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Regional analysis of groundwater nitrate concentrations and trends in Denmark in regard to agricultural influence

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Abstract

The act of balancing between an intensive agriculture with a high potential for nitrate pollution and a drinking water supply almost entirely based on groundwater is a challenge faced by Denmark and similar regions around the globe. Since the 1980s, regulations implemented by Danish farmers have succeeded in optimizing the N (nitrogen) management at farm level. As a result, the upward agricultural N surplus trend has been reversed, and the N surplus has reduced by 30–55 % from 1980 to 2007 depending on region. The reduction in the N surplus served to reduce the losses of N from agriculture, with documented positive effects on nature and the environment in Denmark. In groundwater, the upward trend in nitrate concentration was reversed around 1980, and a larger number of downward nitrate trends were seen in the youngest groundwater compared with the oldest groundwater. However, on average, approximately 48 % of the oxic monitored groundwater has nitrate concentrations above the groundwater and drinking water standards of 50 mg l^{-1}. Furthermore, trend analyses show that 33 % of all the monitored groundwater has upward nitrate trends, while only 18 % of the youngest groundwater has upward nitrate trends according to data sampled from 1988–2009. A regional analysis shows a correlation between a high level of N surplus in agriculture, high concentrations of nitrate in groundwater and the largest number of downward nitrate trends in groundwater in the livestock-dense northern and western parts of Denmark compared with the south-eastern regions with lower livestock densities. These results indicate that the livestock farms dominating in northern and western parts of Denmark have achieved the largest reductions in N surpluses. Groundwater recharge age determinations allow comparison of long-term changes in N surplus in agriculture with changes in oxic groundwater quality. The presented data analysis is based on groundwater recharged from 1952–2003, but sampled from 1988–2009. Repetition of the nitrate trend analyses at five-year intervals using dating of the groundwater recharged in the coming years and a longer time series of the nitrate analyses can reveal the evolution in nitrate leaching from Danish agriculture during the

past 10 yr. Similar analyses can be carried out to compare with other regions internationally.

1 Introduction

Intensive agriculture is a major source of environmental N (nitrogen) pollution with severe N losses to soil, water and air. The environmental effects include a decline in biodiversity, eutrophication of ecosystems and surface waters, acidification, global warming, air pollution and diffuse nitrate pollution of groundwater. N pollution from intensive agriculture not only affects the environment, but may also affects human health due, for example, to the presence of N-containing particles in the atmosphere, which may give rise to respiratory health problems and diseases, or nitrate in drinking water, which may pose risks for some types of cancer although no firm conclusions exists (van Grinsven et al., 2010; Erisman et al., 2011).

The manufacture of nitrogen-containing fertilizer for food production and the cultivation of leguminous crops convert atmospheric N_2 into reactive forms that significantly perturb the global nitrogen cycle and threaten the stability of the planet (Rockström et al., 2009). Globally, industrial N fixation has increased exponentially from near zero in the 1940s (Vitousek et al., 1997). The production of nitrogen fertilizers helps keep world crop productivity one step ahead of human population growth, but nitrogen fertilizers also cause N imbalances in agricultural development in all parts of the world (Vitousek et al., 2009).

A global challenge is to produce enough food for the ever-growing population and at the same time minimizing the loss of N to the environment. Since the 1980s, agriculture in Western Europe has managed to reduce its nitrogen surpluses, owing to stringent national and European Community policies (Vitousek et al., 2009; Grizzetti et al., 2011; Hansen et al., 2011; Dalgaard et al., 2012). However, Vitousek et al. (2009) reckon that regions in Africa continue to extract the nutrient capital of what were once highly fertile soils with low yields, while in contrast intensive agricultural production in Northern

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China has a very high input of N to agricultural fields and high yields, but also a very high N loss to the environment.

Nutrients in the soil are leached when the supply exceeds the nutrient demand of the plant. Since 1980, agriculture in Denmark has been able to reduce its N surplus by approximately 40 % while maintaining crop yields. The result of the reduction in the agricultural N surplus is reflected in respective reductions in nitrate leaching of on average 33 % (Kronvang et al., 2008), the N load in surface waters of approx. 29–32 % and groundwater nitrate concentrations of approx. 40 % (Hansen et al., 2011). Also other countries as The Netherlands (Visser et al., 2007), Belgium (Aguilar et al., 2007) and the US (Rupert, 2008; Burow et al., 2010) have observed effects on groundwater nitrate concentrations due to impact from fertilizer use in agriculture. Several Danish initiatives have been taken to reduce the N pollution from agriculture. Some of the most effective environmental measures have been a reduction in the statutory and crop-specific N fertilisation standards and N utilization requirements of manures which has raised the overall N use efficiency from 27 % in 1985 to 40 % in 2008 (Dalgaard et al., 2011a).

