

RECENT TRENDS IN NO₃-N CONCENTRATION AND LOADS IN BRITISH RIVERS

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ABSTRACT Data from routine water quality surveillance of many unpolluted headwater reaches during the period 1974-1986 are analyzed to investigate the pattern and controls of recent changes in NO₃-N concentration and flux (load) in British rivers. Mapping of mean annual values reveals contrasting countrywide patterns for NO₃-N concentration and load. British rivers are typified by a pronounced annual cycle of NO₃-N concentrations, but statistically significant rating relations between NO₃-N concentration and flow are less well developed. Analysis of trends in the period 1980-1986 revealed a significant increase, a significant decrease and no significant change in NO₃-N concentrations at 51, 11, and 38% respectively of the study sites. Comparable percentages for NO₃-N loads were 34, 17, and 49%. Rates of increase in mean concentration were highest in the intensively farmed lowland areas and in winter. Analysis of trends in the period 1974-1986 highlighted the complicating influences of the 1975-1976 drought, and extrapolation of recent trends suggests that unacceptably high NO₃-N concentrations (>11.3 mg l⁻¹) will be a problem in a large part of Eastern England by the Year 2000.

INTRODUCTION

Recent years have witnessed growing concern over the trend of increasing nitrate concentrations which has been documented for the last 30 to 40 years in a number of river systems of Europe and elsewhere (e.g. Paces, 1982; Hagebro *et al.* 1983; White, 1983; STACWQ, 1984; Smith & Alexander, 1985; D.O.E., 1986a; Roberts & Marsh, 1987). Fears have been expressed concerning eutrophic degradation of water courses enriched in nitrogen (Meybeck *et al.* 1989), and the health risks associated with nitrates in drinking water (e.g. Forman *et al.* 1985; ECETOC, 1988) have led to the setting of stringent standards for concentrations of this nutrient in potable supplies (e.g. WHO, 1970).

Investigations of long-term trends in river NO₃-N concentrations, however, have been hampered by the relatively few suitable data sets which are available for analysis and by the problems which attend the recognition and interpretation of trends in water quality conditions. In the United Kingdom, for example, extended time series exist for some of the larger rivers (Casey & Clarke, 1979; Wilkinson & Greene, 1982) but rarely are available for small catchments (Burt *et al.* 1988). Studies of trends in river NO₃-N concentrations also have almost exclusively focussed on changes in concentration rather than on fluctuations in the flux of nitrogen through the fluvial system. Assessment of changes in NO₃-N concentrations over time is complicated by strong seasonal and inter-annual fluctuations and by periods of rapid change interspersed with periods of stability, which are often evident in available long-term records (Roberts & Marsh, 1987; Toms, 1987). Interpretation

of the causes of $\text{NO}_3\text{-N}$ trends in rivers also is uncertain because of the complexity of inputs, transformations, and outputs involved in the nitrogen cycle (Smith & Alexander, 1985), and marked increases in $\text{NO}_3\text{-N}$ concentrations generally have been ascribed in Europe to increasing fertilizer usage (Roberts & Marsh, 1987) and in parts of North America to increasing atmospheric deposition of nitrogen (Smith *et al.* 1987).

Data gathered from a number of rivers in the UK and elsewhere suggest that the nature of trends in $\text{NO}_3\text{-N}$ concentrations varies considerably among catchments. With the exception of recent studies based on the United States Geological Survey's National Stream Quality Accounting Network and the United States Environmental Protection Agency's National Stream Quality Surveillance System (Smith *et al.* 1987), there have been few investigations of countrywide variations in trends of $\text{NO}_3\text{-N}$ concentrations. Data available from routine water quality surveillance by statutory authorities at many sites on rivers of mainland Britain provide an opportunity to investigate the pattern and controls of recent changes in both the concentration and the flux (load) of $\text{NO}_3\text{-N}$ across the UK. Such data are inevitably limited because they do not provide a long time series for investigation. However, data from routine water quality sampling do have the advantage of providing a geographically extensive perspective and, therefore, permit spatial variation in recent trends in $\text{NO}_3\text{-N}$ concentrations to be examined.

THE DATA BASE

The present study is based on the conflation of data from the routine surveillance and monitoring of water quality by the Regional Water Authorities in England and Wales and the River Purification Boards in Scotland during the period 1974-1986. Since privatization of the water industry in 1989, the statutory responsibility for sampling water quality lies with the National Rivers Authority and its regional sections. Attention was focussed on $\text{NO}_3\text{-N}$ concentrations characteristic of waters sampled near source rather than at downstream locations on major rivers. To this end, data were collected, as far as possible, for headwater sites located on reaches classified as being unpolluted in national surveys (D.O.E., 1986b) and fed by relatively small and homogeneous catchment areas ($<100 \text{ km}^2$).

