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Nitrate leaching to groundwater

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Groundwater provides over 30% of developed supplies of potable water in Britain. The outcrops of the important aquifers form extensive tracts of agricultural land. Groundwater resources largely originate as rainfall that infiltrates this land. During the 1970s, growing concern about rising, or elevated, groundwater nitrate concentrations, in relation to current drinking water standards, stimulated a major national research effort on the extent of diffuse pollution resulting from agricultural land-use practices.

The results presented derive from intensive and continuing studies of a number of small groundwater catchments in eastern England. It is in this predominantly arable region that the groundwater nitrate problem is most widespread and severe. The distribution of nitrate in the unsaturated and saturated zones of the aquifers concerned is summarized. These data have important implications for the water-supply industry, but their interpretation is discussed primarily in relation to what can be deduced about both the recent and long-term histories of leaching from the more permeable agricultural soils.

INTRODUCTION

Occurrence and significance of groundwater resources

In Britain, groundwater abstraction, predominantly for potable water-supply, averages about 7000 Ml d^{-1}, of which some 50% is from the Cretaceous Chalk, 35% from the Triassic Sandstone and the remainder from smaller aquifers, notably the Jurassic Limestone and the Cretaceous Lower Greensand. The main aquifer outcrops (figure 1) form extensive areas of valuable farmland in eastern, central and southern England. Groundwater resources, which largely originate in these areas as infiltrating excess rainfall are, and always have been, directly vulnerable to diffuse pollution by agricultural practices.

Over much of the aquifer outcrop areas (the unconfined aquifers), the groundwater table, even at its highest seasonal level, is below 10 m depth and quite widely exceeds 20 m depth. Rainfall infiltrating below the depth of influence of plant roots will pass vertically downward through the aerated (or unsaturated) zone before recharging the main groundwater body (or saturated zone). Such recharge then becomes incorporated in the lateral flow of the regional groundwater system and, if not abstracted for water-supply, will eventually discharge to form the baseflow of streams and rivers. In eastern and central England the major aquifers are extensively covered by a variable suite of Pleistocene glacial (drift) deposits (figure 1), which to varying degrees confine groundwater, reduce recharge rates from infiltrating rainfall and thus form the semi-confined aquifers.

Although a fraction of the groundwater and dissolved salts in natural circulation may be discharged in the same year that they originated as infiltration from agricultural soils, it must be stressed that groundwater residence times for British aquifers will more often exceed 10, or even 20, years (Foster & Crease 1974; Foster 1976). At considerable depth below the groundwater table and in the deeper confined aquifers, circulation may be exceedingly slow and, in certain areas, groundwater has been dated back to at least 10 ka B.P. Thus groundwater systems are quite different from surface watercourses in that the effects of pollution incidents become apparent much more slowly in water supplies but persist much longer.

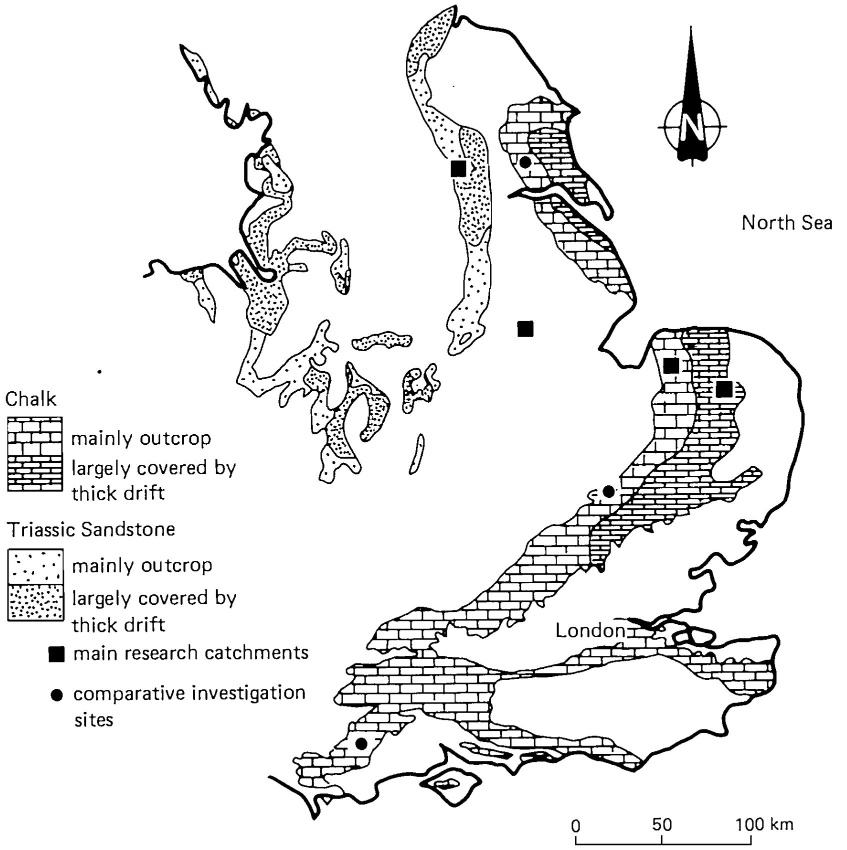


FIGURE 1. Distribution of major British aquifers.

