**	3.347	3.348	1.674	1.674
03	Kent Downs	**7.828**	3.914	3.914	1.958	1.957
04	PEMBROKESHIRE COAST NATIONAL PARK	**6.708**	3.354	3.354	1.677	1.677
05	BRECON BEACONS NATIONAL PARK	**7.194**	3.596	3.597	1.799	1.798
06	Northampton	**6.452**	3.226	3.226	1.613	1.613
07	NORFOLK BROADS NATIONAL PARK	**5.456**	2.728	2.728	1.364	1.364
08	SNOWDONIA NATIONAL PARK	**11.373**	5.686	5.686	2.843	2.843
09	PEAK DISTRICT NATIONAL PARK	**7.060**	3.530	3.530	1.765	1.765
10	YORKSHIRE DALES NATIONAL PARK	**8.908**	4.454	4.454	2.227	2.227
11	NORTH YORK MOORS NATIONAL PARK	**6.437**	3.218	3.219	1.609	1.609
12	LAKE DISTRICT NATIONAL PARK	**13.091**	6.545	6.545	3.273	3.273
13	Galloway	**9.244**	4.621	4.622	2.311	2.311
14	NORTHUMBERLAND NATIONAL PARK	**5.588**	2.794	2.794	1.397	1.397
15	Kyles of Bute	**6.342**	3.171	3.171	1.586	1.585
16	Perth	**3.529**	1.765	1.764	0.882	0.882
17	Glen Affric	**8.946**	4.473	4.473	2.236	2.236
18	Fyvie	**4.955**	2.477	2.478	1.239	1.239
19	Isle of Lewis	4.948	2.473	2.474	1.237	1.237
20	The Orkneys	**3.815**	1.908	1.907	0.953	0.954


a) Drax Deposition Foot Print
> 2.25
1.50 – 2.25
0.75 – 1.50
0.00 – 0.75
kg S ha -1 year -1
b) Sites Chosen for Linearity Analysis
20
19
18
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10
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2
1

FIGURE 1.

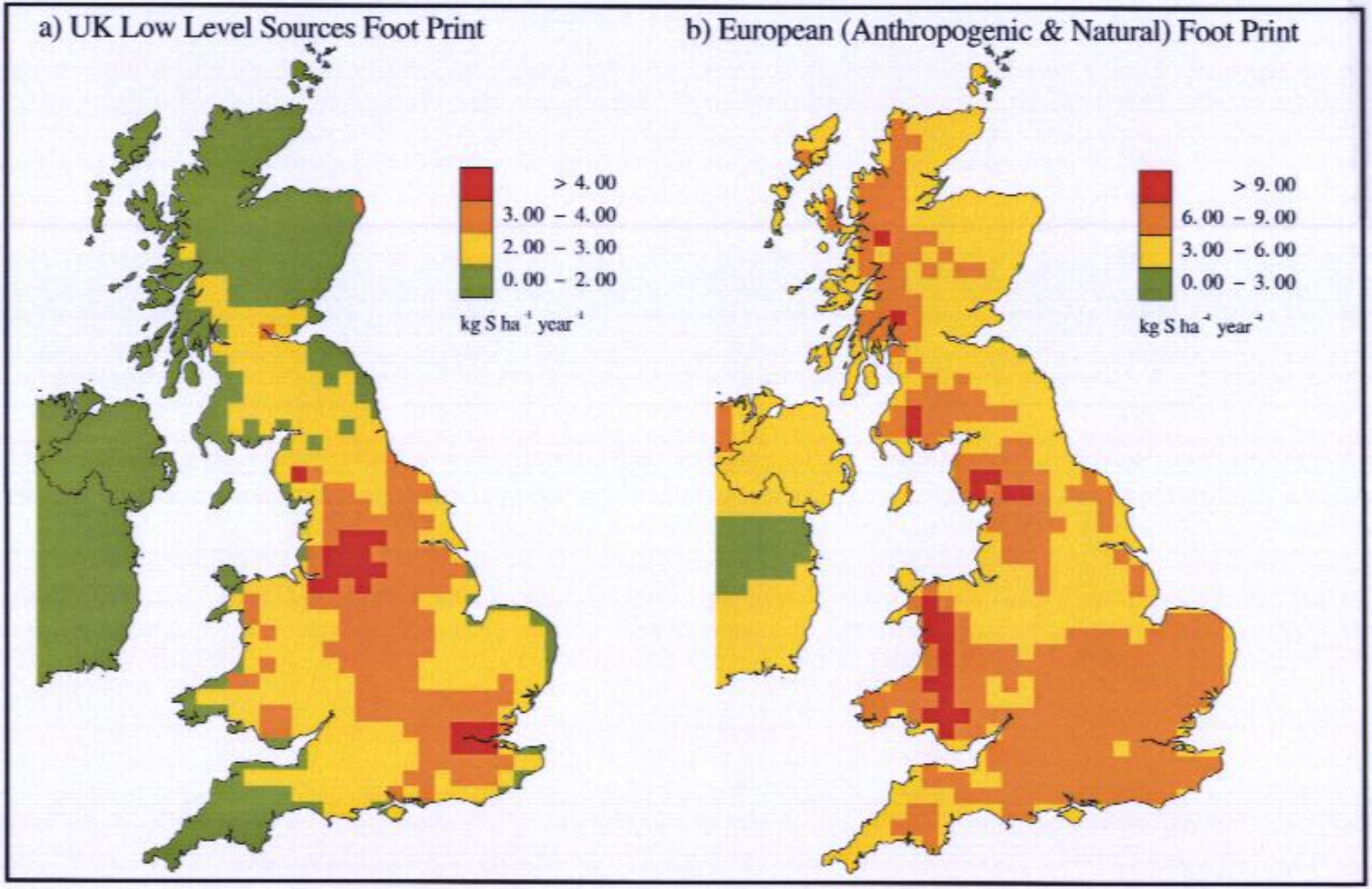
a) UK Low Level Sources Foot Print
> 4.00
3.00 – 4.00
2.00 – 3.00
0.00 – 2.00
kg S ha -1 year -1
b) European (Anthropogenic & Natural) Foot Print
> 9.00
6.00 – 9.00
3.00 – 6.00
0.00 – 3.00
kg S ha -1 year -1

FIGURE 2.