Sampling frequencies in the study period varied considerably among the different Water Authorities and River Purification Boards involved and typically sampling was not conducted on a regular schedule throughout the year. Most stations that were selected for the present analysis were sampled at least eight times annually, which yielded >100 observations in the period 1974-1986. In two of the Water Authority areas (Anglian and North-west), however, water quality records were not available for the period before 1980.

Information was collected to examine trends in $\text{NO}_3\text{-N}$ concentrations at 743 sites across mainland Britain, but a more restricted data-base of 430 stations was available to investigate changes in $\text{NO}_3\text{-N}$ loads, reflecting the fact that discharge measurements were undertaken at or near about half of the water quality sampling stations considered in the present study. Annual $\text{NO}_3\text{-N}$ loads were calculated by an interpolation method proposed for computing dissolved and other fluxes from information collected on river concentrations in the UK Harmonized Monitoring Network (Rodda & Jones, 1983). In this approach, the product of concentration and discharge (either the instantaneous or daily mean value) on the day of sampling is considered to be representative of conditions occurring between sampling occasions.

The data-base established for the present study is, to a certain extent, limited by several

factors. These relate especially to the adoption of different laboratory analysis techniques by the various statutory authorities involved to differences among these authorities in: (1) spatial density and distribution of sampling stations and (2) the frequency and timing of sampling, and to the overall design of monitoring networks, which were established primarily to identify the incidence of pollution rather than to assess background water quality. Nevertheless, the results of routine surveillance and monitoring by the statutory authorities in the UK provide a valuable, and hitherto largely unexploited, source of information on $\text{NO}_3\text{-N}$ behavior at the countrywide scale.

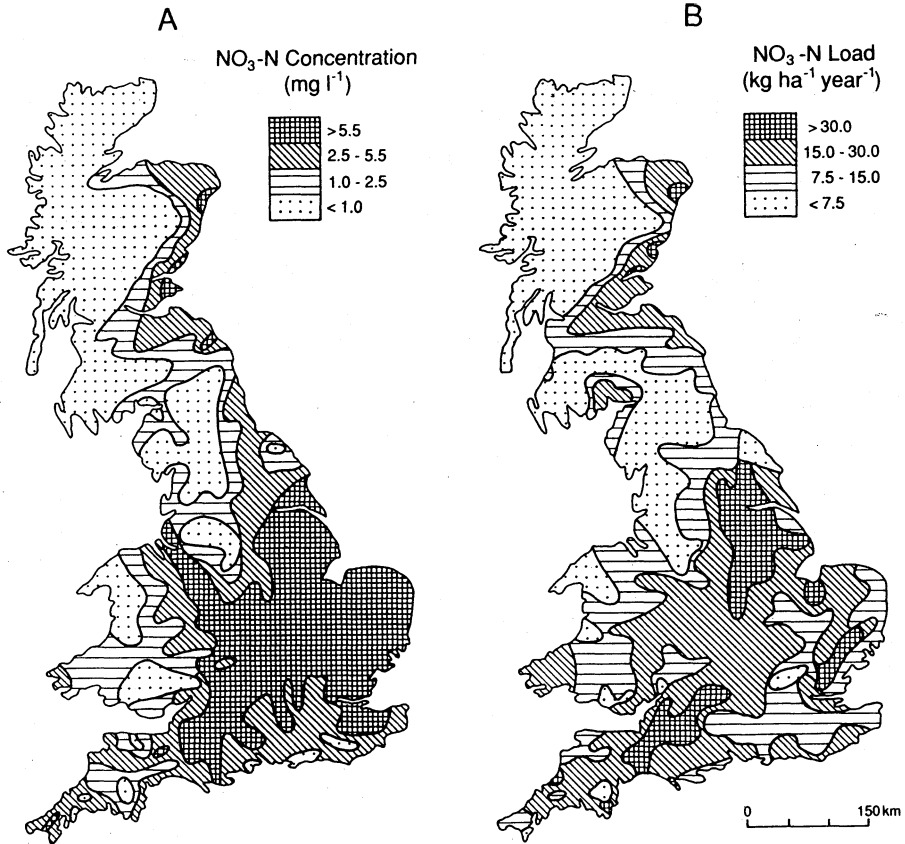


FIG. 1 Mean annual $\text{NO}_3\text{-N}$ concentrations (A) and loads (B) in British rivers.

