
The Water Industry and the Nitrogen Cycle [and Discussion]

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The water industry and the nitrogen cycle

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Owing to a rising trend of nitrate concentrations in many rivers and groundwaters that are used for water supply and at the same time the introduction of legislation that tightens limits, the water industry's attention has been focused particularly on this component of the nitrogen cycle. In some rivers in eastern England that are used for potable water supply, nitrate concentrations have, after extreme meteorological conditions, exceeded 100 mg l^{-1} for short periods and, as a consequence, supply abstractions have at times been curtailed. The nitrate concentration in at least 100 groundwater sources in the United Kingdom exceeds 50 mg l^{-1} either continuously or intermittently, and several of the wells most severely affected have been withdrawn from service and replaced by other sources.

Over the last few years the water industry has mounted a substantial programme of investigations directed towards:

- (a) identifying the source of nitrate and the reasons for the rising trends in surface and groundwater;
- (b) predicting the rate at which nitrate concentrations will rise in future and the eventual levels;
- (c) controlling the problem by blending of supplies, treatment to remove nitrate, storage, and other methods, and the cost of such options.

1. INTRODUCTION

The water industry has a direct effect on the nitrogen cycle through transformations in the N compounds brought about from activities such as sewage treatment, sewage sludge disposal to land, and water storage. This is relatively small in comparison with the effect that agricultural land use or natural processes such as fixation of N_2 have on the cycle. However, changes in the cycle that have occurred in the last 45 years, particularly those relating to agricultural land use, are now having a considerable impact on the quality of freshwater, and this is a major concern to the water industry.

Increasing concentrations of nitrate in many rivers and groundwaters that are used for water supply have been observed (S.T.A.C.W.Q. 1979). The introduction of legislation imposing tighter quality limits has caused the water industry to focus its attention on nitrate. Other problems that may result from an increasing input of nutrients to lakes and reservoirs leading to algal blooms and possibly associated treatment problems have not been overlooked (Youngman & Lack 1981).

The nitrate problem is a very real one and in some rivers in eastern England that are used for potable water supply, nitrate concentrations have, for short periods after extreme meteorological conditions, exceeded the W.H.O. maximum recommended limit ($22.6 \text{ mg NO}_3\text{-N l}^{-1}$) and, as a consequence, supply abstractions have been curtailed. The nitrate concentrations

in at least 100 groundwater sources in the United Kingdom either continuously or intermittently exceed the W.H.O. acceptable limit ($11.3 \text{ mg NO}_3\text{-N l}^{-1}$) and several of the wells most severely affected have been withdrawn from service and other sources of water have had to be substituted. The industry has been sufficiently concerned to mount a substantial programme of investigation directed towards:

identifying the sources of nitrate and the reasons for the rising trends in surface and groundwater;

predicting the rate at which nitrate concentrations will rise in future and the eventual levels;

controlling the problem by blending supplies, treatment to remove nitrate, storage and other methods;

determining the cost of such options.

Many organizations have been involved in these investigations with the work coordinated by the Nitrate Sub-Committee of the Department of the Environment/National Water Council, Standing Technical Advisory Committee on Water Quality. After the drought of 1976 a report dealing particularly with the treatment aspects was prepared by S.T.A.C.W.Q.'s sister committee concerned with water treatment.

The purpose of this paper is to review these investigations and to identify areas of uncertainty where additional work is required.

2. PUBLIC HEALTH STANDARDS FOR NITRATE AND NITROGEN COMPOUNDS IN DRINKING WATER

The 1945 Water Act placed a statutory duty on water supply undertakings in England and Wales to supply 'wholesome' water for domestic use; the responsibility for ascertaining the 'wholesomeness' of supplies rests with the local authority, and compliance in respect of these duties is a matter of legal interpretation. The water industry reorganization after the 1973 Water Act incorporated the requirements of the 1945 Act.

Regional Water Authorities, in common with all pre-1973 water undertakings, adopted the W.H.O. European Standards for Drinking Water (1970) as their guidelines for assessing the 'wholesomeness' of public supplies; the standard for nitrates is detailed as follows.

Less than $50 \text{ mg l}^{-1}(\text{NO}_3)$ ($11.3 \text{ mg NO}_3\text{-N l}^{-1}$): 'recommended'.

$50\text{--}100 \text{ mg l}^{-1}(\text{NO}_3)$ ($11.3\text{--}22.6 \text{ mg NO}_3\text{-N l}^{-1}$): 'acceptable'.

If the nitrate content is within the acceptable range and the water is otherwise chemically and bacteriologically satisfactory, it may not give rise to trouble, but physicians in the areas should be warned of the possibility of the occurrence of infantile methaemoglobinaemia.

Greater than $100 \text{ mg l}^{-1}(\text{NO}_3)$ ($22.6 \text{ mg NO}_3\text{-N l}^{-1}$): 'not recommended'.

The recommendations were reiterated in 1977 and a rider was added to the effect that infants who are bottle-fed should be supplied with water containing less than 50 mg l^{-1} ($11.3 \text{ mg NO}_3\text{-N l}^{-1}$). W.H.O. guidelines for drinking water quality are currently being redrafted and may provide a guideline level for combined nitrate-nitrite of 10 mg l^{-1} expressed as N (45 mg l^{-1} nitrate).

The E.E.C. adopted a Community directive on 'the quality of water intended for human consumption' in 1980. This directive specifies a guide level of 25 mg l^{-1} nitrate ($5.7 \text{ mg NO}_3\text{-N l}^{-1}$) and a maximum acceptable concentration of 50 mg l^{-1} nitrate ($11.3 \text{ mg NO}_3\text{-N l}^{-1}$). If the guide level is exceeded, bottled water should be provided for infants. E.E.C. directives are binding upon Community members, and U.K. water industry legislation is such that this

directive can be accommodated within U.K. law without additional parliamentary legislation. The U.K. water industry is thus faced with mandatory water quality standards at the consumer's tap for the first time. Derogations are available in respect of certain criteria 'due to the nature and structure of the ground', provided there is no identified detriment to public health. Water authorities in the U.K. are currently awaiting advice from the Department of the Environment on the interpretation of this directive and it is evident that too rigid an interpretation would require the expenditure of many millions of pounds to achieve the specified nitrate standards at all times.