The main British aquifers are all of the porous bedrock type, in which discontinuities (fissures) have a varying, but often overriding, influence on overall groundwater, solute and pollutant movement, but in which the pore water of the rock matrix will often play an important role in retarding the movement of solutes and pollutants.

Approach and scope of the paper

Among the substances added to, and/or generated by, agricultural soils, nitrates are most prominent, since they are very soluble, readily leached, and represent a health hazard at a relatively low concentration in water supplies.

Discussion of nitrate leaching to groundwater will be limited to that from agricultural soils, although it should be noted that nitrate may also be leached from most unsewered sanitation units, certain sewage disposal and waste-water reclamation processes and from farmyard livestock effluents during their handling, storage and disposal. Such leaching, however, is more of local than regional significance, at least in the British context.

The results presented derive exclusively from a continuing I.G.S. research project, which began in 1975. It is being executed primarily through the detailed long-term study of four small groundwater catchments (figure 1) to public water-supply boreholes experiencing a significant, and growing, nitrate problem. The research is part of a coordinated programme in which the Water Research Centre and the Regional Water Authorities are also involved. The results to date have important practical implications for the water-supply industry, which have received interim review elsewhere (Foster & Young 1979, 1981; Oakes *et al.* 1981). However, some significant aspects of their interpretation still await clarification through further long-term fieldwork.

In this paper, however, the results are discussed primarily in relation to what can be deduced about both the recent and long-term histories of nitrate leaching from agricultural soils. Reliable estimates of leaching losses from the permeable soils, which occur on most aquifer outcrops, are difficult and costly to determine by agricultural field experiments. At the same time they are likely, other factors being equal, to be greater than those from lower-permeability less-aerated soils, where denitrification losses will be more significant.

Since the aquifer outcrop areas occur mainly in the drier parts of England and their associated soils are well drained, most of the land area involved is used for arable farming. There is a corresponding bias in this paper. In eastern England the widespread ploughing-up of permanent pasture is believed to have taken place in the nineteenth century and had certainly affected more than 80% of the research catchments before 1935. Elsewhere the conversion of pasture to arable land has continued to be a marked feature of British agriculture during this century.

APPRAISAL OF THE RECENT HISTORY OF LEACHING LOSSES

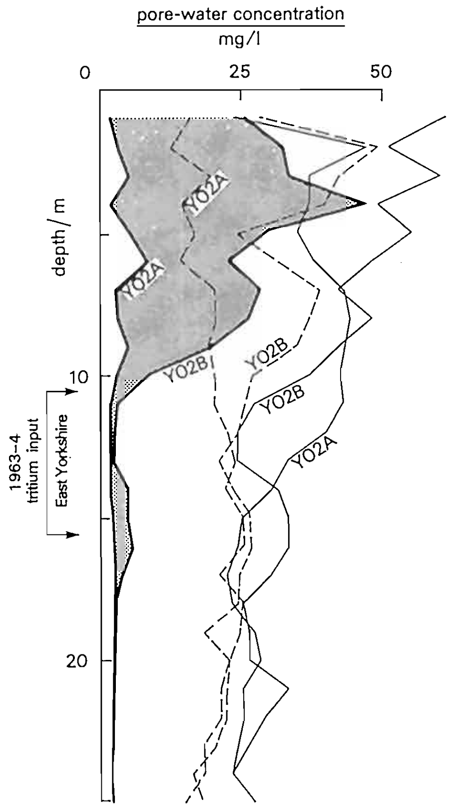
If they could be interpreted confidently, vertical profiles of the distribution of nitrate in the unsaturated zone form a potentially powerful method of evaluating the recent history of leaching losses from permeable agricultural soils subjected to different cropping régimes and fertilizer applications.

It is only feasible to sample the pore water of the unsaturated zone. The methods of sampling and analysis for nitrate (and the other frequent determinands) have been described elsewhere (Foster & Young 1979, 1981). Among unsaturated zone pore-water profiles from British aquifers, those from the Chalk offer the best possibility of detailed interpretation, because of this formation's greater overall areal uniformity in hydraulic properties on the small scale. They have been shown to be related to the history of agricultural practice on the overlying land, and are alone discussed further in this section.