$\text{NO}_3\text{-N}$ BEHAVIOR IN BRITISH RIVERS

A brief consideration of different facets of $\text{NO}_3\text{-N}$ behavior in British rivers provides a background context for the discussion of trends in $\text{NO}_3\text{-N}$ concentrations over recent years.

Spatial patterns

Mean annual $\text{NO}_3\text{-N}$ concentrations and loads have been mapped for mainland Britain in Fig. 1. Mean concentrations ranged across the country from 0.1 to 15 mg l^{-1} , and a marked northwest to southeast gradient is evident (Fig. 1A). The gradient strongly reflects country-wide differences in relief, hydrometeorological conditions and agricultural activity. Upland areas in the north and west generally are associated with mean concentrations less than 2.5 mg l^{-1} . The higher massifs in Southwest England, Wales, Northern England and Scotland, where high annual precipitation dilutes river solutes and land use is least disturbed by agricultural activity, are characterized by mean $\text{NO}_3\text{-N}$ concentrations less than 1 mg l^{-1} . A decreasing ratio of runoff to precipitation and an increasing intensity of agricultural land use towards the south and east of Britain results in higher mean river concentrations of $\text{NO}_3\text{-N}$. Many of the lowland areas are typified by mean concentrations ranging from 5 to 9 mg l^{-1} , although highest values of $>10 \text{ mg l}^{-1}$ are encountered in East Anglia.

The map of mean annual $\text{NO}_3\text{-N}$ loads (Fig. 1B) exhibits considerable differences in comparison with that of mean concentrations because the pattern of flux reflects the relation between river flow and solute concentration. Low mean $\text{NO}_3\text{-N}$ values ($<7.5 \text{ kg ha}^{-1}\text{year}^{-1}$) are encountered in the upland areas of the north and west where very low $\text{NO}_3\text{-N}$ concentrations offset high runoff. However, mean loads also are relatively low ($<15 \text{ kg ha}^{-1}\text{year}^{-1}$) in the extreme east and south of the country where high $\text{NO}_3\text{-N}$ concentrations are counterbalanced by low runoff volumes. Highest mean $\text{NO}_3\text{-N}$ loads ($>30 \text{ kg ha}^{-1}\text{year}^{-1}$) generally are found in a band running from the Southwest, through the Midlands, to the east coast, where both concentrations and flows attain intermediate values between the extremes of northwest and southeast.

Countrywide patterns of $\text{NO}_3\text{-N}$ concentration and load can be accounted for in more detail by the characteristics of the individual catchments used in the present study. Stepwise multiple regression analysis, which employed information on farm type, land capability, soil type, hydrogeology, relief, rainfall, and runoff in each drainage as independent variables, produced models which explained 81 and 83% of the variance in the data-sets for $\text{NO}_3\text{-N}$ concentrations and loads, respectively (Betton, 1990).

Seasonal régime

A pronounced annual variation in $\text{NO}_3\text{-N}$ concentrations is typical of many British rivers, and this variation can be characterized by the fitting of simple harmonic curves to data collected over several years at a sampling station (e.g. Casey & Clarke, 1979; Walling & Webb, 1985). First-order harmonic curves were significant at $>80\%$ of the sites analyzed here. The variance in concentrations explained by the harmonic model generally decreased at sites in the north and west of the country where $\text{NO}_3\text{-N}$ concentrations remained low throughout the year. Consistently, the majority of sites where a significant sinusoidal trend could not be found were associated with upland catchments marked by very low concentrations. Other sites, where insignificant trends were found, were dominated by groundwater inflows and where concentrations remained relatively high except periodically during storm events.

The amplitude of the fitted first-order harmonic ranged from 0.1 to 7.2 mg l^{-1} , but was generally <0.5 in upland regions and $>2 \text{ mg l}^{-1}$ in the lowland areas of the Midlands and East Anglia (Fig. 2A). Countrywide variation in the magnitude of seasonal fluctuations in $\text{NO}_3\text{-N}$ concentrations may reflect differences in leaching patterns which exist between the

west and east of Britain (Burns & Greenwood, 1982). At a great majority of the sites (>80%) concentrations vary seasonally with the maximum occurring in the winter months from December to March. The timing of this maximum reflects factors such as the leaching of nitrate, which accumulated in the soil in the summer and autumn, by high rates of soil water movement in the winter months, the absence of nitrogen uptake by plants in the dor-

