

A CHRONOLOGY OF NITROGEN DEPOSITION IN THE UK BETWEEN 1900 AND 2000

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Abstract. Measurements of the concentrations of nitrogen compounds in air and precipitation in the UK have been made since the mid-19th century, but no networks operating to common protocols and having traceable analytical procedures were established until the 1950s. From 1986 onwards, a high-quality network of sampling stations for precipitation chemistry was established across the UK. In the following decade, monitoring networks provided measurement of NO₂, NH₃, HNO₃ and a satisfactory understanding of the dry deposition process for these gases allowed dry deposition to be quantified. Maps of N deposition for oxidized and reduced compounds at a spatial scale of 5 km × 5 km are available from 1986 to 2000. Between 1950 and 1985, the more limited measurements, beginning with those of the European Air Chemistry Network (EACN) provide a reasonable basis to estimate wet deposition of NO₃⁻-N and NH₄⁺-N. For the first half of the century, estimates of deposition were scaled with emissions assuming a constant relationship between emission and deposition for each of the components of the wet and dry deposition budget at the country scale. Emissions of oxidized N, which dominated total nitrogen emissions throughout the century, increased from 312 kt N annually in 1900 to a peak of 787 kt for the decade 1980–1990 and then declined to 460 kt in 2000. Emissions of reduced N, largely from coal combustion were about 168 kt N in 1900, increasing to a peak of 263 kt N in 2000 and by now dominated by agricultural sources. Reduced N dominated the deposition budget at the country scale, increasing from 163 kt N in 1900 to 211 kt N in 2000, while deposition of oxidized N was 66 kt N in 1900 and 191 kt N in 2000. Over the century, 68 Mt (Tg) of fixed N was emitted within the UK, 78% as NO_x, while 29 Mt of nitrogen was deposited (43% of emissions), equivalent to 1.2 t N ha⁻¹, on average, with 60% in the reduced form. Deposition to semi-natural ecosystems is approximately 15 Tg N, equivalent to between 1 and 5 t N ha⁻¹, over the century and appears to be accumulating in soil. The N deposition over the century is similar in magnitude to the total soil N inventory in surface horizons.

Keywords: deposition, emissions, historical, nitrogen

1. Background

Nitrogen emissions to the global atmosphere from combustion and agricultural activities now exceeds the N emitted by natural processes by almost a factor of 2 (Galloway, 1998). Atmospheric N compounds are important reactants for the production of tropospheric ozone and other photochemical oxidants, and deposited N is changing the species composition of semi-natural plant communities at regional



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scales in Europe. Effects on both atmospheric chemistry and ecosystem function are now widespread and are regarded as indicators of the global perturbation of the N cycle (Cowling *et al.*, 2002). Human health effects resulting from the use of increased use of nitrogen in agriculture, and from combustion of fossil fuels are also widespread geographically and in the range of different effects (Townsend *et al.*, 2003).

Atmospheric emissions of N grew rapidly during the agricultural and industrial revolutions of the 19th century, but early records are poorly documented (Sutton *et al.*, 1993). Measurements of N compounds in air and precipitation are also very limited and of variable quality between 1800 and 1950. There were no networks of air and precipitation chemistry samplers until 1950 (Eriksson, 1952).

Given the importance of N compounds to atmospheric composition and ecosystem function (see papers in this issue), it is important to quantify the magnitude of N inputs to terrestrial surfaces, its change with time and the fate of the deposited N.

In this paper, we develop a chronology of N deposition to terrestrial surfaces in the UK between 1900 and 2000, and where possible, indicate the probable emission and deposition further back towards the start of the industrial revolution. In the limited space of this special volume, we have concentrated on the essential details of the approach. A comparison between this chronology and measurements available from a variety of locations throughout the UK is considered in a more detailed analysis (O'Donoghue, manuscript in preparation).