No direct health hazard has been demonstrated due to the presence of ammonia in drinking water but it must be removed during treatment so as not to interfere with disinfection by chlorine. The 'Water for human consumption' directive specifies a maximum admissible concentration of $0.5 \text{ mg l}^{-1} \text{ NH}_4^+$.

A further E.E.C. directive concerning the quality of surface water intended for abstraction for the supply of drinking water in member states became Community law in July 1977. The requirements are compatible with those of the 'human consumption' directive, although there is some latitude in that a 95% (of samples) compliance with the mandatory 50 mg l^{-1} nitrate ($11.3 \text{ mg NO}_3\text{-N l}^{-1}$) level is specified.

The W.H.O and E.E.C. standards express nitrate concentrations as NO_3 , but in the remainder of this paper nitrate is expressed as equivalent N.

3. SOURCES OF N IN FRESH WATER

The principal sources of N compounds in fresh surface waters and groundwater are rainfall, leaching of agricultural soils and sewage effluent discharges.

(a) *Rainfall*

Fixed N in precipitation may make a significant input to the land surface. However, while comprehensive data are available at the meteorological office on rainfall quantity and its distribution, measurements of the chemical quality of rainwater and temporal variations are very limited. It is possible to show in a broad way that N in rainfall varies across the U.K., generally being highest close to industrial areas. There is also evidence from the few long-term records to show a significant increase in the mineral N content of rain with time (Russell & Richards 1919; Williams 1976). This is most probably related to the increased use of fossil fuel. The average concentration in rainfall for England and Wales is about 1 mg N l^{-1} , which represents an input to the land surface of about 10 kg ha^{-1} annually. If these average values are extended to the U.K. as a whole, they give an annual contribution of about 260 kt of inorganic N.

(b) *Leaching of agricultural soils*

Land drainage is an important factor governing nitrate concentrations in fresh waters. Nitrogen may be lost from the soil-plant system in a gaseous form to the atmosphere, in crop removal, or by leaching below the root zone where it passes either to a surface water course or to deep aquifer drainage. Although there have been relatively few detailed studies on catchments in the U.K. that allow precise values of N losses in drainage water to be calculated, there is a substantial amount of information from lysimeter and plot studies. It is thus possible to identify the most important factors influencing leaching. Usually almost all the N in drainage

water is present as nitrate, but exceptions occur where livestock excreta or crop wastes cause direct contamination of surface water.

A considerable quantity of N is present in the soil down to the base of the root zone, but this is held in an organic form and is only released slowly by mineralization. To obtain optimum crop yields it is necessary to add N fertilizer and in the U.K. some 1 Mt of inorganic N is applied annually. An equivalent amount of organic N is added in animal excreta. The rates of application of inorganic fertilizers to root crops and cereals currently range from 90 to 200 kg N ha⁻¹, having increased by between five and ten times during the past 35 years.

TABLE 1. NITRATE CONCENTRATIONS IN DRAINAGE WATER AND THE EQUIVALENT N LOAD, ASSUMING AN EFFECTIVE RAINFALL OF 300 mm

	[NO ₃ -N] mg l ⁻¹	N load kg ha ⁻¹
rough grazing, including mountains, moorland and unfertilized grassland	< 1-2	< 3-6
fertilized grassland	4-10	12-30
fertilized arable land	10-40	30-120

The amounts of N leached are affected by the crop, meteorological conditions, cultivation methods, the amount of fertilizer applied and the drainage conditions. Values in drainage water generally fall within the ranges given in table 1.

Well drained soils with an arable régime remain well aerated, and losses by denitrification appear to be small, while mineralization of organic matter occurs readily. However, poor drainage of soils, particularly in the wetter parts of the country, may lead to anaerobic conditions with denitrification in the lower soil layers. The difference between the grass and arable land in such situations may not be as great as the above values indicate.

It is not possible to assess at present what proportion of inorganic N fertilizer is leached from the soil under long-term equilibrium conditions. The results of experiments with the aid of isotopically labelled N applied to lysimeters are awaited. However, empirical evidence indicates that in well drained arable soil an equivalent of about 40-50% of added inorganic fertilizer may be leached.

Another important source of nitrate may result from the ploughing of established grassland, leading to the mineralization of the organic N. For example, ploughing of a virgin grassland plot on the Chalk in Sussex released 200 kg N ha⁻¹ of nitrate during 1978-9, whereas less than 5 kg N ha⁻¹ was lost from adjacent unploughed grass (C.P. Young, personal communication 1981). A marked feature of British agriculture over the last 100 years has been the conversion of pasture to arable land. In the 1900s about 30% of the land in England and Wales was under arable cultivation and 40% under grass. During the 1940s a major shift occurred: arable acreage increased to 40% and grassland was reduced to 30%. Another important factor to be considered is the increase in drainage work that has taken place in recent years. Whether or not this will lead to a substantial increase in the N in surface waters is still uncertain.

It is widely accepted that agricultural activities make a most significant contribution to the nitrate concentrations in surface and groundwater. Detailed quantification of the loss of N from agricultural and other land by leaching is not possible, but crude estimates give a figure in excess

of 700 kt per year. The losses tend to be greatest during the autumn and winter months at times of high river flow and when, owing to low temperatures, there is little denitrification. During these months the water authorities are replenishing surface water storage and the occurrence of the maximum concentration of nitrate at this time causes problems.

(c) *Sewage*

Sewage effluent from about 35 million people in the U.K. is discharged to the non-tidal reaches of rivers. The remainder, from about 21 million people, goes to estuaries or the sea. There are about 4500 separate sewage treatment works. A high proportion of N in sewage is derived from human excreta. Each person produces about 10 g of N per day. Nitrogen occurs as soluble salts in sewage and is also contained in solid faeces. During wastewater treatment, solid material is removed for disposal as sludge and this decreases the amount of N in the sewage to a lower daily amount of about 8 g per person.

