PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY B BIOLOGICAL SCIENCES

# Modelling Nitrate from Agriculture into Public Water Supplies [and Discussion]

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# Modelling nitrate from agriculture into public water supplies

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## SUMMARY

A wide range of models and techniques are briefly reviewed within the context of the Thames nitrates study in which models were developed to assess impacts of agricultural practices on nitrate levels in the river system. Here a semi-distributed approach was adopted in which a series of component models was developed to simulate hydrological and chemical behaviour of the Thames River basin. These components included:

(*a*) a daily hydrological model for the Thames basin, which included 17 tributary sub-catchments and several major aquifer systems. The model provided input flows such as tributaries, groundwater, surface runoff, effluent returns as well as abstraction flows;

(*b*) a soil zone and aquifer model for calculating the nitrate concentrations of surface runoff and groundwater given a particular land-use and fertilizer application rate;

(*c*) An integrated model of flow and water quality for the main river, which provided a mass balance along 22 reaches of the main river, allowed for denitrification processes and incorporated all inputs from the non-point sources derived by (*a*) and (*b*) above.

Model results will be presented together with an assessment of the major problems of nitrate modelling and predictions, which occur within the hydrologically active soil zone.

## INTRODUCTION

Although, in general, river water quality in the U.K. has been improving in recent years, in certain rivers the situation with regard to nitrate has deteriorated. In the Anglian Water Authority, for example, concern over increasing nitrate concentrations has led to the installation of a denitrification plant to treat water above the EEC standard of 10 mg $l^{-1}$ nitrate-N. Green (1978) reported that a significant upward trend in nitrate concentrations was evident in many groundwater and surface-water abstractions used for public-supply purposes in the Anglian region; current information appears to indicate that these increases may be primarily associated with the increasing intensity of and/or improvements in arable farming in the region in the past 20 years. Approximately 50% (by volume) of the Anglian Water Authority's abstractions have consistently exceeded 10 mg $l^{-1}$ nitrate-N with a small number of groundwater sources in strategic locations approaching 20 mg $l^{-1}$; the majority of surface abstractions, both direct river and reservoir, have exhibited concentrations in excess of 10 mg $l^{-1}$ nitrate-N for transient periods (days to months).

In the Thames River system similar problems have arisen. For example, the mean annual nitrate concentration rose from 4.2 ml $^{-1}$ nitrate-N in 1968 to 7.7 mg $l^{-1}$ nitrate-N in 1979 at Walton, the intake for the lower Thames reservoirs serving London. In the winter of 1973–74 the concentration was above the EEC limit for two weeks and in 1976–77 the limit was exceeded for four weeks.

A range of modelling techniques have been developed to simulate and predict the movement of nitrate from agricultural sources to public water supplies. These techniques are briefly reviewed and then an application to the Thames River basin is described.

## MODELLING TECHNIQUES

The modelling techniques that have been used to simulate nitrate movements fall into various categories such as time series analysis, lumped conceptual models and distributed or semi-distributed approaches.

### (*a*) *Time series approaches*

Time series techniques are suitable where the overall input–output behaviour is of prime importance and where internal mechanisms are particularly complex. It is assumed that a law of large systems supplies whereby the combinations of all the complex nonlinear and distributed elements gives rise to an aggregated system behaviour that is relatively simple in dynamic terms. In terms of modelling nitrate behaviour time series techniques can be used to relate nitrate loadings to nitrate concentrations in rivers. For example, Onstad & Blake (1980) related nitrate availability data for the River Thames basin to the river quality data time series and developed a relation of the form:

$$N_t = a_1 N_{t-1} + b_1 U_t + b_2 U_{t-7}. \qquad (1)$$

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Here $N_t$, the mean annual average nitrate concentrations in the Thames in year $t$ are related to the previous year's concentrations $N_{t-1}$ and the current year's nitrogen loading $U_t$. In this study an additional term $U_{t-7}$ was required to allow for the delayed affect of groundwater inflows. Although relatively simple, this model could be used to predict the effects of agricultural changes on future river water nitrate concentrations. Figure 1*a* shows the land use change in the Thames basin and Onstad & Blake (1980) derived a time series of available nitrate per unit area as shown in figure 1*b*. A particularly striking change in nitrogen availability from the Thames catchment appear to have occurred in the early 1940s when one third of the Thames Basin was ploughed for arable farming. This gave rise to double the nitrogen concentrations in the river as shown in figure 1*c*. However, it was not until the late 1950s and 1960s that increased use of agricultural fertilizers had a major impact and increased nitrate concentrations in runoff and rivers to current high values.