2. Emission of Oxidized and Reduced Nitrogen in the UK

Current inventories provide spatially disaggregated emissions of NO_2 and NH_3 for the UK at a resolution of $5 \text{ km} \times 5 \text{ km}$ (Goodwin *et al.*, 2001). However, this spatial resolution is limited to the period 1995–2000 and even the national totals are limited to the period 1970–2000 in the current inventory. Earlier, less-sophisticated inventories have been developed, and these provide estimates of annual NO_x emissions for the UK for the period 1900–1980 (RGAR, 1983). Emissions of reduced N as NH_3 are much less certain and are available for a shorter period in the national UK inventory (Dragosits *et al.*, 1998). The sources of information on NO_x and NH_3 emissions in the UK used in the study are summarised in Table I.

2.1. REDUCED N EMISSIONS

The main source of atmospheric NH_3 is livestock farming, and as reliable statistics on livestock numbers in the country are available (Misselbrook *et al.*, 2000), these data may be used to estimate emissions using emission factors for the different livestock and management systems. Using this simple approach (Asman *et al.*, 1988), animal numbers for 1870–1950 were multiplied by emission factors described by (Misselbrook *et al.*, 2000). This method however does not account for changes in

TABLE I
A summary of nitrogen emission data for the UK from 1830 to 2000

Date	Information	Source	Comment
1830–2000	Coal consumption statistics (NH _x)	Ashworth and Pegg (1986), Church <i>et al.</i> (1986), Court (1951), NationalStatistics (2003), PEP (1936)	Combustion emission factor (Geadah, 1985; Bauer and Andren, 1985)
1830–2000	Agricultural statistics (animal numbers, fertiliser use) (NH _x)	GSS (1973), GSS (1996)	Emission factor approach
1830–1870	Population statistics	NationalStatistics (2001)	Used for scaling animal numbers pre-1870
1900–1980	Expert estimate on NO _x emission trend	RGAR (1983)	
1980–2000	NO _x emission	Goodwin <i>et al.</i> (2002)	1980 emissions used as baseline

the animals' diet and livestock husbandry. In a second approach, an estimate for total N excretion by livestock (Webb, 2001) and fertiliser use is used to partition the total 1995 NH₃ emissions into emissions from "basic manure" and emissions from "fertiliser-induced" manure. The fraction for "basic manure" ammonia emissions is 46% of the "emission factor approach" NH₃ emissions.

Reduced N emissions from NH₃ from domestic coal combustion have been neglected in the UK emissions inventories to date, and appear to be a substantial source between 1800 and 1950. A large proportion of the recorded national coal consumption was used for residential/domestic purposes before 1950. Thus, coal consumption data from 1830 to 1990 were divided into combustion at high temperatures (industry) and combustion at low temperatures (domestic) and total NH₃ emissions calculated, respectively.

Industrial high-temperature combustion results in trivial NH₃ emissions (Bauer and Andren, 1985) with an average emission factor of 0.23 g NH₃-N t⁻¹ coal consumed. These reduced N emissions were negligible for the period 1830–1990.

However, the domestic coal combustion emission factor, estimated to be 0.82 kg NH₃-N t⁻¹ coal burnt (Geadah, 1985) is a substantial source, but associated with considerable uncertainty. Discussions on NH₃ coal combustion emission factors can be found in Sutton *et al.* (2000), Lee and Dollard (1994), Battye *et al.* (1994) and Warn *et al.* (1990).

For the period 1830–2000, this approach provides an estimate of UK NH₃ emissions from coal combustion (Figure 1), showing that throughout the period 1850–1950 coal was the main source of NH₃ emission in the UK. An examination of rain chemistry data reported by Smith (1872) and Crowther and Ruston (1913) for the urban areas provides evidence of substantial sources of reduced nitrogen in