In a survey of 65 sewage treatment plants in the U.K. (E. J. Tomlinson, personal communication 1981), the average concentration of inorganic N in the effluents was 25 mg N l⁻¹, of which 60% was in the ammonium form and 40% was nitrate. These proportions can vary widely depending on the works and the circumstances of discharge. At the Rye Meads plant on the river Lee, for example, the effluent during the period 1974–8 was well oxidized and 99% of the N was present as nitrate. The concentrations of inorganic N are reduced by dilution as the effluent enters a water course. During the summer months the ammonia may be relatively rapidly oxidized to nitrate (Garland 1981), and further the nitrate may be reduced by biological denitrification and the growth of algae. In autumn and winter the natural nitrification and denitrification processes taking place in rivers slow down, and the sewage works may also have difficulty in producing a nitrified effluent, thus causing ammonia levels in rivers to increase.

In recent years ammonia levels have tended to fall as sewage treatment has become more efficient (Lester 1971), and river pollution generally has been subject to much closer control. Ammonia levels in relatively unpolluted rivers such as the Severn and Thames are under 0.5 mg l⁻¹ on average, while more polluted rivers such as the Trent had an average of 1–1.5 mg l⁻¹ ammonia up to the mid to late 1970s. Consequently, in most river systems in the U.K. the total oxidized N may be equated to the nitrate N with little loss of accuracy.

Exceptions to the general situation are the rivers Thames and Lee, where the proportion of sewage effluent may exceed 50% under conditions of low flow. However, even during the peak nitrate period of December 1976 to February 1977 the sewage contribution to the nitrate load in these rivers was not greater than 15%.

It is estimated that 155 kt of N from sewage treatment plants are discharged annually to the rivers in the U.K. and that about 88 kt are disposed of directly to estuaries or to the sea.

Almost 70% of the sewage sludge from treatment plants is either used as a fertilizer and soil conditioner on agricultural land or deposited in landfill. The remainder is dumped in the sea. Sludge contains 3–4% N. The total amount of nitrate applied to agricultural land in this way is about 20 kt per year. The N would be combined with organic matter or be present as ammonia, but slow oxidation would eventually convert much of this to nitrate.

4. TRENDS IN NITRATE CONCENTRATIONS IN SURFACE WATERS AND GROUNDWATER

In the context of public water supplies the immediate need of any water undertaking is to know when nitrate concentrations in their sources are likely to exceed the limits of $11.3 \text{ mg NO}_3\text{-N l}^{-1}$ and $22.6 \text{ mg NO}_3\text{-N l}^{-1}$. To maintain wholesome supplies it is necessary to study historic trends and predict future nitrate levels so that medium- to long-term strategies can be developed.

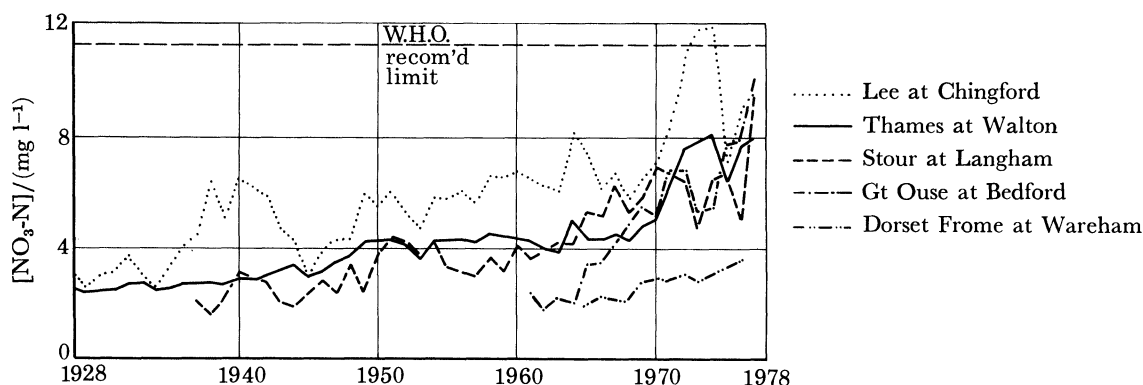


FIGURE 1. Trends in mean annual nitrate concentration in five rivers for which long-term data are available. (After Marsh (1980).)

(a) Surface water

Information on the quality of river water in the U.K. is collected by the water authorities in England and Wales, the Department of the Environment in Northern Ireland, and the River Purification Boards in Scotland. Many of the collection points form part of the network of 245 Harmonized Monitoring Stations for the sampling of a wide range of determinands, including inorganic but excluding organic N. This scheme fulfils certain international obligations. A wealth of contemporary data on river water quality is thus being collected, but historical data on which to assess long-term trends is rather limited.

Nevertheless, a number of sites with long-term records do exist (figure 1) and the progressive increase in the nitrate concentrations in these river waters over the last 20 years is evident (S.T.A.C.W.Q. 1979).

An analysis of 12 rivers for which data are available for a 20-year period shows nitrate concentration increasing by between 50 and 400%. The River Thames, for example, which provides the major water supply for London, has shown an increase in mean annual nitrate concentrations from 2.5 mg N l^{-1} in 1928 to 8 mg N l^{-1} in 1978.

Figure 2 shows the mean annual nitrate concentration in the principal rivers in England and Wales. There is a strong regional pattern. The highest values are found in the Midlands and the southeast of England and the lowest in more mountainous regions in the north of England, the Lake District, Wales and the northwest of England. By combining this information with the mean annual flows in the rivers, it is possible to estimate the total nitrate load reaching tidal waters. This calculation has been carried out with some precision for England and Wales

by Marsh (1980) to give a value of 275 kt of nitrogen per year for the period 1974–7. Much cruder estimates for Scotland and Northern Ireland give 60 and 15 kt per year respectively.