Profiles from pasture land

Profiles from unfertilized and fertilized permanent grassland (e.g. figures 2 and 3) have been shown to be characterized by low and uniform nitrate concentrations, frequently less than 2 mg NO_3-N l^{-1}. They suggest that corresponding leaching losses do not generally exceed 10 kg N ha^{-1} annually, except perhaps where rates of inorganic N fertilizer application are, by current standards, abnormally high (over 250 kg N ha^{-1} annually).

Profiles from beneath a large, now partitioned, field in East Yorkshire (figure 2), show the marked effect of ploughing-in of pasture with subsequent continuous cereal cropping. The enhanced mineralization of organic N in the soil biomass consequent upon the aeration caused



Year	site YO2A	site YO2B
1978	G(0:0:0)	C(75:28:180)
1977	G(72:26:140)	C(72:26:140)
1976	G(0:0:0)	C(?)
1975	G(50:19:120)	C(75:23:150)
1974	G(65:0:0)	C(75:23:150)
1973	G(63:19:120)	C(94:28:150)
1972	G(87:0:0)	C(76:23:140)
1971	G(94:28:150)	C(94:28:150)
1970	G(65:0:0)	C(84:23:120)
1969	G(87:0:0)	C(75:28:180)
1968	G(50:19:120)	C(75:28:180)
1967	G(43:0:0)	C(20:23:120)
1966	G(0:0:0)	
1965	G(0:0:0)	
1964	G(0:0:0)	
1963	G(53:0:0)	
1962	G(40:15:100)	
1961	G(0:0:0)	
1960	G(0:0:0)	
1959	G(0:0:0)	
1958	G(0:0:0)	

FIGURE 2. Pore-water profiles for Chalk unsaturated zone in East Yorkshire showing the effects of partial conversion from pasture to arable land in autumn 1966 (G, permanent grassland; C, winter or spring cereals; (N: Cl: SO_4) ratio (kilograms per hectare) applied annually in fertilizers.)

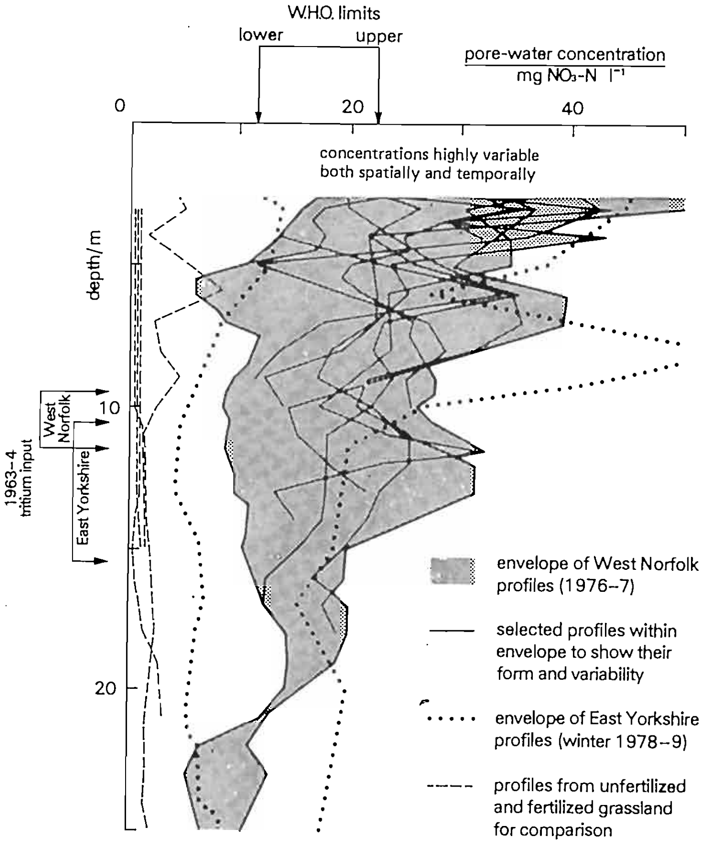


FIGURE 3. Summary of nitrate profiles for Chalk unsaturated zone beneath long-standing arable land.

by ploughing-in of established grassland is an important source of nitrate that might be lost by leaching, especially from the more permeable agricultural soils. The quantity potentially available is very large (Reinhorn & Avnimelech 1974; Smith & Young 1975).

Profiles from arable land

High nitrate concentrations have invariably been encountered in pore-water profiles from beneath arable land, even with a history of short grass leys (Foster & Young 1979, 1981). The profiles for long-standing arable fields are characterized by a major 'nitrate front', which has reached, or formed at, around 3–8 m below ground level, with concentrations decreasing to 10–20 mg NO_3-N l^{-1} below 10 m depth. When viewed in relation to the W.H.O. (and E.E.C.) recommendations for potable water (figure 3), these profiles give rise to serious concern about future Chalk groundwater quality. Within a given arable field, the pore-water nitrate profiles (and those for other determinands) show overall uniformity (figure 4), but in detail significant variations in the magnitude of peak concentrations (reflecting very localized differences in soil leaching) and in the depth to peak concentrations (reflecting minor differences in the net groundwater flux) are present. Profiles from long-standing arable fields in the same catchment, each of which will have had a somewhat different history of cropping and fertilizer application, are much more variable, but have a characteristic shape (figure 3).