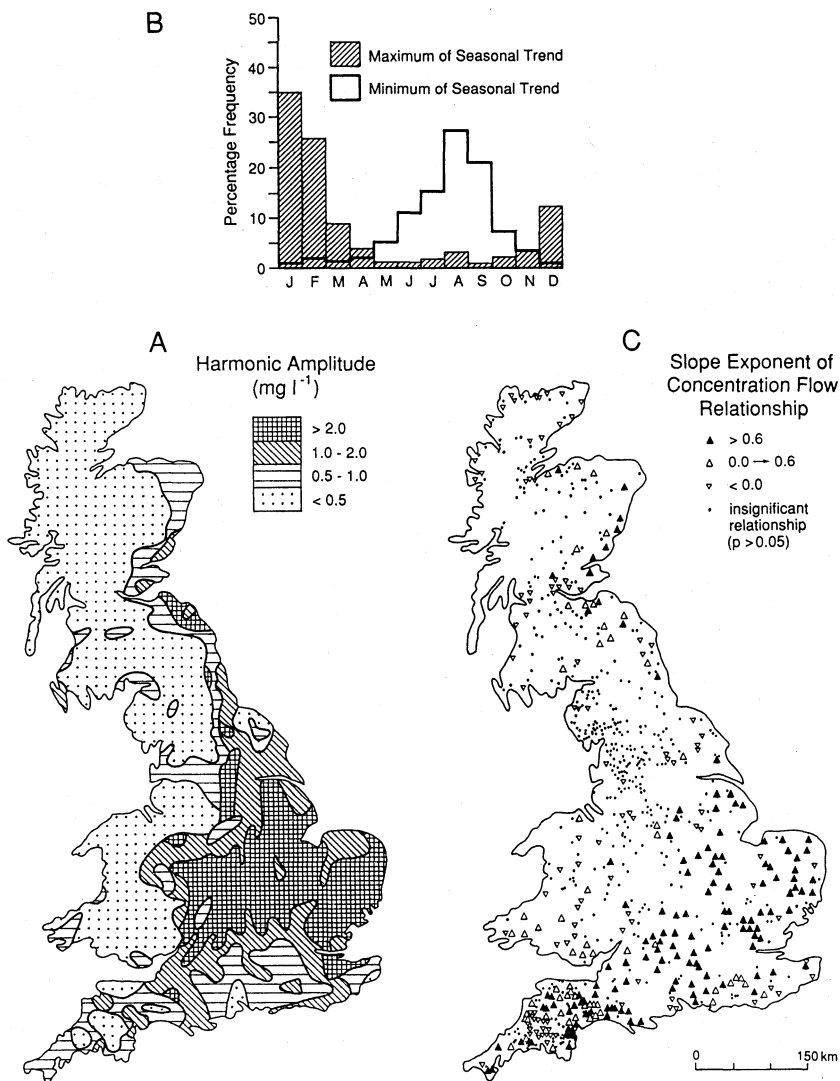


FIG. 2 Amplitude of first-order harmonic function fitted to $\text{NO}_3\text{-N}$ concentration data (A), timing of peak and trough in annual $\text{NO}_3\text{-N}$ concentration cycle (B) and slope of $\text{NO}_3\text{-N}$ concentration-flow rating relationship (C) in British rivers.

mant season, and the continued high rates of mineral nitrogen production throughout the early winter period (Roberts *et al.* 1983). In contrast, diminished soil water movement, increased plant uptake of nitrogen and the occurrence of denitrification during low flow conditions cause $\text{NO}_3\text{-N}$ concentrations to decrease to a minimum in July and August. The annual variation in concentration at most sites is not symmetrical (Fig. 2B), and the data fit a second-order harmonic curve better. A seasonal régime in which the occurrence of peak concentrations in December or March is followed by a slow decline to a minimum in August and then a rapid rise during autumn and early winter was most common, but, at some sites, a faster decrease in spring and slower increase in autumn were encountered.

Fitting of harmonic curves to $\text{NO}_3\text{-N}$ load data also indicates that peak flux occurs in the months of December to March at the great majority of the study sites. However, maximum loads typically occurred in December and January in western areas and in February and March in eastern regions and this reflects a geographical control of the flow régime across Britain.

Concentration-flow relations

The relations between $\text{NO}_3\text{-N}$ concentration and river discharge are often associated with a considerable degree of scatter, which reflects complicated storm-period behavior (Webb & Walling, 1985; Burt & Arkell, 1986), hysteresis in nitrogen availability (Walling & Foster, 1975), and the influence of other factors such as antecedent soil moisture content and temperature (Casey & Clarke, 1979; Thomson, 1979). In the present study, statistically significant power-function rating relations could only be fitted at 61% of the sampling sites (Fig. 2C). For most sites, concentration increased with increasing flow (positive slope). The most statistically significant relations between concentration and flow were encountered mainly in the south and east of the country, and positive slopes were found especially in East Anglia, the Midlands, and parts of Southwest England. In contrast, negative slopes, signifying a dilution effect, typically were found at sites in the Thames Basin, Northeast England, and Southwest Scotland (Fig. 2C).