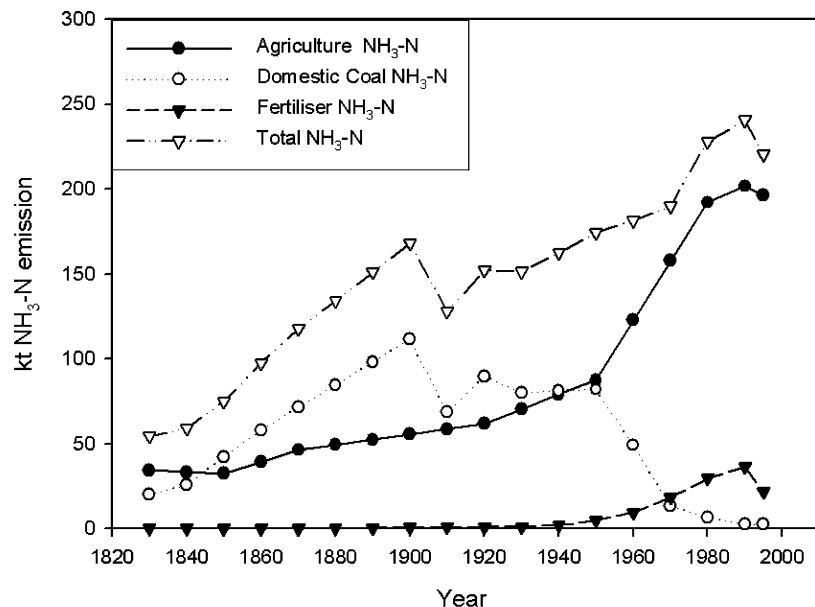


Figure 1. Reduced nitrogen emissions in the UK between 1830 and 2000.

urban areas. The data show NH_4^+ concentrations in rain of $767 \mu\text{eq l}^{-1}$, exceeding those in rural areas by more than an order of magnitude, and in general support of large urban emissions. The rural concentrations in rain range from 10 to $40 \mu\text{eq l}^{-1}$ (Smith, 1872), which is in the same range as present day values. These urban concentrations are consistent with a very large urban source, which includes domestic coal combustion, a substantial source from the urban horse population, and poor sanitation facilities.

Emissions of reduced N rose steadily from 1820, reflecting use of domestic coal, which exceeded agricultural emissions by 1845. Domestic coal usage peaked in 1900, and associated reduced N emissions gradually declined. Since 1940 agricultural NH_3 emissions have dominated the UK sources of NH_3 which peak in the early 1980s and have declined slightly since then.

2.2. OXIDIZED N EMISSIONS

Inventories of NO_x emissions are currently among the best known of the pollutant emissions, supported by extensive measurement programmes and a detailed separation into different sectors (Goodwin *et al.*, 2001). Most of the NO_x is emitted by vehicles and large combustion plant, but there is a natural component due to emissions of NO from soil as a consequence of nitrification. Emissions in this paper were based on Goodwin *et al.* (2001) (National Atmospheric Emissions Inventory, NAEI) from 1970 to 2000, and prior to 1970, values were taken from RGAR (1983),