From the point of view of water supply, a complicating factor is that there may be considerable year-to-year and seasonal variation in the concentration of nitrate in rivers, reflecting climatic influences (figure 3). Public water supply relies heavily on river abstraction in the southeast of England and the Midlands, and these high values approaching or occasionally exceeding the limits for nitrate give the water undertakings cause for concern in that the application of water supply nitrate standards could impose restrictions on the availability of these resources in the future.

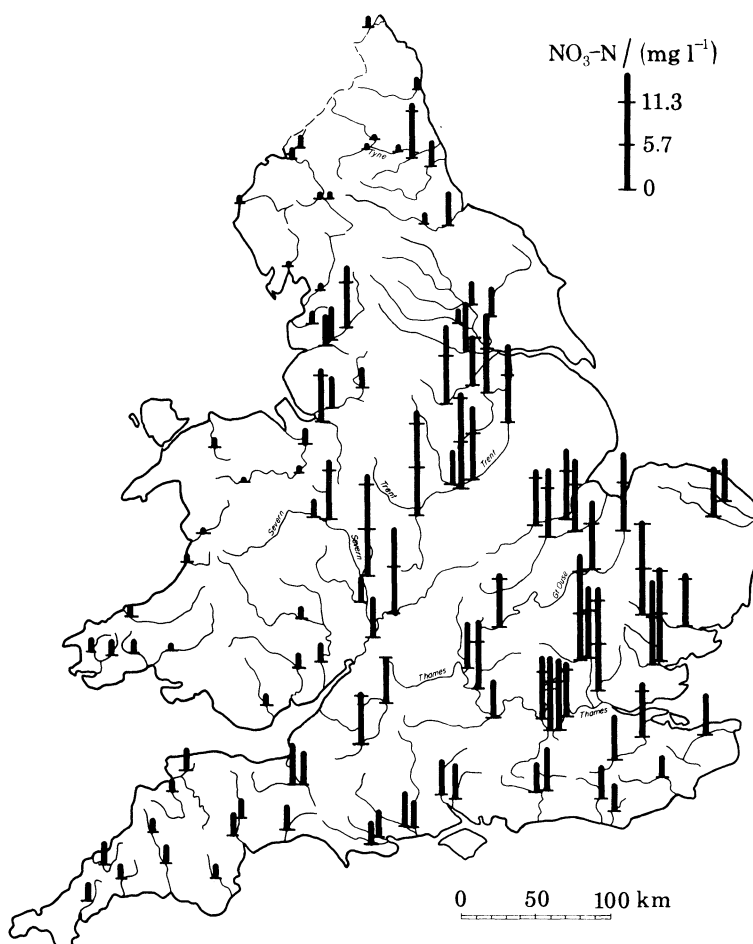


FIGURE 2. Mean nitrate concentrations for the period 1974–7 for rivers in England and Wales. (After S.T.A.C.W.Q. (1979).)

The Thames Water Authority has developed a lumped parameter model based on land use history for the Thames catchment (13 100 km²) to forecast future trends (Onstad & Blake 1980). Figure 4 gives a typical set of results and shows that if the present land use practice continues, the average annual nitrate concentration in the river would exceed 11.3 mg N l⁻¹ in the mid-1990s. Maximum concentration during a year would of course be well in excess of 11.3 mg N l⁻¹, but it is not possible to predict this at present. The models are being improved and a much more deterministic approach is being adopted.

In the Anglian Water Authority, a small ($13 \times 10^3 \text{ m}^3$ per day) spring-fed direct river abstraction from a wholly agricultural catchment is predicted to reach 100 mg l^{-1} nitrate by the late 1980s. For the remainder of the region, statistical predictions (A. E. Warn, personal communication 1981) indicate that the major reservoir resources are 'safe' but that seasonal effects related to high natural flows will continue to cause nitrate quality problems of short duration (days to weeks) for direct river abstractions with a slight progressive increase in severity.

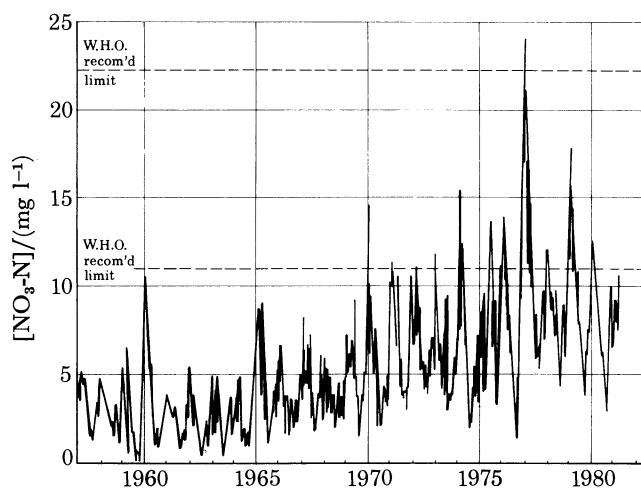


FIGURE 3. Concentrations of nitrate in the river Great Ouse, showing seasonal variations and a long-term trend.

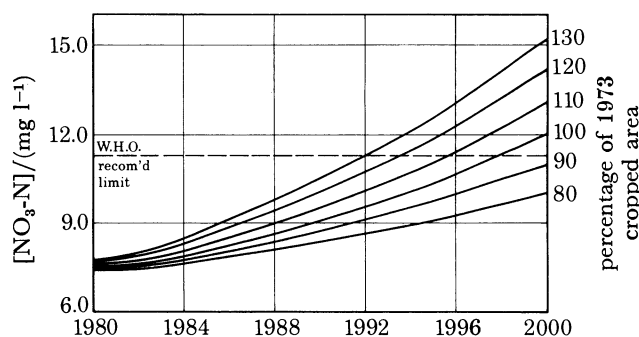


FIGURE 4. Model predictions of the average annual nitrate concentrations in the river Thames at Walton, assuming 1.5% annual growth in production per unit area. (After Onstad & Blake (1980).)