Discussion of profile interpretation

Natural tritium constitutes one of the best available tools with which to evaluate the mechanisms of movement of more-soluble, non-reactive, pollutants through the unsaturated zone, because of its unique temporal distribution in rainfall (and thus infiltration) with a pronounced peak associated with thermonuclear fallout in the springs of 1963 and 1964. The preservation of this peak at relatively shallow depth in Chalk unsaturated zone pore-water profiles, with apparently little dispersion, has been widely observed (see, for example, Smith *et al.* 1970; Foster & Smith-Carington 1980), although it should be noted that profiles with much broader peak width have also been observed at some sites in eastern England. If it could be assumed that solutes and isotopes were moving uniformly downwards in a non-dispersive ('piston') flow, such that all of the pore-water nitrate in the profile above the base of this tritium peak was of post-1963 origin (figures 3 and 4), then the detailed history of recent leaching losses for the given cropping record could be deduced from the corresponding moisture content profiles and estimates of actual infiltration. The implication would be that leaching rates have risen steadily from about 1960 onwards to exceed 80 kg N ha^{-1} annually in the mid-1970s. Average annual rates of leaching since 1963 would be mainly in the range 50–70 kg N ha^{-1} (table 1).

Although direct leaching of fertilizer nitrate by late spring infiltration will occur in some years, and any unused nitrate would be leached in the following autumn, the bulk of leaching losses are probably derived from the mineralization of organic matter in the soil biomass, especially in autumn. Nevertheless, such leaching losses are large when compared with current annual fertilizer application rates on arable land (80–130 kg N ha^{-1}). If verified they would represent an enormous loss of nutrients: assuming current costs to be about £0.30 kg^{-1} N and an average annual loss of 80 kg N ha^{-1}, the annual loss would exceed £15 M, considering only the drift-free aquifer outcrop areas (figure 1) in eastern England alone, some 80% of whose land area is dedicated to arable farming. Nitrate concentrations of Chalk groundwater in much of this