### (*b*) MAGIC – *A lumped conceptual model*

The MAGIC (model of acidification of groundwaters in catchments) model has been developed to study the acidification processes and chemical trends in catchments. Extensive details of the background theory and equations used in MAGIC have been by Cosby *et al.* (1985*a*). However the dominant processes incorporated include:

(i) anion retention by catchment soils (e.g. sulphate adsorption);

(ii) adsorption and exchange of base cations and aluminium by soils;

(iii) alkalinity generation by dissociation of carbonic acid (at high carbon dioxide partial pressures in the soil) with subsequent exchange of hydrogen ions for base cations;

(v) control of $Al^{3+}$ concentrations by an assumed equilibrium with a solid phase of $Al(OH)_3$.

MAGIC simulates these processes by using:

(i) a set of equilibrium equations that quantitatively describe the catchment input–output relations for base cations and strong acid anions in precipitation and stream water;

(ii) a set of mass balance equations that quantitatively describe the catchment input–output relations for base cations and strong acid anions in precipitation and stream water;

(iii) a set of definitions that relate the variables in the equilibrium equations to the variables in the mass balance equations.

It provides a tool by which soil processes and simplified hydrological processes can be quantitatively linked to examine chemical changes over time scales of years to decades. The model was originally developed and tested for catchments in the Shenandoah National Park, U.S.A. (Cosby *et al.* 1985*a*, *b*), but it has now been adapted for catchments in Scotland (Loch Dee, Whitehead *et al.* (1987)) and Wales (Plynlimon, Llyn Brianne, Whitehead *et al.* (1988*a*, *b*). Regional studies, using MAGIC have also been undertaken in Scotland, Wales and Norway. Although originally developed for acidification studies the model contains all the principal chemical processes controlling anion and cation behaviour. Most importantly, it can be used to investigate the effects of land use change (such as afforestation) on water quality. However, the model is a lumped or aggregated representation of a catchment and is limited, at present, to simulating up to two soil types in catchments, arranged vertically or horizontally.

MAGIC is particularly useful in that it can account for the dry and wet deposition of nitrates which can be very significant in the southeast and central regions of the U.K. Thus MAGIC could be used to assess jointly the nitrate availability from soils and from atmospheric sources.

### (*c*) *Lumped conceptual soil zone models*

Another approach to modelling nitrate in the soil zone is that developed by Addiscott & Whitmore (1987) in which the key processes controlling nitrogen transformation in the soil zone have been modelled. Processes included are nitrate leaching, mineralization, nitrification and N uptake by crops (see Greenwood 1982). Inputs and outputs are incorporated such that model simulations can be obtained for a wide range of weather regimes, application rates, soil types and crop types. The model has been validated against extensive field data.

### (*d*) *Distributed runoff models*

Another class of models have been developed to simulate the complex behaviour of catchment hydrology and hence chemistry. These include the Institute of Hydrology Distributed Model (IHPM) and Systeme Hydrologique European (SHE) developed jointly between the Institute of Hydrology the Danish Hydraulics Institute and SOGREATH of France. Both IHDM and SHE are physically based models that solve the equations governing flow in surface or near surface systems. The models include information on topography, channel network geometry and soils and were designed initially to investigate the effects of land use change and for application to ungauged catchments. They are fully distributed and can represent processes at the field scale and also at large catchment scale. Extensions to the models to predict nitrates are currently being investigated.

### (*e*) *Distributed groundwater models*

There are many groundwater models available for simulating flow but relatively few that have been established, calibrated and validated for jointly simulating flow and nitrate. Kaluarachchi & Parker (1988) describe a two-dimensional finite element model for predicting nitrogen species transformation and transport in unsaturated soil and groundwater. Nitrification, denitrification, mineralization and immobilization are treated as first order kinetic processes as is plant uptake of ammonium and nitrate. Simultaneous transport of the solution phase is described by the convection diffusion equation.