For many rivers a lack of long-term historical data makes nitrate predictions difficult and effort has tended to concentrate on those rivers with an immediate problem. However, more deterministic predictive models requiring fewer data are being developed by the water industry.

The increase in the nitrate concentrations in rivers has also led to an increase in nitrate concentrations in reservoirs, although the effects are less marked. Appreciable denitrification, possibly giving a 50% decrease in nitrate, occurs within the stored water body from early summer through to late autumn. The effect is demonstrated in figure 5 for Grafham Water, which receives a pumped input from the Great Ouse. River water storage in reservoirs accounts for a large proportion of water used for public supplies, and when nitrate concentrations are high it is important to be able to predict the amount of denitrification that will occur so that water supply sources can be most effectively managed. Simulation models have been developed

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for this purpose (Rutherford & Davis 1979). For natural impoundments the potential control options are more limited, and with short-term storage, when no control of the input volumes and nitrate loads is practicable, the means of control is in water treatment.

(b) Groundwater

Groundwater accounts on average for about 30% of the public supply in England and Wales. In some regions of southern England the proportion may be as high as 70%. Available long-term data on the concentration of nitrate in groundwater are less common than for surface waters.

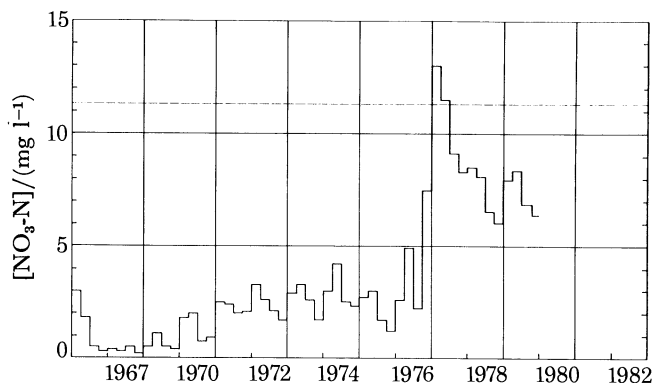


FIGURE 5. Quarterly mean concentrations of nitrate in Grafham Water (Anglian Water Authority pumped storage reservoir) for the period 1967-77.

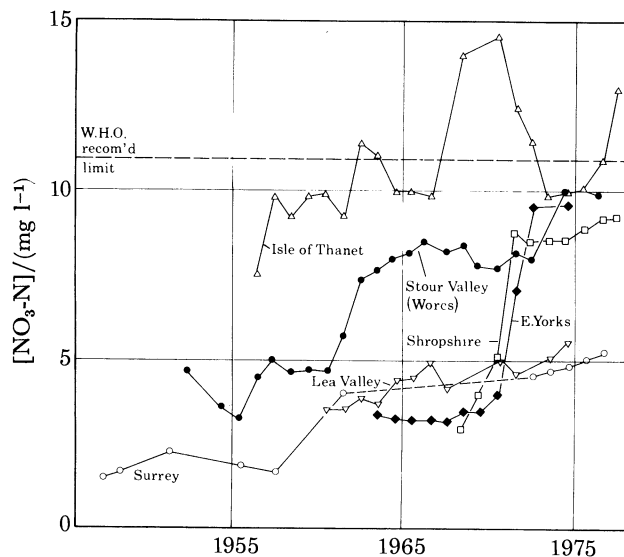


FIGURE 6. Nitrate concentrations in selected public water supply abstraction boreholes in the Chalk and Triassic sandstone aquifers in the U.K.

Nevertheless there is a marked upward trend in some catchments (figure 6). In certain areas W.H.O. limits have been exceeded and wells have been temporarily shut down (Young *et al.* 1979; Greene 1980). These increases are particularly serious in the dry eastern areas of England. In the Anglian Water Authority area, for instance, the problem is very evident in some of the sources abstracting from the Lincolnshire Limestone aquifer (figure 7).

Extensive field and laboratory investigations have been undertaken by the water industry

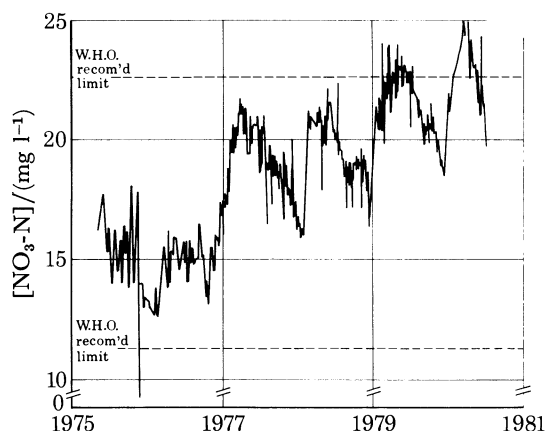


FIGURE 7. Nitrate concentrations in a public water supply well in the Lincolnshire Limestone.

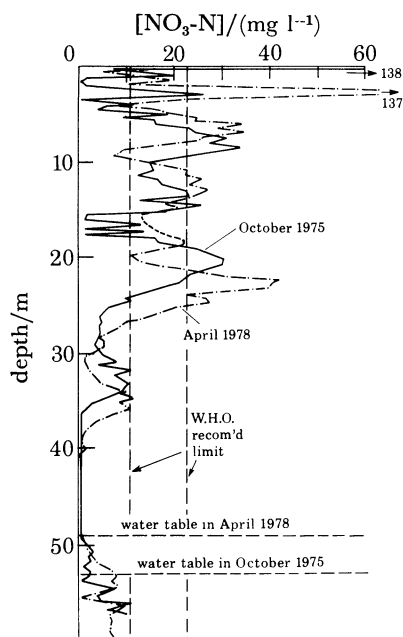


FIGURE 8. Variations in nitrate concentrations with depth in the pore water of the unsaturated and saturated zones of the Chalk at an arable site in Hampshire. The profile was originally measured in October 1975 and redrilled and sampled in April 1978, by which time a downward movement of about 3 m had occurred.