$\text{NO}_3\text{-N}$ TRENDS

Considerable attention has been paid in recent years to statistical procedures for the detection of trends in water quality time series (e.g. Smith *et al.* 1982; Hock, 1986; Montgomery & Loftis, 1987; Berryman *et al.* 1988). Some of these techniques were developed specifically (1) to analyze monthly water quality data and (2) to remove variations in water quality resulting from changes in season and streamflow, so that concentration changes resulting from human activities could be evaluated (e.g. Hirsch *et al.* 1982; Smith & Alexander, 1985). In the present study, trends due to both natural and anthropogenic causes were of interest, and a simple method of identifying changes in $\text{NO}_3\text{-N}$ concentrations over time was adopted. The method consisted of a linear regression of the 3-year moving average concentration on time. This approach was used in an earlier study to investigate changes in nitrate concentrations of 18 English rivers (Tomlinson, 1970). The analysis provides information on the significance, magnitude and direction of the trend. A trend was regarded as being significant when its probability of occurring by chance was less than 5% ($P < 0.05$).

Trend analysis of $\text{NO}_3\text{-N}$ concentrations and loads for British rivers was undertaken with reference to changes over two periods. The spatially most extensive information was

available for the 7-year period 1980-1986, and although the trend analysis was conducted for a relatively short time period, it provides scope for a countrywide analysis of recent trends similar to that recently undertaken for an 8-year period in major US rivers (e.g. Smith *et al.* 1987). Data covering the years 1974-1986 were available from many of the statutory authorities and provided an opportunity to compare trends derived from the shorter period and to evaluate the impact of the extreme 1975-1976 drought on recent trends in $\text{NO}_3\text{-N}$ concentrations. An evaluation of precipitation and runoff records for selected British rivers during the two study periods did not reveal any systematic fluctuation in hydrometeorological conditions that could readily explain trends in $\text{NO}_3\text{-N}$ concentrations.

1980-1986 period

Trend analysis revealed a significant increase, a significant decrease, and no significant change in $\text{NO}_3\text{-N}$ concentrations at 51, 11, and 38% of the 743 sites, respectively (Fig. 3A). At 430 sites analyzed for trends in $\text{NO}_3\text{-N}$ loads, a significant increase, a significant decrease, and no significant change was detected at 34, 17, and 49% of them for the period 1980-1986, respectively (Fig. 3B). A trend of decreasing concentrations was found mainly in the Thames Basin, parts of northern Scotland, and Southwest England. The latter region also exhibited several adjacent sites where $\text{NO}_3\text{-N}$ loads decreased during the 7-year period. Recent decreases in $\text{NO}_3\text{-N}$ concentrations in parts of the UK may be caused by the 'clean up' campaigns of the statutory authorities, which have encouraged the agricultural community to take more care in preventing point-source pollution of water courses.

Rates of increase in annual mean concentration were highest in lowland regions at elevations of <100 m, and the largest increases ($>0.2 \text{ mg l}^{-1}\text{year}^{-1}$) occurred in parts of East Anglia and the Midlands (Fig. 3A), where agricultural activity is most intensive and mean $\text{NO}_3\text{-N}$ concentrations also are the highest in the country. With respect to relative changes in concentration, concentration increases which occurred in the agricultural lowlands of Britain, generally, were less than 10%. In contrast, concentration increases greater than 10% were common at sites in the upland areas of Northern England and the Scottish Highlands, where the environment, characterized by low $\text{NO}_3\text{-N}$ concentrations, is more sensitive to changes which promote accelerated nitrogen losses in rivers. The recent increase in $\text{NO}_3\text{-N}$ loads, expressed as a percentage of mean annual load for the period 1980-1986, also exceeds 10% for many rivers in the uplands of northern and western Britain, although the greatest rates of increase ($>2.0 \text{ kg ha}^{-1} \text{ year}^{-1} \text{ year}^{-1}$) were found in eastern and Southwest England (Fig. 3B). However, the difference in recent concentration increases between lowland and upland areas, in general, has less impact on loads.