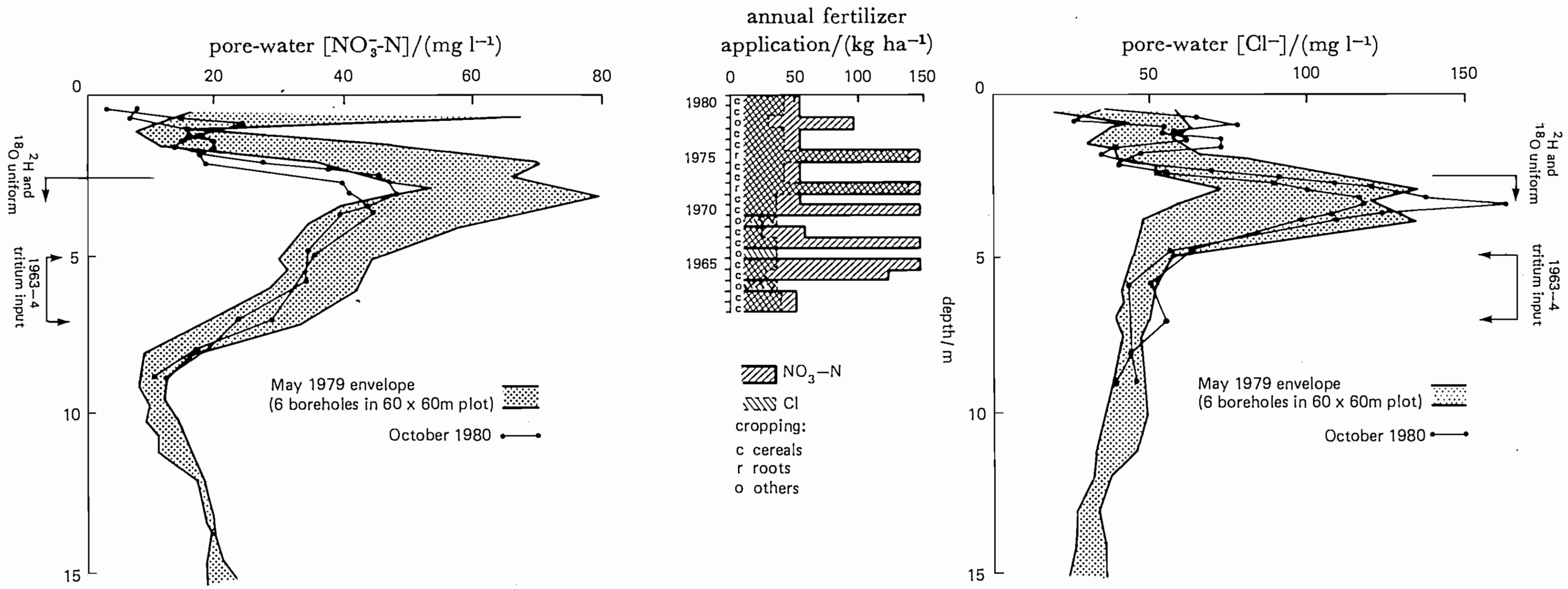


FIGURE 4. Multiple and sequential unsaturated zone pore-water profiles for an individual long-standing arable field on Cambridge Chalk.

region would, by implication, rise steadily to reach intolerable levels (in excess of 22.6 mg NO_3-N l^{-1}) from the 1990s onwards, and in other regions to somewhat lower levels at later dates (Young *et al.* 1976), necessitating major capital expenditure on alternative water-supply development or water treatment facilities, or both.

TABLE 1. ANALYSIS OF CHALK UNSATURATED ZONE PORE-WATER PROFILES (ASSUMING NON-DISPERSIVE FLOW) FOR RECENT LEACHING LOSSES FROM ARABLE LAND

	NO_3^--N/(kg ha^{-1})			Cl^-/(kg ha^{-1})		
site (sampling date)	mean annual application in fertilizer†	profile total (depth range)‡	equiv. mean annual loss	mean annual application in fertilizer†	profile total§ (depth range)‡	equiv. mean annual loss
N01X (Feb. 1976)	132	830 (1–11 m)	64	72	1180 (1–11 m)	91
N60 (Apr. 1977)	143	980 (1–11 m)	70	77	1460 (1–11 m)	104
Y08 (Oct. 1978)	125	780 (1–16 m)	52	78	1290 (1–16 m)	86
Y09–10 (Oct. 1978)	153	600 (1–16 m)	40	88	1000 (1–16 m)	67
C01–06 (May 1979)	78	980 (1–7 m)	61	50	1170 (1–7 m)	73
Y02B‖ (Oct. 1978)	75 100¶	1020 (1–11 m)	93	25	1150 (1–11 m)	104

† During period 1963 to sampling date indicated.

‡ From below main root zone to apparent depth of 1963–4 tritium penetration.

§ After deducting background corresponding to pore-water concentration of 15 mg Cl l^{-1}.

‖ Figures for Y02B relate to period of continuous arable cultivation since 1966 when pasture ploughed in.

¶ Allowing 300 kg N ha^{-1} release on ploughing of permanent grassland.

The possibility of substantial nitrate attenuation through denitrification within the unsaturated zone, beneath arable land at least, seems rather unlikely (Foster & Young 1979, 1981). Biochemical denitrification cannot yet be discounted, however, since significant (insoluble) organic carbon concentrations and, occasionally, nitrate-reducing bacteria populations have now been measured at a considerable depth below the soil interface at a few sites (Whitelaw & Rees 1980; Hall 1981).

In view of the scale of its implications, the profile interpretation above warrants a detailed re-examination. Two difficulties are immediately apparent.

First, in eastern England arable farming practices underwent their most important post-war change, to more continuous cereal cropping sustained by much higher N fertilizer applications, during the period 1955–65, and have been relatively constant since then. The 'implied date' of major increases in the pore-water nitrate profiles is from the late 1960s. Thus a considerable time-lag in the response of soil biomass mineralization to increased inputs of organic N from crop residues would have to be invoked. Bearing in mind the complexity of the soil system this may be feasible. At some sites in southern England with rather different cropping histories, however, nitrate profiles have been simulated by using surprisingly simple 'release rules' (Oakes *et al.* 1981), based on the observation that an equivalent of only 50 % of applied fertilizer N is taken up by the crop (Kolenbrander 1977) and the assumption that, for permeable arable soils, an amount equivalent to all the unused fertilizer N will be released and leached from the soil, by one process or another, during the subsequent 3 years.

Secondly, in the analysis of the pore-water profiles for a non-nutrient, non-reactive, ion such as chloride, it is only possible to approach a satisfactory mass balance for the post-1963 period (table 1) by assuming rather extreme values for the chloride content of rainfall and of the various brands of soil fertilizers and additives.