to determine the extent of the problem, predict future concentrations and propose solutions. The investigations have shown a clear relation between the nitrate concentration in the pore water of the unsaturated zones of the Chalk and Triassic sandstone aquifers (where they are unconfined) and historic land use (Young & Gray 1978). High concentrations of nitrate ($15\text{--}50\text{ mg NO}_3\text{-N l}^{-1}$) occur below fertilized arable land (figure 8) and lower concentrations (generally less than $3\text{ mg NO}_3\text{-N l}^{-1}$) below long-term unfertilized grassland (figure 9) or woodland. On the basis of these findings it is estimated that there may be some 2–3 Mt of nitrate currently stored in the unsaturated zones of the principal aquifers in the United Kingdom (Chalk and Triassic sandstone). The important questions are: What is the rate at which this nitrate is moving down through the unsaturated zone? and What is its future impact on

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groundwater supplies? The exact mechanism governing the movement of water and solutes through the unsaturated zone is currently the subject of much debate (Oakes 1981; Agricultural Research Council 1978; Barker & Foster 1981).

Several investigation sites have now been redrilled after a period of several years (figure 8) (Young *et al.* 1979), and these have shown that the nitrate is moving down at a rate of about 1 and 2 m per year in the Chalk and Triassic sandstones, respectively. On the basis of such

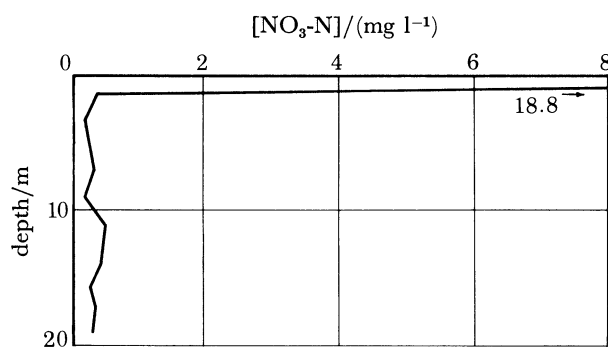


FIGURE 9. Variations of nitrate concentration with depth in the pore water of the unsaturated zone of the Chalk at an unfertilized permanent grassland site.

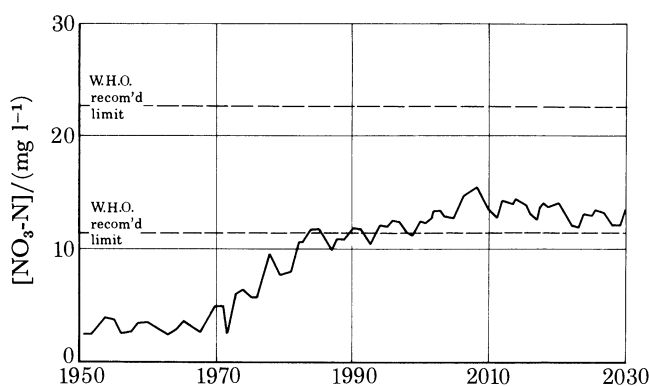


FIGURE 10. Model predictions of the change in nitrate concentrations in the groundwater of a Chalk catchment draining to the river Itchen, Hampshire.

studies, mathematical models have been developed to predict the future nitrate concentrations at any point within a groundwater catchment (Young *et al.* 1976; Oakes 1981). An example of such a prediction for a groundwater catchment draining to the River Itchen is shown in figure 10. This catchment is typical of many in the south of England and these predictions give cause for concern.

Most groundwater investigations have been concerned with aquifers at their outcrops because it is here that nitrate concentrations may be particularly high. Groundwater pumped from confined aquifers, that is aquifers covered by more impermeable strata, have very low nitrate concentrations. It has been suggested that, where possible, relocation of wells in the confined zones of aquifers may partially alleviate the nitrate problem. This, however, may only be a short-term solution because high-nitrate water may be drawn from the outcrop areas. Research into the possibility of developing the confined zones of aquifers in this way is proceeding.

5. REMEDIAL MEASURES AND COSTS

Methods of overcoming the problem of high nitrate concentrations in water supplies depend upon the nature of the supply and duration for which high (unacceptable) nitrate levels persist. Remedial options include blending with low nitrate supplies, water treatment by ion exchange or biological denitrification, the provision of raw water storage, or the supply of low-nitrate bottled water for infants. Realistically, biological denitrification and the provision of raw water storage for induced natural denitrification are only applicable to surface water supplies where 'downstream' physico-chemical treatment facilities already exist. For groundwater supplies the remedial options are therefore restricted to blending, ion exchange treatment or the provision of bottled water for infants. For river gravel abstraction, any of these remedies may be extremely expensive and the possibility of aquifer land purchase and restriction of farming activities should not be immediately discounted.

(a) Blending

This solution depends upon the availability of low-nitrate supplies within reasonable proximity. It was adopted by the Anglian Water Authority for Bedford's river-derived supplies in the post-1976 drought period when nitrate levels reached a peak of $24.1 \text{ mg NO}_3\text{-N l}^{-1}$. Supplies from Grafham reservoir were used for dilution purposes. Similar solutions were adopted by Essex Water Company when supplies from the rivers Stour and Chelmer exceeded $22.6 \text{ mg NO}_3\text{-N l}^{-1}$ for 3 months with the maximum recorded concentrations on the Stour approaching $30 \text{ mg NO}_3\text{-N l}^{-1}$. More recently the Anglian Water Authority have spent more than £4M on a 'spine' main in central Lincolnshire to provide low-nitrate Bunter sandstone groundwater for blending with the troublesome central Lincolnshire Limestone borehole sources (figure 7).

In general, blending is a fairly expensive option owing to the high capital costs of mains and new source developments, but where an adequate source of low-nitrate water is available within a short distance there is little need to examine other options.