In Denmark, public drinking water supplies almost entirely originate from groundwater and approximately 15 % of the total area of Denmark has therefore been classified as nitrate-vulnerable abstraction areas (Hansen and Thorling, 2008) with many waterworks and wells having been turned off due to nitrate pollution. Groundwater protection is therefore a high priority and since 1985 it has been one of the most important drivers of regulation of the Danish agricultural sector through national action plans (Kronvang et al., 2008) and EU policies (Uthes et al., 2011; Happe et al., 2011).

The present paper continues the analysis initially presented and published in Hansen et al. (2011). The focus is still on nitrate in the oxidized zone of the Danish groundwater because nitrate in oxic groundwater can be regarded as a conservative compound directly comparable to the nitrate leached from the root zone when the age of the groundwater recharge is known. The aims of the present study are to better understand the geographic distribution of nitrate in groundwater and the evolution of nitrate trends in Denmark at different groundwater recharge ages by examining the influence from (1) regionally calculated N losses from agriculture and (2) the local amount of groundwater recharge.

2 Methods

2.1 Agricultural, geological and hydrological conditions

Denmark has a total land area of about 43 000 km^2, and about two thirds of this is under agricultural use. The fertilization rate in Denmark is high compared with other European countries (OECD, 2010; European Environmental Agency, 2005). The average livestock density is about 0.8 livestock units per hectare (Dalgaard et al., 2011b), and the average input of N to agricultural land is about 180 $kg\,ha^{-1}\,yr^{-1}$ in 2008 (Statistics, 2010; Kronvang et al., 2008). The land surface has a modest topography where the highest point is 170 m above sea level. The climate in Denmark is coastal temperate, and the precipitation varies from about 600 to 1000 $mm\,yr^{-1}$. The upper geologic layers are mainly 50–200 m thick Quaternary glacial deposits underlain by Tertiary marine and fluvial deposits or Cretaceous limestone and chalk. The aquifers thus consist of either unconsolidated sands and gravels or fractured limestone and chalk.

2.2 Nitrate reduction in Danish groundwater

The Danish groundwater can be divided into an upper oxic zone and a, usually, deeper reduced zone (Fig. 1). Nitrate reduction takes place in an intervening zone, called the nitrate-containing anoxic zone, between the oxic and the reduced zones. Hydrogeological heterogeneity and variation in the reduction capacity of the sediments can locally result in a complex transition between the oxic and the reduced zones (Hansen and Thorling, 2008). These circumstances also give rise to variation in the thickness of the anoxic zone from a few mm to more than 15 m across the country. In Denmark,

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the oxic zone in the Quaternary deposits has developed after the latest glaciations. On exposed residues of Saalean landscapes in Western Denmark, the oxidation processes have been active for more than 100 000 yr, whereas in the rest of the country, they have only been active for about 12 000 yr due to differences in the extension of the latest glaciations.

Nitrate reduction in the soil and groundwater is often microbially controlled. In the unsaturated zone, the denitrification processes take place in the reduced microenvironments due only to reduction of organic matter (Ernstsen, 1999). In the groundwater aquifers, organic matter, ferrous ions and pyrite (Postma, 2001) are the dominating nitrate-reducing agents causing the denitrification processes that take place in the nitrate-reducing anoxic zone.

2.3 Nitrate concentrations in oxic groundwater

Data on nitrate concentrations from all types of wells (monitoring, investigations, abstraction, etc.) have been integrated to obtain a national overview of the geographic distribution of the nitrate in oxic groundwater in Denmark set out in Fig. 2. Like many other data used for chemical analyses of the Danish groundwater, nitrate concentrations are being reported to the National database JUPITER. The data used in Fig. 2 were downloaded in January 2011 and consist of 3757 analyses sampled in the period 1890–2010. Data from such a long period are used in order to obtain as many nitrate analyses as possible from the oxic zone to create a national overview. Data stored in JUPITER have been analyzed by professionally certified laboratories.

Before nitrate concentrations are determined in the laboratories, the groundwater samples undergo normal analysis in the field which includes online measurements of pH, redox potential, oxygen concentration, temperature and conductivity. This approach ensures a high analytical quality and representative groundwater samples. Performing field analyses has been normal procedure over the last approximately 20 yr in the Danish groundwater monitoring programme. The sampling and the field analyses are performed according to Danish technical standards.