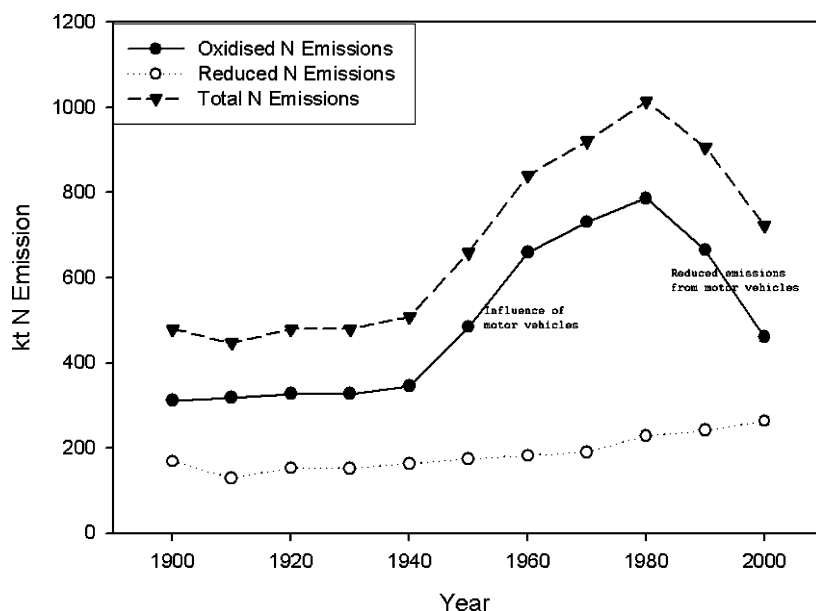


Figure 2. Oxidised, reduced and total nitrogen emissions in the UK between 1900 and 2000.

adjusted on the basis of the NAEI values for the common year 1980. This process yields an estimate of the UK emissions of oxidized and reduced nitrogen for the 20th century during which the total emission is dominated by oxidized N (Figure 2). The different trends reflect the very different source activities and the social and industrial development associated with them.

3. Precipitation Chemistry Measurements

Measurements of the nitrogen content of precipitation in the UK began in the mid-19th century with the work of Lawes and Gilbert (1854) and Smith (1872) following the discovery by Rutherford in the 18th century of the presence of nitrogen in air. The measurements made then by weight and in isolation are difficult to compare with present day protocols, but provide an indication of amounts deposited (Erisman, 2000). The first European network of precipitation chemistry, operating to common protocols and reporting the N in air and precipitation was that established by (Eriksson, 1952) and known as the European Air Chemistry Network (EACN). The EACN originated in Sweden in 1947 as part of an agricultural research programme on nutrient cycles and by 1955 sampling sites had been established throughout Europe. The network in the British Isles comprised 17 sites including seven in Ireland and 10 sites in the UK. In 1986, two acid deposition monitoring networks became operational in the UK (Devenish, 1986). The primary

TABLE II
Precipitation chemistry measurements in the UK from 1850 to 2000 and their source

Date	Information	Source	Comment
	Very early measurements of rain chemistry	Lawes and Gilbert (1854)	N compounds are wet deposited
1869–1870	Early measurements of rain chemistry	Smith (1872)	Show urban air to be highly polluted by NH_4^+
1900–1950	Occasional measurements, e.g. Rothamsted (rural), Leeds (urban)	Russel and Richards (1919) (see Goulding <i>et al.</i> , 1998) Crowther and Ruston (1913)	Some valuable N deposition estimates
1950–1964	Rain chemistry: EACN	Eriksson (1952), Stevenson (1986)	The first UK networks, up to 12 sites, insufficient for UK deposition mapping
1978–1986	UK air chemistry measurements	RGAR (1983, 1987)	
1986–2000	UK precipitation chemistry measurements and NO_2 concentration	NEGTA (2001)	Suitable for estimating wet N deposition and NO_2 dry deposition
1996–2000	NH_3 network	Sutton <i>et al.</i> (2001)	Provides basis for UK deposition estimated using FRAME
1999–2000	HNO_3 network	NEGTA (2001)	Provides basis for dry deposition

network sampled with wet-only collectors daily at nine rural sites and the secondary network used bulk precipitation collectors with a sampling frequency of weekly or fortnightly. Precipitation chemistry from 59 sites comprising this secondary network was used to produce annual wet deposition maps. From 1989 onwards, the number of the secondary network sites was reduced to 32 (+ a reduction to five sites for the primary network). Measurements of UK N deposition are summarized in Table II, along with references to publications and reports in which details of the methods and sampling protocols may be found.

4. Wet Deposition Maps

Maps for the UK are derived from the product of the mean volume weighted concentrations of NH_4^+ and NO_3^- and annual precipitation. Concentration maps are obtained using linear Kriging interpolation. The wet deposition maps are then

created at a 5 km grid resolution as the product of the ion concentration grid with the 5 km rainfall grid (1961–1990), and corrected for the effects of seeder feeder enhancement of wet deposition using the method described by Dore *et al.* (1992).