A seasonal analysis of $\text{NO}_3\text{-N}$ concentrations suggests that, on average across the UK, the increase of winter concentrations ($0.159 \text{ mg l}^{-1} \text{ year}^{-1}$) during the period 1980-1986 was almost double that of summer concentrations ($0.089 \text{ mg l}^{-1}\text{year}^{-1}$). In contrast, winter loads over the same period, decreased, on average, $-1.041 \text{ kg ha}^{-1} \text{ year}^{-1} \text{ year}^{-1}$, but summer loads increased, on average, $1.378 \text{ kg ha}^{-1} \text{ year}^{-1} \text{ year}^{-1}$, which balances the winter decrease.

Multiple regression analysis was performed to establish relations between trends in $\text{NO}_3\text{-N}$ concentrations and loads at the sites and the physiographic, hydrometeorological and land-use characteristics of the associated catchment (Betton, 1990). This analysis, however, proved disappointing, in that the regression models did not explain much of the variance in corresponding concentrations or loads. Furthermore, few statistically significant relations resulted. The significant positive relations were found between the magni-

tude of the concentration trend and the extent of dairy farming in the catchment, and between the magnitude of the load trend and the proportion of Grade 1 agricultural land in the catchment. These relations suggest that the largest recent increases in $\text{NO}_3\text{-N}$ concentrations may be caused by the intensification of land use in catchments, which is characterized by good quality agricultural land, especially grassland, where there has been an increasing tendency to apply more fertilizer (Royal Society, 1983). However, an analysis of the controls on recent trends in $\text{NO}_3\text{-N}$ concentrations in British rivers is hampered by a lack of appropriate information at the catchment scale on previously identified factors controlling $\text{NO}_3\text{-N}$ concentrations, such as changing rates of fertilizer usage and fluctuations in atmospheric deposition of nitrogen (Roberts & Marsh, 1987; Smith *et al.* 1987).

1974-1986 period

Data were available for the 13-year period at 425 and 254 stations for $\text{NO}_3\text{-N}$ concentrations and loads, respectively. A comparison of the rate of change of concentration and loads between the 1974-1986 and 1980-1986 periods at these sites, in general, revealed (1) no

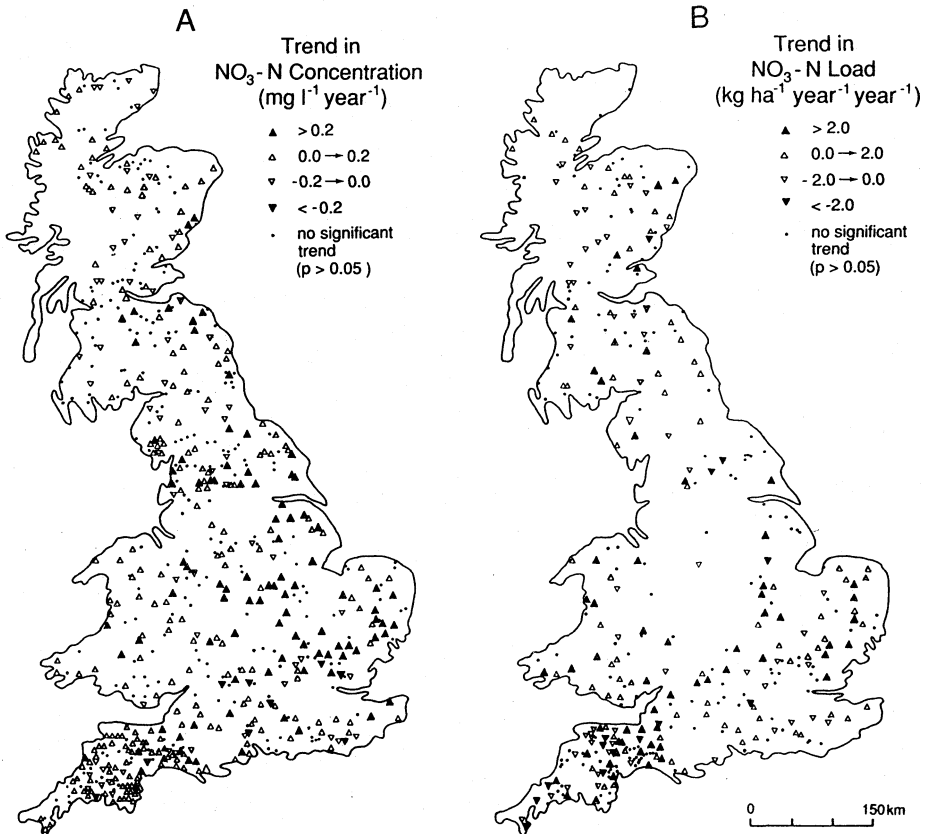


FIG. 3 Trend in $\text{NO}_3\text{-N}$ concentration (A) and load (B) in British rivers in the period 1980-1986.