An alternative approach developed by Oakes (1981) considers the movement of water and nitrates from soils in groundwater and then into public water supply boreholes. The model consists of a flow module for estimating water movements from surface into groundwaters and a nitrate module for estimate nitrate leaching from soils into subsurface water.

The quantity of nitrate leached from the soil into the underlying aquifer is calculated on an annual basis over the entire catchment area. It is also necessary to calculate leaching losses back to 1950 or even earlier in some cases because of the long time delays in some aquifers. For example, if it takes 40 years for water and nitrate to move from the land surface into water supplies, then the leaching rate 40 years ago is needed to be able to calculate groundwater nitrate concentrations today. In the model leaching rate has been related to land use and fertilizer applications. The relations between land use and leaching were derived from the results of a field measurement programme undertaken in the past 10 years. About 100 boreholes have been drilled by the Water Research Centre (WRC), the British Geological Survey (BGS) and the water authorities in different aquifers and beneath a wide variety of land use types. By extracting water from the rock borehole samples recovered, and measuring the chemicals in the water from different depths it was possible to calculate directly the amount of nitrate leached. By relating the amounts of nitrate leached to present and antecedent land use and fertilizer applications it was possible to postulate some relations between land use and nitrate leaching. It was found that for arable land the leaching rate was equivalent to between 20% and 50% of the fertilizer application rate, and for grassland between 10% and 15%. Ploughing of grassland results in a leaching loss of between 100 and 300 kgN $ha^{-1}$†. These findings, together with results from other research organizations have been incorporated in the WRC model to calculate leaching losses.

The WRC model has been criticized because of the empirical nature of these leaching relations and indeed there is no attempt to include a detailed knowledge of process interactions within the model. On the other hand it has been applied to over 20 catchments in chalk, sandstone and limestone areas with some success in simulating the historic nitrate trends in public supply boreholes (Oakes 1981).

### (*f*) *River quality models*

In the U.K. some 70% of water to public supply is obtained from direct river abstraction. Within rivers, there are major transformation of nitrate which determine the nitrogen balance. Nitrogen transformations in rivers are complex as shown in figure 2. The principal processes involve the nitrification of ammonia to nitrite and hence to nitrate under aerobic conditions and the reduction of nitrate to nitrogen or ammonia under unaerobic conditions. Plants and algae also assimilate nitrate from the water column during growth periods and this nitrogen is recycled eventually during the decay of cellular organic matter.

Most modelling studies have been restricted to the nitrification process since the discharge of ammonia into rivers from industrial or domestic effluents has a significant effect on fisheries, water supply and, as shown in the following equations, dissolved oxygen levels.

$$NH_4^+ + (\tfrac{3}{2})O_2 \xrightarrow{\text{Nitrosomonas}} NO_2^- + 2H^+H_2O, \quad (2)$$

$$NH_4^- + (\tfrac{1}{2})O_2 \xrightarrow{\text{Nitrobacter}} NO_3^-. \quad (3)$$

*Nitrosomonas* and *Nitrobacter* are autotrophic bacteria responsible for the oxidation process. Since this process is a two-stage biochemical reaction most mathematical models that have been developed are in the form of a set of coupled differential equations such as:

$$\frac{\partial N_1}{\partial t} = -u\frac{\partial N_1}{\partial x} + D\frac{\partial^2 N_1}{\partial x^2} - k_1 N_1, \quad (4)$$

$$\frac{\partial N_2}{\partial t} = -u\frac{\partial N_2}{\partial x} + D\frac{\partial^2 N_2}{\partial x^2} + k_1 N_1 - k_2 N_2, \quad (5)$$

$$\frac{\partial N_3}{\partial t} = -u\frac{\partial N_3}{\partial x} + D\frac{\partial^2 N_3}{\partial x^2} + k_2 N_2, \quad (6)$$

where $N_1$, $N_2$ and $N_3$ represent ammonia (as $N$), nitrite (as $N$) and nitrate (as $N$) concentrations, $u$ represents velocity, $D$ is the dispersion coefficient, $x$ represents distance along the river, $t$ is time and $k_1$ and $k_2$ are reaction rates. These equations are often simplified to first-order lumped parameter models where advection forces dominate.