For the period 1950–1960, the EACN data based on at most 12 stations for the UK, provide a reasonable estimate of the spatial patterns of concentration for the major ions, which are similar to the values in recent maps based on 35 sampling stations. However, the limited site network leads to substantial interpolation errors, with areas of high concentration extrapolated into regions, shown in later networks to be relatively unpolluted. The errors introduced in wet deposition estimates using these early data were too large to use them for development of the chronology.

5. Dry Deposition

Dry deposition of NO_2 and HNO_3 is currently calculated from the measured concentration fields using a process-based deposition model (Smith *et al.*, 2000). The measured concentration measurements are limited to the period 1999 onwards, and no measurements of HNO_3 are available to estimate the importance of this very reactive gas in the UK.

Adding wet and dry deposition gives the total deposition seen in Figure 3 which shows the annual N deposition fields for oxidized and reduced nitrogen for the UK at a resolution of 5 km \times 5 km.

6. Extending the Deposition Chronology back from 1986–2000 to 1900

In the absence of measured deposition fields for the UK for the period prior to 1986, the trends with time can be approximated using the emissions data, assuming the relationships between emission and deposition remain reasonably constant with time.

Reconstruction for oxidized N deposition was based on the data for 1980s, the earliest decade in which high-quality monitoring networks provided the basis for accurate estimates of both wet and dry deposition. For reduced N emissions, the base year 1990 was chosen, as NH_4^+ -N budget estimates are appreciably more reliable in 1990 than for 1980.

Having established reduced and oxidized N emissions, deposition was scaled linearly according to the proportions in the UK deposition budget estimates for the base year 1980 for oxidized N and 1990 for reduced N. The emissions estimate for oxidized N is taken from Goodwin *et al.* (2002) and for reduced N the scaled emissions estimate (see above) is used. Deposition is based on the decadal average of wet deposition (measurement-based) and scaled dry deposition. Thus, a budget