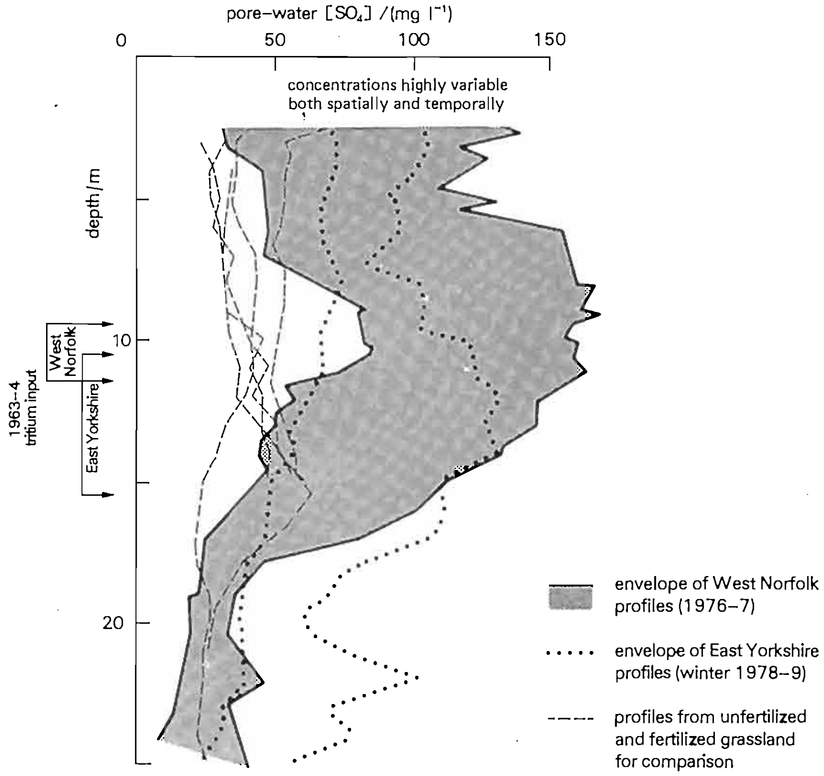


FIGURE 5. Summary of sulphate profiles for Chalk unsaturated zone beneath long-standing arable land.

Similar can be said of sulphate, which is present in the profiles from beneath arable land in enormous quantities (figure 5) with maximum annual leaching rates perhaps reaching 250 kg SO_4 ha^{-1}. Although the rate of migration of sulphate is likely to differ significantly from that of chloride, because of their differing physicochemical properties, the sulphate distribution does not appear to be solubility controlled nor related to the oxidation of naturally occurring sulphides. Thus this sulphate was probably derived largely from the original manufactured forms of N fertilizer, $(NH_4)_2SO_4$, and P fertilizer, $Ca(H_2PO_4)_2$–$CaSO_4$, with the implied somewhat lower leaching rates in recent years (figure 5) reflecting gradual replacement by NH_4NO_3 and $NH_4H_2PO_4$ during the 1960s. (In passing it must be mentioned that there is accumulating evidence of a concomitant increase in the leaching or mobility, or both, of Ca, Na, Mg, K, Ba, Sr, B and perhaps other ions, as a direct result of modern arable farming practices or indirectly through modification of unsaturated zone water–rock equilibria caused by the changing composition of soil leachates; these are likely to confront the water supply industry with additional future problems.)

The Chalk unsaturated zone pore-water to which the solute profiles relate is generally believed to be essentially immobile and, below about 2 m depth, the rock matrix is expected to remain almost fully saturated because of the very small pore sizes (Foster 1975), although this may not be true at all sites (Wellings & Bell 1980). Solute transport downwards from the zone of plant influence is thought to be controlled by a mechanism involving solute exchange, by lateral molecular diffusion, between mobile microfissure water and immobile pore water, the former eluting solutes from high-concentration 'regions' in the matrix and transferring them to lower-concentration 'regions' along its direction of flow. This mechanism has been shown to be very sensitive to variations in the rate and duration of fissure flow, the fissure geometry and the porosity and aqueous molecular diffusion coefficient in the rock matrix (Barker & Foster 1981). Non-dispersive solute movement in such formations was shown to be a rather special case. Detailed analysis of tritium profiles (Foster & Smith-Carington 1980) has also shown that perhaps up to 20 % or more of infiltration may completely 'bypass' the above slow mode of downward movement. It is suspected that such 'bypass' is associated with high-intensity winter rainfall exceeding the infiltration capacity of the microfissure system, decreasing soil moisture potentials and allowing very rapid groundwater flow in the larger fissures with insufficient time for significant solute exchange with the Chalk matrix. The 'bypass flow' could often contain relatively low nitrate concentrations, since these are prevalent in soil infiltration during most of the winter period. The tritium profile analysis also suggested that the top 2–3 m of the profile, including the soil itself, must behave in a complex fashion (Foster & Smith-Carington 1980), with rapid mixing and highly dispersive flow. This has been borne out by recent pore-water profiling for stable isotopes (^{2}H and ^{18}O), which exhibit strong seasonal fluctuations about a fairly constant long-term mean concentration in rainfall. Corresponding fluctuations in their concentration in the top 2–3 m of sequential pore-water profiles are observed (Bath *et al.* 1981), with relatively constant concentration, corresponding to that in the groundwater of the saturated zone, below (figure 4).