change in direction of trend between the two periods, and (2) small differences in the magnitude of change between the two periods. The trend in concentration and in load became significant at a few additional stations when the longer time period was considered, but there also were many sites where a statistically significant trend was evident only for the 1980-1986 period (Figs 4A & 4B). The latter phenomenon reflects the impact of the 1975-

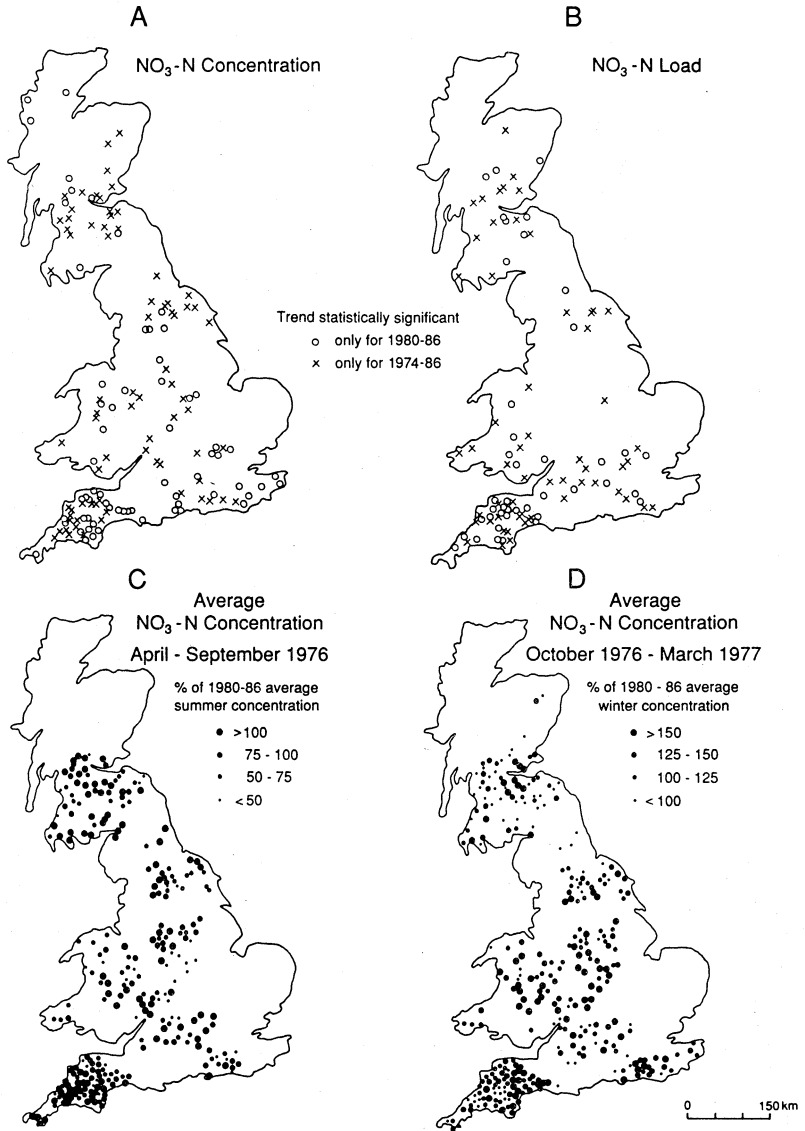


FIG. 4 Comparison of NO₃-N concentration (A) and load (B) trend significance in 1980-1986 and 1974-1986, and magnitude of average NO₃-N concentration during drought (C) and post-drought (D) periods in comparison with longer-term mean concentrations.

1976 drought, which was a major hydrological perturbation affecting Britain (Doornkamp *et al.* 1980) and had a pronounced effect on stream $\text{NO}_3\text{-N}$ concentrations (Slack, 1977; Walling & Foster, 1978).