Surprisingly, there has been relatively little modelling research on the nitrate reduction process in rivers. The process of denitrification is represented by the following equation:

$$6NO_3^- + 5CH_3OH \rightarrow 3N_2 + 5CO_2 + 7H_2O + 6OH^-. \quad (7)$$

Biological denitrification is promoted by a large number of bacteria that contain nitrate reductases, enzymes that mediate the reaction. The nitrogen gas formed by this reaction may be transferred from the water to the atmosphere if the nitrogen concentration exceeds the saturation concentration. The reduction processes occur in the mud or at the mud–water interface and as shown in equation (7) the reaction requires an organic carbon substrate. Because of the complexity of the reaction mechanisms it is normally assumed that the reaction kinetics are first-order with the transport equation for nitrate written as:

$$\frac{\partial N_3}{\partial t} = -u\frac{\partial N_3}{\partial x} + D\frac{\partial^2 N_3}{\partial x^2} + k_2 N_2 - k_4 N_3, \quad (8)$$

where $k_4$ is the denitrification rate.

Empirical evidence for the first-order reaction kinetics is given by Toms *et al.* (1975), but with the modification that the reaction rate is a function of mud

† 1 hectare = $10^4$ $m^2$.

surface area and temperature. In addition, there is considerable spatial variation in the denitrifying capacity of sediments in lowland eutrophic rivers.

## THE THAMES NITRATES STUDY

Within large river basins nitrate is derived from many sources such as effluents, agricultural runoff, and groundwaters and all of these must be incorporated into any river basin model to be used for predictive purposes. A fully distributed modelling approach is clearly impractical given the size and complexity of the large river basin. Thus a semi-distributed approach was adopted in a major study of the Thames River basin.

In the Thames River basin there has been a major change in land use over the past forty years as shown in figure 1. Onstad & Blake (1980) estimated that the major land-use change from permanent grass top cereal crops together with significant use of nitrogen fertilizers released large loads of nitrate as shown in figure 1 *b* and these nitrate loads have been reflected in higher river nitrate concentrations as indicated in figure 1 *c*.

To evaluate future trends and management options within the Thames basin a series of component models for the river basin were developed. These included:

(i) a daily water quantity model for the thames system, which includes 17 tributary sub-catchments and several major aquifer systems. The model provides river input flows such as tributaries, groundwater, surface runoff, effluent returns as well as abstraction flows;

(ii) a soil zone and aquifer model developed by the Water Research Centre for calculating the nitrate concentrations of surface runoff and groundwater given a particular land use and fertilizer application rate;

(iii) an integrated model of flow and water quality for the main river developed by the Institute of Hydrology. The model provides a mass balance along twenty two reaches of the main river, allows for denitrification processes and incorporates all inputs from the non-point sources derived by models (i) and (ii) above. Details of the model are given by Whitehead & Williams (1984).

The dynamic water quality model for the Thames is based on a mass balance approach for a non-conservative variable and can be written for a reach as follows:

$$\frac{dn(t)}{dt} = \frac{Q_{\mathrm{i}}(t)}{V_{\mathrm{e}}}u_{\mathrm{i}}(t) - \frac{Q_{\mathrm{o}}(t)}{V_{\mathrm{e}}}n(t) + \frac{Q_{\mathrm{G}}(t)}{V_{\mathrm{e}}}n_{\mathrm{G}}(t) + \frac{Q_{\mathrm{T}}(t)}{V_{\mathrm{e}}}n_{\mathrm{T}}(t) + \frac{Q_{\mathrm{S}}(t)}{V_{\mathrm{e}}}n_{\mathrm{S}}(t) + \frac{Q_{\mathrm{E}}(t)}{V_{\mathrm{e}}}n_{\mathrm{E}}(t) + \frac{Q_{\mathrm{A}}(t)}{V_{\mathrm{e}}}n_{\mathrm{A}}(t) - \frac{K}{d}(10^{0.02930})\,n(t), \quad (9)$$

where $n(t)$ is the output nitrate concentration, $u_1(t)$ is the input nitrate concentration, $Q_{\mathrm{i}}(t)$ and $Q_{\mathrm{o}}(t)$ are upstream and downstream flow rates, $K$ is the denitrification rate, $d$ is river depth, and the subscripts G, S, T, E and A refer to groundwater inflows, surface runoff, tributaries, effluents and abstractions respectively. Inputs from groundwater, daily surface runoffs and tributaries are defined by the WRC soil zone and groundwater models $V_{\mathrm{e}}$ represents the effective volume of the cell. This is to allow for short circuiting effects in the river and the presence of deal zones. Typical simulations of flow and nitrate in the Thames are shown in figures 3 and 4.