It is concluded that the complexity of the Chalk unsaturated zone is such that final interpretation must await the results of long-term sequential profiling. Such work is in hand but has so far produced conflicting results. Although clear downward movement of tritium and nitrate have been observed at some sites (Foster & Smith-Carington 1980; Oakes *et al.* 1981), there is as yet no definite evidence for the systematic, non-dispersive, downward movement of the main 'nitrate front' beneath long-standing arable land. Indeed, results from a site of very detailed investigation on the Cambridge Chalk show marked changes in peak concentration more consistent with dispersive flow, possibly coupled with denitrification (figure 4).

If such a flow mechanism predominated the downward movement of nitrate, it is possible that the more recent rates of leaching loss, while increasing from earlier (pre-1965) values of about 40 kg N ha^{-1} annually, are significantly less than those implied by non-dispersive flow. Since long-term mean annual infiltration is mainly in the range 200–300 mm and arable land occupies up to 80 % of the aquifer outcrop area, in the absence of denitrification, leaching rates become critical to water-supply interests in eastern England in the range 55–80 kg N ha^{-1} annually.

Current outlook

Among current trends in agricultural practice, the move from spring-sown to winter-sown cereals and perhaps also the introduction of minimal cultivation techniques should reduce nitrate leaching to groundwater from permeable arable land.

The forecast renewed increase in fertilizer application to cereals and the continued major increases on grass, however, may well prove detrimental to water-supply interests.

LONGER-TERM HISTORY OF LEACHING LOSSES

The distribution of nitrate and other solutes in the saturated zone of unconfined aquifers, and particularly at depth in that zone, relates in a general way to the longer-term history of leaching losses from agricultural soils. The data cannot be interpreted directly since they reflect the influence not only of variable leaching rates but also varying land-use, especially increasing arable acreages, unknown but perhaps significant denitrification (since nitrate-reducing bacteria have been recovered at depths of more than 20 m below groundwater table), and complex groundwater flow régimes with unknown dispersion, in which vertical, as well as lateral, flow components are present.

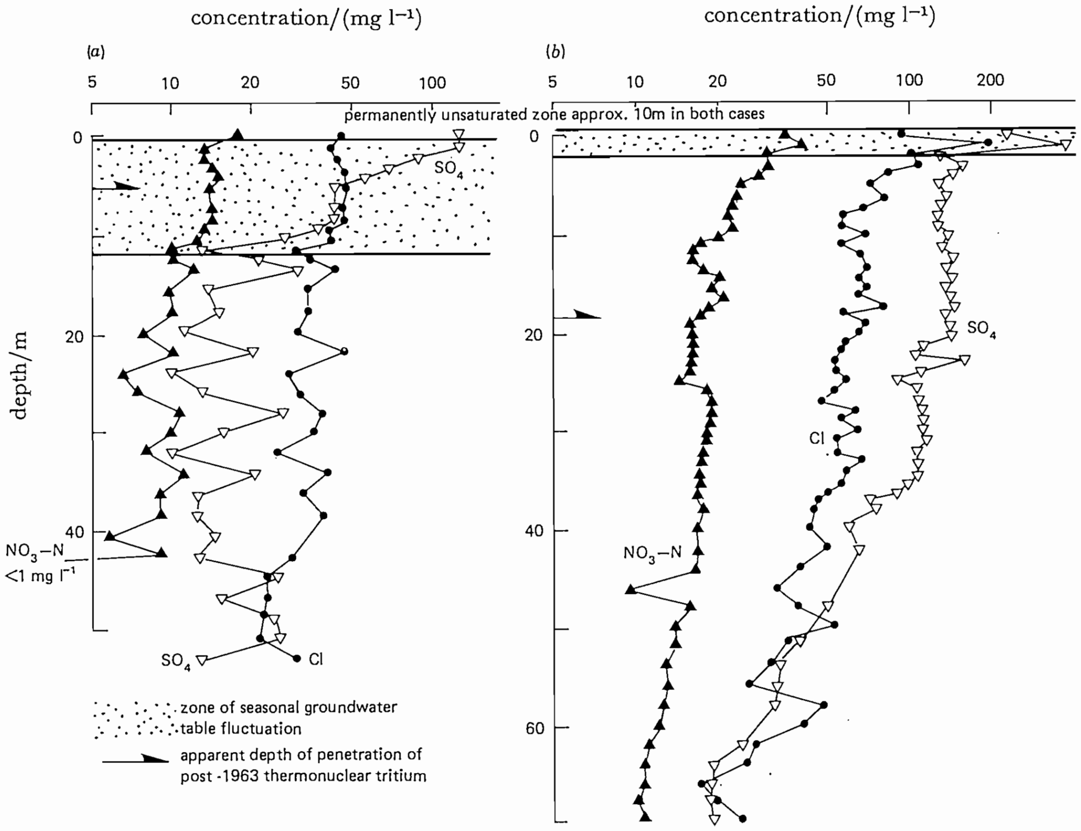


FIGURE 6. Chemical profiles of saturated zone pore-waters from outcrop areas of (*a*) West Norfolk Chalk in 1976 and (*b*) South Yorkshire Triassic Sandstone in 1980.