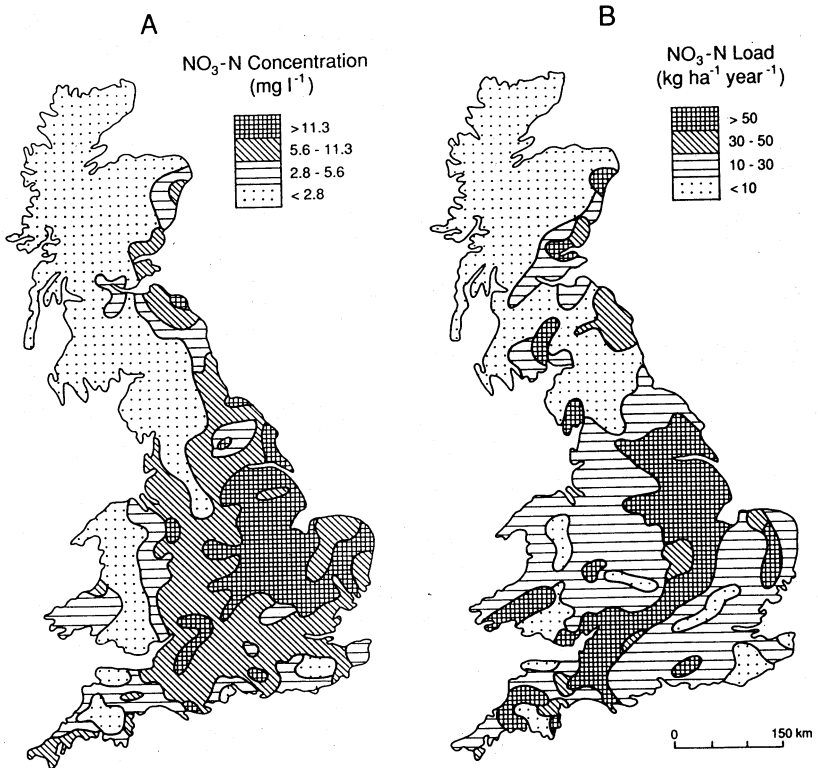


FIG. 5 Average $\text{NO}_3\text{-N}$ concentrations (A) and loads (B) predicted for the Year 2000 in British rivers.

The 1975-1976 drought did not affect streams uniformly across Britain and, therefore, had a varying impact on trends in $\text{NO}_3\text{-N}$ concentrations. At the height of the drought in the summer of 1976, mean $\text{NO}_3\text{-N}$ concentrations from April to September in rivers of selected statutory authority regions were less than 50% of 1980-1986 average summer concentrations in much of the English Midlands but exceeded those in parts of southern Scotland (Fig. 4C) where the effect of the drought was reduced by thunderstorms. The immediate post-drought period, October 1976-March 1977, was exceptionally wet and was associated with rapid fluctuations in $\text{NO}_3\text{-N}$ concentrations as caused by the flushing of $\text{NO}_3\text{-N}$ that accumulated during the protracted dry spell. Mean concentrations for this period exceeded the mean winter concentrations for 1980-1986 period by 150% in central Wales, the Midlands, South Yorkshire, and east-central Scotland. Mean concentrations were less than the longer-term mean winter values typically in parts of Scotland, Yorkshire and southern Britain (Fig. 4D), where high rainfall and runoff in the winter after the drought diluted the $\text{NO}_3\text{-N}$ in the rivers. Similar contrasts are evident for $\text{NO}_3\text{-N}$ loads for the drought and post-drought period. Loads generally increased across the country in the winter

of 1976-1977 but differences between the drought and post-drought period were most extreme for the Northumbrian Water Authority area. In this area, mean loads from April to September 1976 were only 10% of the long-term mean summer concentrations, but from October 1976 to March 1977 were 375% of 1980-1986 average winter concentrations. $\text{NO}_3\text{-N}$ concentrations immediately after the drought were the highest recorded in the 1974 to 1986 period. These high loads are associated with concentrations higher than those previously monitored in rivers with long-term records (e.g. Slack & Williams, 1985). The drought is likely to have an impact which will continue for several years and it complicates interpretation of factors controlling long-term trends in $\text{NO}_3\text{-N}$ concentrations and loads.

IMPLICATIONS

The present study of UK rivers has revealed that recently $\text{NO}_3\text{-N}$ concentrations have significantly increased at about 50% of the sampling sites analyzed, and confirms predictions that maintenance of current trends would result, on average, in an average increase from 2 to 3 mg l^{-1} for $\text{NO}_3\text{-N}$ concentrations over the next decade (e.g. STACWQ, 1984). However, the results also suggest that, on average, $\text{NO}_3\text{-N}$ concentrations are either constant or decreasing at 50% of many headwater sites. These results suggest that caution is necessary regarding concerns about increasing nitrogen in larger river systems. An analysis of a limited amount of data indicate that significant increases in $\text{NO}_3\text{-N}$ loads have occurred over recent years at about 33% of the study sites.

Extrapolation of trends computed for the period 1980-1986 to the Year 2000 (Fig. 5) suggests that much of Eastern England where mean concentrations exceed the EEC acceptable level (11.3 mg l^{-1}) may be a particular problem and that a zone from Somerset to Lincolnshire may be affected by high mean annual loads ($>50 \text{ kg ha}^{-1} \text{ year}^{-1}$).

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