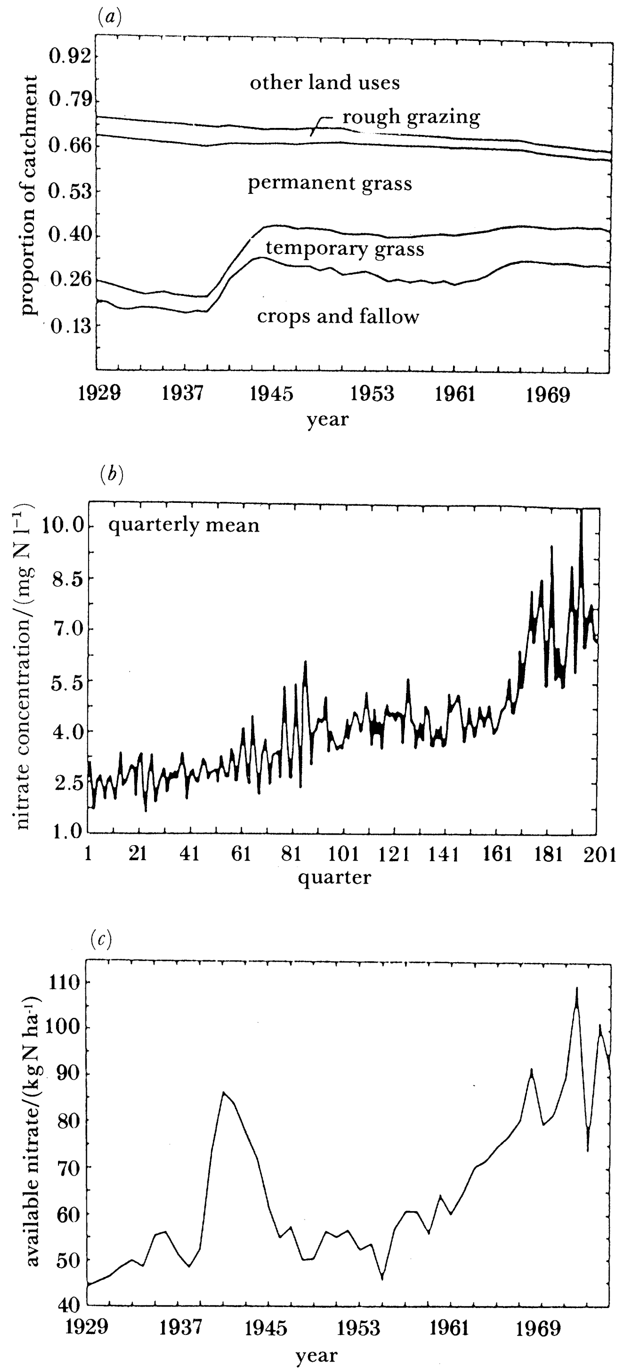


Figure 1. (*a*) Land use change in the Thames River Basin since 1929. (*b*) Nitrate availability in the Thames River basin. (*c*) Nitrate concentrations in the River Thames at Walton since 1929.

The integrated model has been used in a collaborative study with Thames Water Authority to investigate future nitrate concentrations in the Thames and has been run using 56 years of hydrological data to reproduce river flows and nitrate concentrations at