However, in eastern England where, for example, more than 80 % of the research catchments have been under continuous arable cropping since 1935 (the earliest land-use survey at field level) and probably throughout this century, some interesting observations can be drawn from chemical pore-water profiles in the saturated zone (figure 6). (In passing it should be mentioned that pore-water chemistry has been shown generally to agree quite closely with that of groundwater pumped from the corresponding depth interval, although, as would be expected from the above discussion, significant differences have been sometimes observed in the Chalk at depths of known major fissuring.)

At depths where chloride and sulphate concentrations of less than 20 mg l^{-1} occur, probably corresponding to the period before the widespread introduction of inorganic fertilizers (i.e. before 1940), concentrations of 8–10 mg NO_3-N l^{-1} persist and probably reflect annual leaching rates of at least 25 kg N ha^{-1} from arable land, presumably at that time fertilized more by organic manures. Steadily increasing concentrations above that depth probably reflect increasing overall use of naturally occurring and manufactured inorganic fertilizer compounds, such as $NaNO_3$, KCl and $Ca_3(PO_4)_2$, and $(NH_4)_2SO_4$ and $Ca(H_2PO_4)_2$–$CaSO_4$ respectively. The marked increase in sulphate concentrations with decreasing depth (from 10 m and 40 m upwards in figure 6*a* and *b* respectively) probably corresponds to the widespread introduction after 1940 of manufactured inorganic N fertilizers (the $(NH_4)_2SO_4$ types) by when the annual rates of nitrate leaching had risen to approach 40 kg N ha^{-1}.

CONCLUDING SUMMARY

1. A major, and in England predominant, source of nitrate leaching to groundwater is arable farmland.

2. Unfertilized and moderately fertilized pasture is estimated generally to lose no more than 10 kg N ha^{-1} annually.

3. Arable agriculture has had, and will continue to have, a major impact on groundwater quality, especially in unconfined aquifers.

4. The hydrogeological data presented provide clear evidence that the rate of nitrate leaching from the more permeable soils has increased, almost uninterruptedly.

5. They also show little evidence of denitrification *in situ* but this cannot, at present, be entirely discounted.

6. Nitrate leaching has been accompanied by increasing leaching or mobilization of chloride, sulphate and numerous other ions, some of which will also confront the water-supply industry with future problems.

7. In the days of mainly organic N fertilizer, average annual leaching losses from permeable soils were probably not much more than 25 kg N ha^{-1} but increased steadily to around 40 kg N ha^{-1} with the increasing use of inorganic forms.

8. The interpretation of the more recent rates of leaching depends on the precise groundwater flow régime and solute transport mechanisms in the Chalk unsaturated zone, which remain contentious.

9. If an essentially non-dispersive flow is assumed, as suggested by the evidence of tritium profiles, then annual leaching rates since 1963 average 50–70 kg N ha^{-1}.

10. In the 1970s, with modern arable farming practices, annual leaching rates would exceed 80 kg N ha^{-1}, an annual loss of nutrient valued in excess of £15 M from the arable land of the aquifer outcrops in eastern England alone.

11. Such losses would also imply the widespread need for alternative water supplies or water treatment facilities or both, from the 1990s onwards.

12. The régime of solute transport in the Chalk unsaturated zone is highly sensitive to the detailed properties of the fissured porous rock and to the molecular diffusion coefficient of the solute in question.

13. It is possible that the characteristic shape of Chalk unsaturated zone nitrate profiles under long-standing arable land could reflect a more dispersive mode of solute transport.

14. In this case the annual leaching losses, although increasing and well in excess of 40 kg N ha^{-1}, could be significantly less than indicated above.

15. Long-term sequential profiling in individual arable fields is required to resolve the contention; however, the limited results so far available are conflicting.

16. Even at best the nitrate leaching losses are substantial, when viewed in relation to the cost of the equivalent manufactured nutrient, and warrant continued research on methods of restricting them to a practical minimum.

17. Of direct relevance to water-supply and environmental interests would be large-scale medium-term experiments on leaching losses in well monitored groundwater catchments, involving cultivation of new winter cereal strains with the best available husbandry to maximize crop yields but not necessarily to minimize labour costs.

18. In the formulation of detailed guidelines for future agricultural practice it may be pertinent to consider independently the outcrop–recharge areas of the main water-supply aquifers.

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