(b) Raw water storage

Raw water storage can ensure a supply of low-nitrate raw water for blending or exclusive use when the nitrate concentration exceeds a specific limit. The size of the reservoir should be determined by the duration of the high nitrate concentration in the river and the water supply demand. For short-term reservoir storage of less than 6 months duration, little reliance can be placed upon natural denitrification effects and the reservoirs would need to be managed such that an adequate supply of low-nitrate water was available at the beginning of the late autumn–winter problem period. This is contrary to the standard practice of using stored water in dry periods and refilling in autumn. Such a policy could impose quality constraints on the reliable yield of reservoirs.

*(c) Treatment for removal of nitrate**(i) Ion exchange*

Following the development of pilot plant at the Water Research Centre for the removal of nitrate by ion exchange (Gauntlett 1975), the Anglian Water Authority has initiated full-scale plant evaluations (Greene 1981). A continuous ion-exchange process was built in Lincolnshire

and a fixed-bed system incorporating countercurrent regeneration was installed in west Norfolk. Both plants used strong base anion-exchange resin, regenerated by brine. Each plant cost approximately £120 000 (1976 prices). The resins have a poor selectivity for nitrate in preference to sulphate and this resulted in high running costs (£0.005–0.012 m⁻³ treated) and effluent disposal problems in areas where chloride discharged to water courses could severely affect agricultural spray irrigation. It has been shown that a 40% reduction in brine effluent volumes is possible by using reverse osmosis to treat the rinse waters, and on this basis this treatment method is economically viable for plant located more than 15 miles from the coast. Nevertheless the high cost of transporting disposable waste by tanker to the nearest large river or sea outfall could double product water treatment costs. The success of the groundwater ion-exchange projects and the potential control difficulties with the biological denitrification process prompted the Anglian Water Authority to look at the application of ion exchange to nitrate removal from surface waters. This work was commissioned under contract with Hatfield Polytechnic in 1978 and was completed early in 1981. The results show considerable promise, particularly with regard to ease of plant control and the absence of difficulties associated with effluent disposal during the winter when river flows and nitrate concentrations are high.

(ii) *Biological denitrification*

Biological denitrification systems take advantage of the potential for induced development of facultative heterotrophs which, in the absence of free oxygen, will take oxygen from nitrate, reducing it to N₂ or N₂O. Pilot systems have been developed (McCarty *et al.* 1969) for the denitrification of both river water and waste water. With river water a simple carbon source such as methanol or acetic acid must be added. The process is temperature-sensitive and the rate of denitrification doubles for every 10 °C rise in temperature, within limits.

The Water Research Centre and the Thames Water Authority have investigated the use of both fixed beds and fluidized systems incorporating methanol feeds for river water treatment. Fluidized systems operating at upflow rates of 15–25 m h⁻¹ appear to be the most promising in view of their efficiency and limited clogging potential. The use of methanol could present problems in public water supplies, and the Department of the Environment have imposed tentative limits to methanol of 1 mg l⁻¹ in denitrified water and 0.25 mg l⁻¹ in final treated water. Consequently other carbon sources have been examined. Acetic acid appears to be the most promising, but initial work indicates possible problems with high nitrite formations in early bed life.

The Anglian Water Authority, with Water Research Centre support, is developing the first full-scale (2.3 × 10³ m³ per day) biological denitrification plant for the treatment of riverwater on an existing water supply scheme. The carbon source is methanol and a major aspect of the work will therefore be the development of adequate process control monitoring and fail-safe facilities. The capital cost of the scheme is approximately £400 000 (1980 prices).

(d) *The provision of low-nitrate bottled water*

This option would only supply low-nitrate bottled water to consumers with young infants being fed with reconstituted milk. The post-1976 drought nitrate situation prompted the Anglian Water Authority, the Thames Water Authority and the Essex Water Company to purchase bottling machines of the design used in the milk industry. Considerable care needs to be exercised in the bottling process to prevent bacteriological contamination of the bottled

water which could have serious consequences to public health. The costs of providing low-nitrate bottled water will vary widely from urban to rural communities. Given an efficient production and distribution process the costs in an urban area should equal those of domestic milk supplies. In contrast, in 1977 the Anglian Water Authority estimated costs of £0.262 per litre for approximately 1 l delivery by the Authority to a rural community with 250 infants for a 6 month period. Non-profit deliveries by dairies (based on their goodwill) four times per week were estimated to be £0.118 per litre for the same community.

(e) *Cost of remedial action*

The Royal Commission of Environmental Pollution (1979), as part of their study of agricultural pollution, obtained from the Ministry of Agriculture, Fisheries, and Food an estimate of the cost of crop losses from restrictions of nitrogenous fertilizer usage in areas of East Anglia where problems exist with groundwater supplies. Assuming that fertilizer usage was halved, the net loss was estimated at about £17M per year. Anglian Water Authority estimates of costs in 1977 for removing nitrates from the affected water supplies to comply with W.H.O. standards ranged from £10–15M with annual running costs of the order of £3–5M. The Royal Commission concluded that ‘the evidence suggests that if a reduction of nitrate levels in water were deemed necessary, the costs of achieving this through nitrate removed by the water authority would be less than the costs to the nation of severe restrictions on agriculture. In addition, it is known that such action by a water authority will work whereas the results of a reduction of fertilizer usage are exceedingly difficult to predict.’

Capital and running costs are very difficult to estimate and will depend on the cost of applying the nitrate removal process plant to the existing individual works, whether additional land or buildings are required in addition to the influence of manning, and maintenance policies on issues such as plant automation.

The Water Research Centre has estimated (Miller 1981) a capital cost to the water industry of about £20M (at 1980 prices) to provide plant to ensure that nitrate levels in U.K. water supplies are maintained at less than $11.3 \text{ mg NO}_3\text{-N l}^{-1}$ at the present time. As nitrate levels increase, particularly in groundwater supplies, this projected expenditure could increase substantially.

The Anglian Water Authority has some 40 sources with a nitrate problem and is currently reviewing regional estimates for the installation of treatment plant. It appears that a major reorganization of water supply resources over the next decade would be required at an estimated cost of up to £100M if W.H.O. and E.E.C. standards are to be achieved.