The redox condition of the groundwater is used to segregate the relevant subset of data used in this study where only data from the oxic zone are used. This subset of data from the oxic zone represents groundwater with a nitrate content mirroring the original nitrate leaching from the root zone. In oxic groundwater, nitrate is expected to act as an inert tracer due to the presence of oxygen and the generally low reactivity of organic matter below the root zone.

In this study the oxic zone is defined as:

- Nitrate > 1 $mg\,l^{-1}$,
- Iron < 0.2 $mg\,l^{-1}$, and
- Oxygen > 1 $mg\,l^{-1}$.

2.4 Nitrate concentrations used for trend analyses on a national scale

Only nitrate concentrations from the Danish Groundwater Monitoring Programme are used in the national trend analysis presented in Fig. 3. Details about the purpose, construction and hydro-geological conditions of the sites in the Danish Groundwater Monitoring Programme can be found in Jørgensen and Stockmarr (2009) and Hansen et al. (2011). The entire Danish monitoring data set numbers approximately 46 800 nitrate analyses from 1500 groundwater sampling points. Only a subset of the complete dataset is used in the trend analyses in the present study.

The nitrate concentrations used for trend analyses on a national scale originate from 194 groundwater monitoring points with oxic groundwater where the groundwater recharge age has also been determined using the CFC method, typically once during the period 1997–2006. The groundwater recharge age determination allows the comparison of long-term changes in N surplus in agriculture with changes in oxic groundwater quality (Hinsby et al., 2008). The CFC analyses were performed according to the procedure of Laier (2005) as described in Hansen et al. (2011).

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2.5 Nitrate concentrations used for trend analyses on an individual scale

Nitrate concentrations from the Danish Groundwater Monitoring Programme are also used in the trend analyses for individual point measurements depicted in Figs. 2, 4 and 5.

The nitrate concentrations used for trend analyses for individual point measurements comprise 5321 nitrate analyses from 152 sampling points sampled from 1988–2009 where the groundwater (1) had stable oxic conditions, (2) was CFC-dated and (3) had time series of between 8 and 20 yr with approximately one nitrate analysis per year.

The groundwater chemistry data from the Danish Groundwater Monitoring Programme used in the trend analyses were downloaded from the Danish national geodatabase (JUPITER) in October 2009 (www.geus.dk).

2.6 Nitrogen surpluses in agriculture

The surplus of N in agriculture is defined as the balance between inputs (synthetic fertilizer, import of animal feed, organic waste products, net atmospheric deposition and fixation) and outputs (export of plant and animal products). The surplus of nutrients, and especially N, is regarded as the best overall environmental indicator for the changes in the agricultural impact on the environment over a certain time period (European Environmental Agency, 2005). The surplus represents the amount of N pooled in the soil, or not being used up by the production system, and which is therefore at risk of being lost to the environment (Dalgaard et al., 2010; Hansen et al., 2000). The surplus is formulated in Eq. (1):

$$N_{\text{surplus}} = N_{\text{emission}} + N_{\text{accumulation}} + N_{\text{leaching}} \quad (1)$$

In this paper, the regional N surpluses for each of the ten Danish geo-regions shown in Fig. 2 are estimated from the annual, national N surplus as accounted for by Hansen et al. (2011). For each geo-region and for each year from 1950 to 2007, the livestock units are sourced from national county statistics and accord with the linear relationship

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between livestock units and N surplus identified by Dalgaard et al. (2011b). The annual, national N surplus values are apportioned according to the number of livestock units in each geo-region.

2.7 Water balances

Water balance components were estimated by the national water resources model called the DK model (Højberg et al., 2012), which is a coupled surface-groundwater model with a horizontal discretization of 500 × 500 m covering the entire country with the exception of minor islands. The model is set up in the MIKE SHE/MIKE11 model system, where the unsaturated zone is described by a water balance module, while the saturated zone is described by a comprehensive three-dimensional groundwater component to estimate recharge to and hydraulic heads in different geological layers. Stream-aquifer interaction and stream flow-routing are described by MIKE11. The model is constructed on the basis of comprehensive national databases on geology, soil, topography, river systems, climate and hydrology and has recently been updated to include hydrological interpretations from regional- and local-scale hydrological models.

Daily groundwater recharge values (mm day^{-1}) are extracted from the model simulations with MIKE SHE and represent 10-yr average values for the period 1998–2007. For each of the 152 groundwater monitoring points, a groundwater recharge value is found from the 500 × 500 m cell from the groundwater model where the well is situated.