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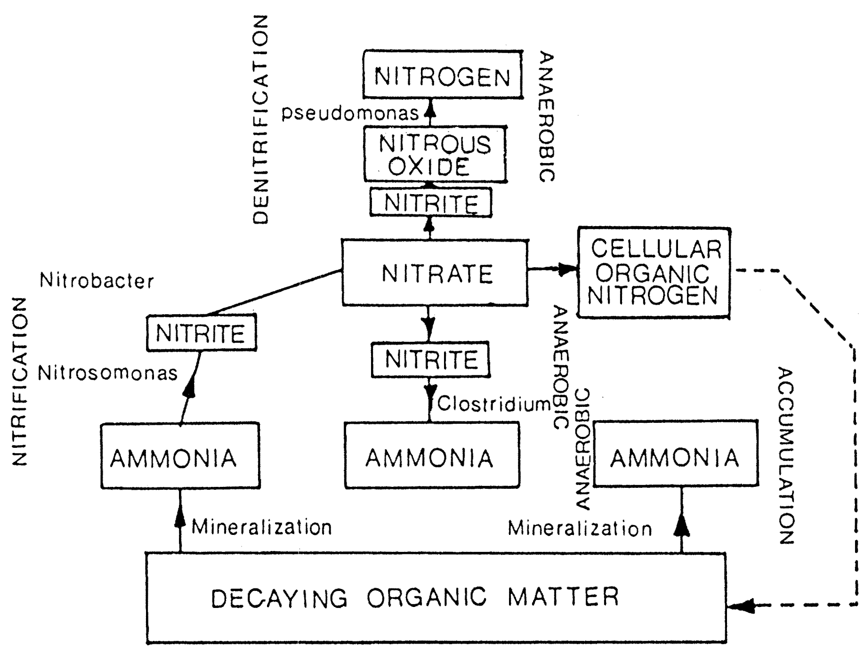


Figure 2. Transformations of Nitrogen in River systems.

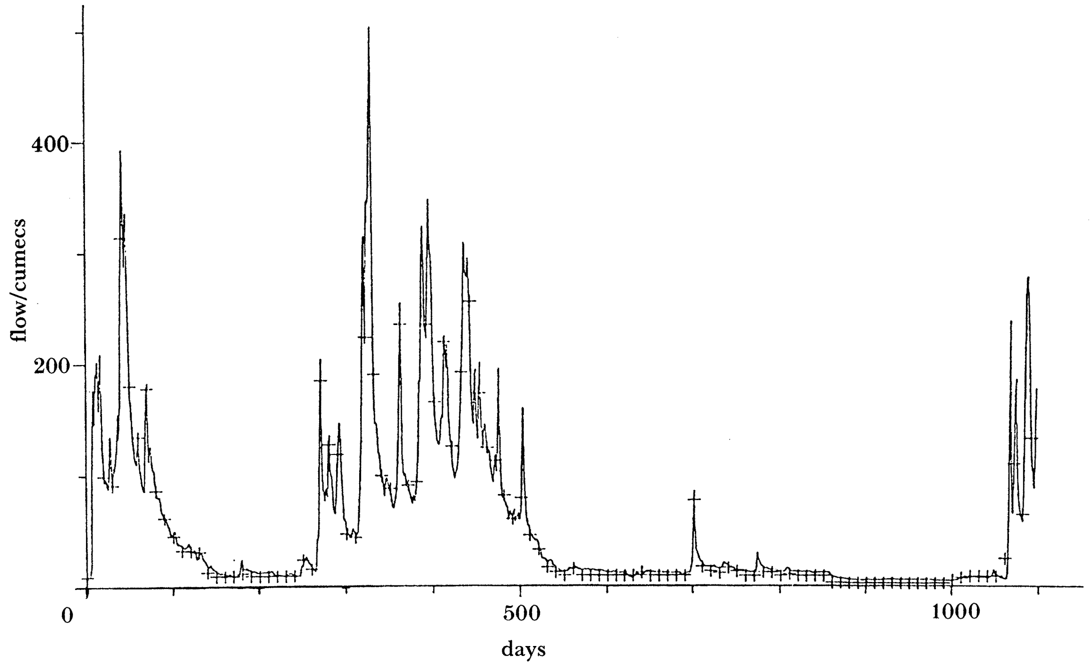


Figure 3. Simulated (–) and observed (+) daily flow in the Thames at Swinford during 1974, 1975 and 1976.

Farmoor and Datchet. These abstractions sites provide drinking water for Oxford and London, respectively. The model was run with two sets of conditions:

(i) 1982 agricultural land-use, fertilizer application rates, population levels and water demands;

(ii) 2006 agricultural land-use, fertilizer applications rates, population levels and water demands.

In 1982 conditions were based on published land-use information and fertilizer surveys, together with inputs from sewage treatment plants based on historical population and average consumption data. Forecasts for the 2006 conditions were made from trends in the historical data and estimates of population growth.