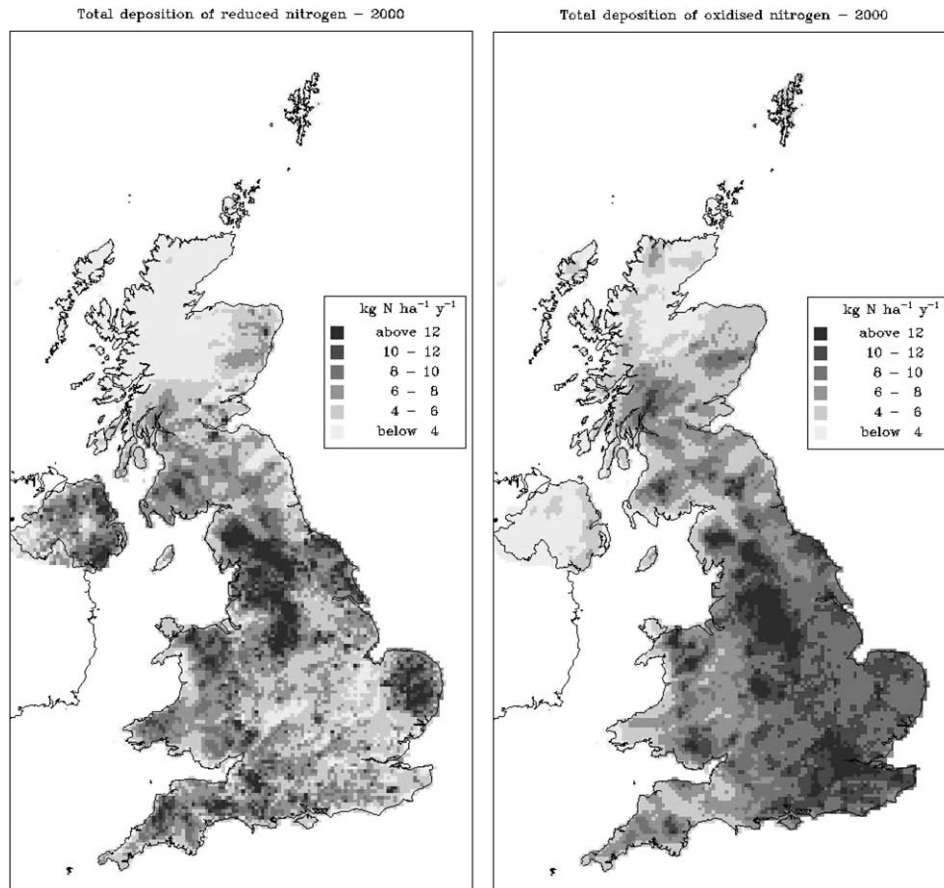


Figure 3. Deposition of reduced nitrogen (wet deposited NH_4^+ and dry deposited NH_3) and oxidized nitrogen (wet deposited NO_3^- and dry deposited NO_2 and HNO_3) for 2000.

for the UK for the period 1900–2000 was established and N exports from the UK were calculated as the difference between emission and deposition, as shown in Table III.

Using the maps for the period 1950–2000, a cumulative N deposition map may be estimated, assuming a constant pattern in deposition throughout the chronology. Such an assumption is clearly subject to considerable uncertainty since the early emissions are so dominated by the urban emissions of reduced N. Thus, the maps developed are likely to have significantly underestimated the deposition in the urban areas and the footprint of the major conurbations. Likewise, deposition in the remote regions has probably been overestimated in the early years, but on average, the deposition in the first half of the century was half of that in the later years.

TABLE III
Emission, deposition and export of oxidized nitrogen (NO_3^- wet deposition, NO_2 and HNO_3 dry deposition) and reduced nitrogen (wet deposited NH_4^+ and dry deposited NH_3) in the UK between 1900 and 2000 as decade averages

	Emission			Deposition			Export		
	Oxidized N emission	Reduced N emission	Total emission	Oxidized N deposition	Reduced N deposition	Total deposition	Oxidized N export	Reduced N export	Total export
1900	312 ^a	168 ^c	480	66 ^e	163 ^g	229	246	5	250
1910	318 ^a	128 ^c	446	67 ^e	125 ^g	192	251	4	255
1920	327 ^a	152 ^c	480	69 ^e	148 ^g	217	258	4	262
1930	327 ^a	151 ^c	479	69 ^e	147 ^g	216	258	4	262
1940	345 ^a	162 ^c	508	73 ^e	158 ^g	231	272	4	277
1950	484 ^a	174 ^c	658	102 ^e	169 ^g	271	382	5	387
1960	659 ^a	181 ^c	840	139 ^e	176 ^g	315	520	5	525
1970	731 ^a	190 ^c	920	154 ^e	185 ^g	339	577	5	582
1980	787 ^a	228 ^c	1015	194 ^f	222 ^g	416	593	6	599
1990	665 ^b	241 ^c	906	196 ^f	234 ^h	430	470	7	476
2000	460 ^b	263 ^d	723	191 ^f	211 ⁱ	402	270	52	321
Total ^j	50014	18026	68040	11475	17484	28959	38539	542	39081

All values in kt N.

^aTrend from RGAR (1983), scaled according to 1980's emissions (NAEI report Nov. 2001).

^bFrom NAEI report (Goodwin *et al.*, 2002).

^cScaled according to agricultural sources (animals), coal consumption (domestic and industrial combustion) and fertiliser use (1950–1990).

^dFrom NAEI report (Goodwin *et al.*, 2002).

^eScaled back according to emissions.

^fBased on measurements (wet deposition measurements/budgets, dry deposition scaled with emissions assuming constant wet-dry partitioning).

^gScaled according to emissions.

^hWet deposition based on measurements, dry deposition scaled mean value of 1990s.

ⁱWet Deposition based on measurements, dry deposition scaled mean value 2000–2001.