Extrapolation of this information in an attempt to estimate future costs on a national basis is difficult owing to the uncertainty about which sources will require attention, the individual nature of sources, and regional differences. Nevertheless an estimated cost to the water industry over the next 20–30 years could be up to £200M.

6. CONCLUDING REMARKS

1. Changes in the nitrogen cycle that have occurred over the last 40 years, particularly those resulting from agricultural activities, are having a direct impact on the water industry. The main concern is with the rising levels of nitrate in surface and groundwaters.
2. Stringent E.E.C. legislation supporting W.H.O. guidelines on nitrate in drinking water

has recently been introduced. This may have major financial implications for the water industry and, although it is not the purpose of this paper to discuss limits, it is important that these should be based on sound evidence in relation to health.

3. Agricultural activity is recognized as the principal source of nitrate in both surface water and groundwaters. Generally the highest concentrations of nitrate arise in winter drainage water from arable land and from the ploughing of long-term grassland. Animal wastes may cause pollution problems, but these are of a local nature.

4. Sewage effluent may make a significant contribution to the nitrate content of some rivers that are used for water supply, but generally the loading is small in relation to that derived from agricultural land run-off. There is no evidence to suggest that the major rise in nitrate concentrations that has occurred in surface water over the last 10–20 years has resulted from sewage effluent.

5. Although the process governing the release of nitrate from different land use situations is not fully understood nor fully quantifiable, the water industry has had to develop models to predict future nitrogen concentrations. These indicate that many sources that are currently just below the statutory limits could rise and require remedial treatment or blending. Such predictions give the water industry cause for concern.

6. The cost to the water industry of dealing with the present problem so as to protect water supplies in accordance with the existing quality criteria has been estimated at £20M for plant. Subsequent, more detailed, appraisals for eastern England, taking account of land purchase, building, effluent disposal costs, etc., show that a major change in the resource development and supply system may be required at a cost of up to £100M. It is difficult to estimate costs on a national basis owing to the many unknowns and variables involved, but a simple calculation suggests a cost to the industry of up to £200M over the next 20–30 years.

7. The water industry has devoted considerable effort to identifying the nature of the nitrate problem and to proposing solutions. The research work and operational studies being undertaken by the research organizations, the water authorities and the water companies are well coordinated by the Department of the Environment and the National Water Council, and strong links have been established with the agricultural industry.

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Discussion

G. D. HOWELLS (*Central Electricity Research Laboratories, Leatherhead, U.K.*). I should like to take up the questions of N sources that were listed in this paper. A notable missing component is that attributable to dry deposition, a significant contribution to the input of S, N, and possibly Cl to the overall budget of these elements, at least in upland catchments. A recent analysis of S budgets, for instance, has demonstrated that the dry deposit of S from atmospheres in the range 5–10 $\mu\text{g SO}_2 \text{ m}^{-3}$ (about a third of the annual mean rural concentration of the U.K.); and deposition velocity in the range 0.5–1.0 cm s^{-1} , would be the same as that contributed by 1000 mm of rain at pH 4.3–4.6. Although some studies include the contribution of ‘bulk deposition’ (i.e. mixed dry and wet), this will still underestimate the dry deposit.

While it is possible on present knowledge to make an estimate for S in dry deposit, N is much more difficult, partly because of a lack of good figures for atmospheric nitrogen oxide concentrations and deposition velocities. It is also difficult to derive from budgets because N becomes so quickly and intimately involved in biological processes. None the less it cannot be ignored and field data for N budgets in catchments suggests that it may indeed be significant.

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W. B. WILKINSON AND L. A. GREENE. Dr Howells is incorrect in suggesting that dry deposition is a missing component from the N sources presented in the paper. The concentration records for rainfall to date have included dust, bird droppings, insect fragments and other particulate matter that has settled on the collection surface of the rain-gauge and has been swept into the vessel during a rainfall event. However, more work would be helpful in distinguishing between the N in rainfall and dry deposition, and the World Meteorological Office has recommended the use of gauges that are open only during rainfall.

J. C. RODDA (*Department of the Environment, Water Data Unit, Reading, U.K.*). Dr Wilkinson and Mr Greene discussed several aspects of the work of water authorities with regard to nitrates: I wish to raise another. This is an estimate of the total nitrate load being delivered by the rivers to the seas surrounding Great Britain.

Since 1974, the Department of the Environment water authorities and river purification boards have been undertaking a regular water-quality sampling programme based on some 250 sites on rivers in England, Wales and Scotland. This programme is known as the Harmonized Monitoring Scheme. Between 30 and 40 different chemical analyses are made on every sample taken at each site, the chemical methods being harmonized by the Water Research Centre. Data are stored on computer at the Water Data Unit where there are now approximately 10^6 chemical determinations, among them nitrate concentrations (mg N l^{-1}). The archive also contains the flow records for each harmonized monitoring site.

From this archive, instantaneous flows and nitrate concentrations were combined as loads for each site. Then, by using the 5 years of data, annual loads were calculated and from them mean loads for individual rivers were aggregated to give the results shown below in table D1.

TABLE D1. NITRATE LOAD (KILOTONNES)

year...	1975	1976	1977	1978	1979	mean
England	90	79	183	147	199	146
Wales	10	18	23	19	17	19
Scotland	13	32	43	38	40	37
Great Britain	113	129	249	204	256	202

Because only about 60% of the area of England and Wales is drained by rivers in the harmonized scheme and some 50% of Scotland, it could be argued that these figures should be increased proportionately. However, it may be more interesting scientifically to note how year-to-year variations in climate influence the delivery rate of nitrate and to consider how this load relates to the application of artificial fertilizers, in terms of both quantity and cost.

W. B. WILKINSON AND L. A. GREENE. This study is of major significance in that it quantifies $\text{NO}_3\text{-N}$ losses to major rivers in Great Britain. The important feature is the magnitude of the loss and the allied annual cost implications for the agricultural industry. Research aimed at developing land-use techniques to reduce N losses of this magnitude could have dual benefit for the United Kingdom water supply authorities and the agricultural industry.