The results from the model run under the two sets of conditions are summarized in tables 1 and 2, which show the number of years the WHO limit of 11.3 mg N $^{-1}$ would be exceeded at the two abstractions sites considered. It is clear that at both abstraction sites there will be a marked increase in the number of years that 11.3 mg $l^{-1}$ would be exceeded if nothing were to be done to control the nitrate problem.

The model also provides information on the relative effects of diffuse pollution. During autumn storm and winter high flow conditions the nitrate diffuse sources such as agricultural runoff completely dominated the point source pollution. The situation is altered during

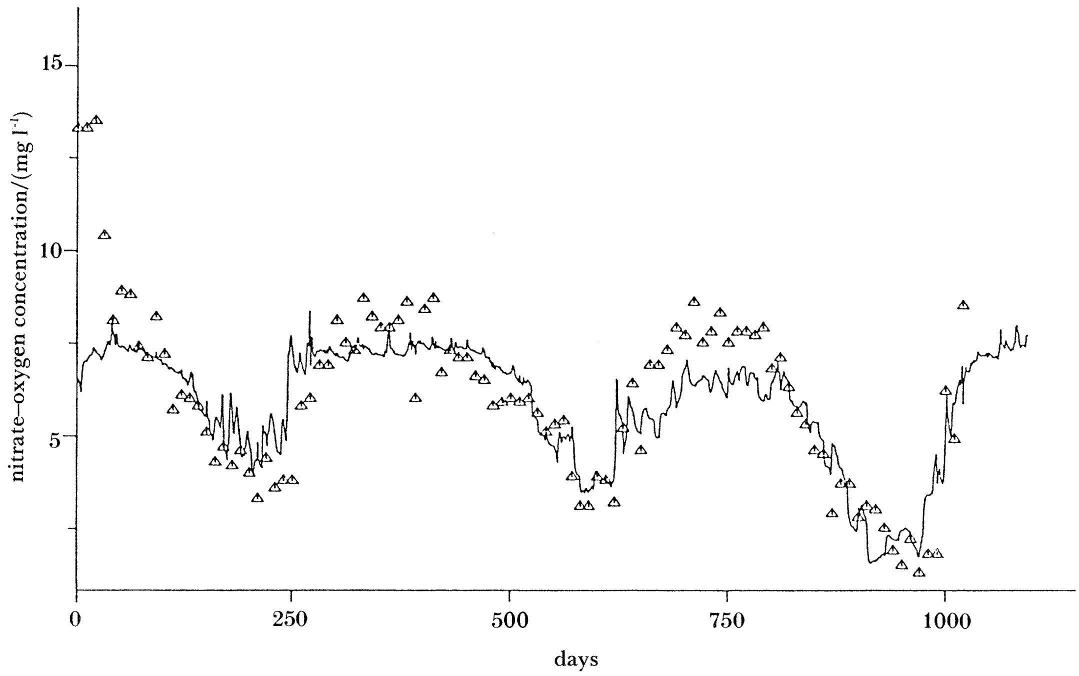


Figure 4. Simulated (—) and observed (△) nitrate concentrations in the Thames at Swinford during 1974, 1975 and 1976.

Table 1. *Oxford intake on River Thames*

| duration of exceedance | number of years 11.3 mg N $l^{-1}$ exceeded (October–September) 1982 land-use, population and damand | 2006 land-use, population and demand |
|---|---|---|
| 1–15 days | 7 | 10 |
| 16–30 days | 3 | 6 |
| 31–45 days | 2 | 2 |
| 46–60 days | 0 | 1 |
| 61–75 days | 0 | 3 |
| 76–90 days | 0 | 0 |
| > 90 days | 0 | 0 |
| as annual average | 0 | 0 |
| total numbr of years in which exceedance is predicted over a 56-year period | 12 | 22 |

low flow conditions when point sources are relatively more significant.

## DISCUSSION AND CONCLUSIONS

To a large extent the future modelling of nitrate in catchments depends on the types of action and policy being contemplated by government to control nitrate pollution and the accuracy with which predictions are required. For example, the Thames semi-distributed model gave adequate information on likely trends in nitrate given broad changes in agricultural strategy. Doubtless the model could be generalized and applied to other major basins to simulate large-scale effects such as changes in land use across the region or regional changes in fertilizer application rates. However, if more detailed information is required on nitrate levels in a particular part of the catchment to protect a surface on sub-surface supply a more sophisticated model, distributed flow model such as IHDM linked to a nitrate process model might be required. Certainly such an approach would be required if individual field land use is to be changed or nitrate fertilization is to be reduced on a small local scale to protect a particular supply.