^jTotal is the cumulative value for the century (100 years), adding the sums of decadal values (e.g., (1900-value * 10) + (1910-value * 10) + ... + 2000-value).

7. The Fate of N Deposited in the UK 1900–2000

The deposited N has three possible fates, being either emitted back to the atmosphere as N_2 , NO, N_2O or NH_3 , or leached from the soil profile as NO_3^- or accumulation in the soil and vegetation mainly as organic nitrogen. A detailed analysis of the likely fate is beyond the scope of this paper. However, some broad conclusions concerning the fate are clear. Nitrogen deposition to managed grassland and arable land is small by comparison with N fertilizer inputs to these land uses and cropping and soil cultivation prevent the long-term accumulation of nitrogen in arable soils (Jenkinson, 1982). However, levels of atmospheric N deposition to semi-natural land are, for large areas, considerably larger than the rates of gaseous loss and few of these areas leach significant quantities of soil N. Thus, in these soils nitrogen

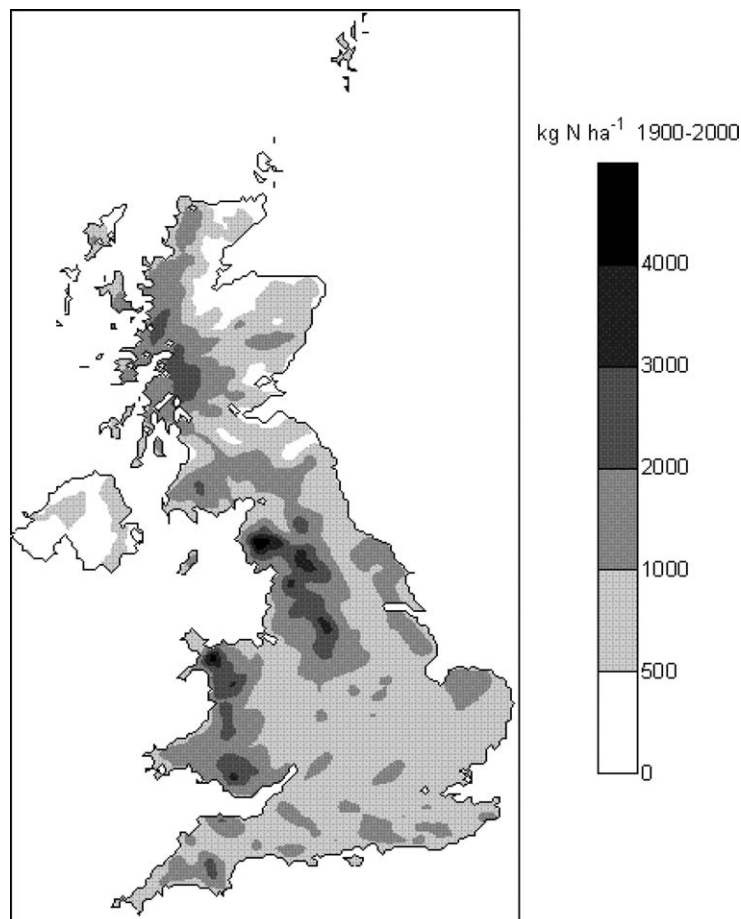


Figure 4. Total cumulative N deposition in the UK between 1900 and 2000.

accumulation is the most important sink and the supply of additional nitrogen may be very important in modifying the carbon economy as well as in changing the species composition of the flora (Pitcairn *et al.*, 1998).

The semi-natural land cover in the UK makes up 46.3% of the land area and the deposition of fixed N to this land between 1900 and 2000 amounted to approximately 15 Mt N, of the total 29 Mt N deposited (Figure 4).

8. Discussion

The exercise presented here combines high-quality measurement and modelling for the latter two decades of the 20th century with progressively less certain values for earlier decades to provide a chronology of nitrogen deposition. Some of the assumptions made to extrapolate back from the 1950 are simple and may be substantially in error. Measurements from the early decades of the 20th century

TABLE IV
NH₄-N concentrations in precipitation at background and rural sites in 1870–1920 and 2000

	1870	2000
Background		
Dalwhinnie	12	3
Aberfeldy	44	8
Crieff	6	9
Achindrain	4	13
Rhu (Row)	21	10
Wemyss Bay	6	10
Mean ^a	16	9
Rural		
Wilcott	52	22
Ludlow	22	19
Rugby	65	25
Tring	21	22
Steatley	16	22
Aldershot	33	18
Mean ^a	35	21
Rothamsted ^b	23	23

All values in $\mu\text{eq NH}_4\text{-N l}^{-1}$.

^aSmith (1872).

^b1888–1916 mean (Russel and Richards, 1919).

support the approach, with concentrations of NH_4^+ during the period 1870–1920 quite similar to those for period covered by the networks (2000), as shown in Table IV. However, the measurements are too few and uncertain in themselves to lend much weight. It is relatively easy to show that the budget derived is unlikely to be in error by more than a factor of two, thus the broad conclusions concerning the relative importance of oxidized and reduced nitrogen and the changes with time are secure.

The quantity of N deposited between 1900 and 2000 is a substantial fraction of the soil N in semi-natural soils, which ranges from 0.5 to 4 t ha⁻¹ as shown in Figure 5 (Jenkinson, 1982). Furthermore, the spatial variability in the deposition, especially close to large sources of NH_3 , creates local hot spots where very much larger accumulations of soil N are likely. Thus, from Figure 4 it is clear that there are substantial areas of the UK where the inventory of soil N in the top 30 cm should have doubled during the century, but with the largest change throughout the latter half of the century, driven by the increasing contribution of reduced N, largely from agriculture.

The uncertainties in this exercise are large, and in the case of the fixed partitioning of the deposition relative to emissions, this probably leads to systematic error. However, the direction of the error is fairly clear, as the very large emissions of reduced nitrogen in urban areas in the early years of the 20th century occurred in a very acidic atmosphere with substantial SO_2 sources, in which the ammonia would deposit very rapidly to terrestrial surfaces, and gas to particle conversion of the remaining NH_3 would be rapid. It is therefore likely that the fraction of N emissions deposited in rural areas in the early 20th century was larger than at the end of the century. Thus, it is likely that the values produced here are underestimates

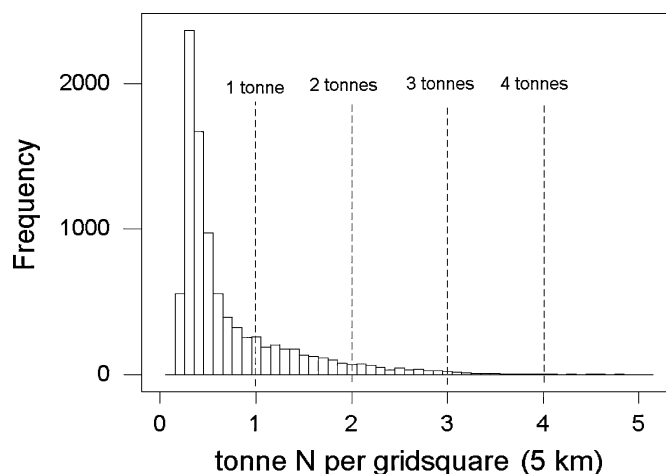


Figure 5. Histogram of N deposited on semi-natural land between 1900 and 2000.

of the actual values. If the emission data deduced in this work can be reliably disaggregated spatially, the application of a chemical, transport model to the early decades of the 20th century would provide a very useful test of the range of possible N budgets and spatial distribution of the deposited nitrogen. The long-term fate and consequences of atmospheric N emissions and deposition remain uncertain and emissions are proving intractable to control. Thus, a more detailed examination of the long-term historical nitrogen deposition in the UK seems likely to provide a valuable guide to the areas of further study, and to the locations where effects of the accumulated soil nitrogen are most likely to be found.

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