Another area of concern is the conversion of land use from cereals to broadleaf afforestation. The additional inputs from fertilization of these trees or inputs from atmospheric sources of nitrates need to be considered and here a detailed chemical model such as MAGIC

Table 2. *London intake on River Thames*

| duration of exceedance | number of years 11.3 mg N $l^{-1}$ exceeded (October–September) 1982 land-use, population and damand | 2006 land-use, population and demand |
|---|---|---|
| 1–15 days | 5 | 8 |
| 16–30 days | 2 | 9 |
| 31–45 days | 0 | 2 |
| 46–60 days | 0 | 1 |
| 61–75 days | 0 | 0 |
| 76–90 days | 0 | 0 |
| > 90 days | 0 | 0 |
| as annual average | 0 | 0 |
| total number of years in which exceedance is predicted over a 56-year period | 7 | 20 |

would be appropriate to predict surface water quality. MAGIC could be extended to handle spatially variability and linked to the distributed quantity models.

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### *Discussion*

P. A. Costigan (*ICI Fertilizers*, *Jealott's Hill Research Station*, *Bracknell*, *Berkshire*, *U.K.*), Dr. Jenkinson's paper showed that organic matter turnover in soils is a very slow process. Consequently the large areas of grassland that were ploughed and converted to arable land during World War II are unlikely to have reached a new equilibrium organic matter content even after 50 years. The gradual oxidation of this organic matter may still be making a significant contribution to nitrate leaching. Is this contribution included in Dr Whitehead's model?

P. G. Whitehead. No, this organic matter is not included at present. However, from the Thames nitrates data and modelling study, the evidence is that the major increases occurred up to seven years after ploughing. Thereafter the effects were minimal.

E. M. Bridges. (*University College of Swansea*, *Wales*, *U.K.*) The role of nitrates figured largely in Dr Whitehead's presentation but these are only one form of pollution. Could Dr Whitehead tells us what other contaminants are monitored by hydrologists.

P. G. Whitehead. Other contaminants include pesticides, heavy metals, anions, cations and effluent discharges which cause particular problems for oxygen balances in lowland river systems.

D. S. Powlson. (*Rothamsted Experimental Station*, *Harpenden*, *Herts*, *U.K.*). Can Dr Whitehead's model deal with the effects on nitrate leading of changes in agricultural practice such as straw incorporation or the growth of winter cover crops? These effects may be very significant.

Does Dr Whitehead think denitrification within ground water aquifed is of great importance?

P. G. Whitehead. At present, the model cannot cope with such changes in agricultural practice. A more detailed process model is required and such a model is under development at the Institute of Hydrology in collaboration with Dr. T. Addiscot at Rothamsted.

British Geological Survey have some evidence that denitrification does occur in groundwater, although the effect may be fairly limited.

M. J. Goss (*The Macaulay Land Use Research Institute, Craigiebuckler, Aberdeen, U.K.*). Dr Whitehead's model predicts greater nitrate concentration in the river Thames water by the year 2000 but what are the separate contributions to this increase from changes in agricultural land use within the catchment and from changing needs for water by the public?

P. G. Whitehead. Both effects are included in the model. The dominant effect is agricultural land use during winter for public use can significantly affect stream quality under low flow conditions.

K. Goulding. (*Soils and Agronomy Department, IACR Rothamsted Experimental Station, Harpenden, Herts, U.K.*). Dr Whitehead stated that, in the River Thames at Walton over the period from 1920 to 1980, there was a good correlation of nitrate concentration with the amount of nitrogen fertilizer applied to land in the Thames catchment. Nitrates can enter rivers from sources other than agriculture, notably sewage effluent, and the population of the Thames catchment will have increased greatly over the period of observation. Did Dr Whitehead attempt to relate nitrate concentration with effluent release or the population of the catchment?

P. G. Whitehead. Yes, the relative effects of agricultural runoff sewage effluent were assessed, particularly in the River Lee. Under high flow conditions nitrogen concentrations in the rivers dominated by agricultural runoff. Similar studies in the Bedford Ouse support this conclusion (see Whitehead *et al.* 1981).

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