2.8 Statistical methods

2.8.1 Gridding of nitrate concentrations

The data on the nitrate concentration map in Fig. 2 were interpolated using the kriging method of the Surfer programme (Surfer, 2002). A semi-variogram was fitted with an exponential function. The search radius of 10 km and a cell size of 2500 m were used.

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2.8.2 Nitrate trend analyses

Determination of nitrate trends was achieved using the SAS software system (SAS, 2008) as described in Hansen et al. (2011).

3 Results and discussion

3.1 Geographic assessment of nitrate in oxic groundwater

Figure 2a shows that nitrate has been found in the oxic part of the groundwater throughout Denmark, with concentrations of up to 360 mg l^{-1} when using average nitrate concentrations based on all available data from 1890–2010. Nitrate concentrations of 25 mg l^{-1} are exceeded in 54 % of the datasets, corresponding to 55 % of the total area of Denmark, and nitrate concentrations of 50 mg l^{-1} are exceeded in 24 % of the dataset, equivalent to 10 % of the total area of Denmark (Fig. 2a). However, if we consider only data from the Groundwater Monitoring Programme sampled from 1989–2009 and representing ages up to maximum 50 yr, then the nitrate concentration of 25 mg l^{-1} and 50 mg l^{-1} are exceeded in 79 % and 48 % of the 152 oxic monitoring points, respectively. The nitrate concentrations in the oxic groundwater data from the Groundwater Monitoring Programme are higher than the concentrations that appear from the data shown in Fig. 2a because the oxic data from the Groundwater Monitoring Programme on average represent younger and more nitrate-polluted groundwater.

However, Fig. 2a shows a tendency towards a regionalization of the highest concentrations of nitrate. The general picture shows that the groundwater is most severely polluted with nitrate in Northern and Western Denmark (geo-regions I, II, III, IV, and V) while Eastern Denmark (geo-regions VI, VII, VIII, IX, and X) is less polluted with nitrate.

An examination of the geographic distribution of the upward and downward nitrate trends and non-significant nitrate trends in the 152 oxic groundwater monitoring points sampled from 1989–2009 reveals no obvious regional pattern (see Fig. 2b, c). However,

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the most pronounced downward nitrate trends seem to be where the concentrations of nitrate in the groundwater are highest in Northern and Western Denmark (geo-regions I, II, III, IV, and V).

The occurrence of oxic groundwater with high concentrations of nitrate is most likely due to a combination of: (1) insufficient protection of the aquifer from overlying clay layers, (2) a low nitrate reduction capacity of the sediments of the aquifer, e.g. low content of potential reduction agents like pyrite, Fe^{II} and organic matter, (3) a high groundwater recharge, and (4) high nitrate leaching from land use. Pronounced pollution of groundwater with nitrate is therefore likely to be found where the nitrate interface has penetrated deeply into the soil layers (see Fig. 1).

3.2 Regional trends of N surpluses and nitrate in oxic groundwater

Denmark can be divided into ten different geo-regions, and regional N balances have been calculated for each geo-region as seen in Fig. 2a. The regional balances are shown together with the national N balance in Fig. 3 and Table 1. All the ten different regional N balances have the same overall oscillation pattern as the national N balance with minimums and maximums occurring at the same time. However, the regional N balances are staggered so that Northern and Western Jutland (geo-regions I, II, III, IV and V) have a higher and Eastern and Southern Denmark (geo-regions VI, VII, VIII, IX, X) have a lower N surplus level than the national average N surplus level.

The higher N surplus level in Northern and Western Jutland and lower N-surplus level in Eastern and Southern Denmark is comparable to the geographic pattern of the nitrate concentration levels in oxic groundwater seen in Fig. 2a.

In almost every geo-region in Denmark, the N-surpluses were approximately 50 kg N ha^{-1} yr^{-1} in 1950, where the country as a whole was characterized by mixed farming and the livestock was distributed evenly across all regions (Dalgaard and Kyllingsbæk, 2003). Over the next 30 yr, N surpluses rose dramatically in every geo-region and they reached a maximum around 1980 where they ranged from 234 kg N ha^{-1} yr^{-1} in Thy (geo-region III) in northern Jutland to 91 kg N ha^{-1} yr^{-1} in

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North Zealand (geo-region VIII) in the eastern part of Denmark. Since 1980, the N surpluses have decreased by 30–55 % in all Danish geo-regions. The national average N surplus reduction since 1980 is about 37 % and the national average N surplus was 117 kg N ha^{-1} yr^{-1} in 2007 (see Table 1).

The trends in the regional N surpluses and the national N surpluses both show the same fluctuations as the nitrate concentration trends in oxic groundwater at the national level (Fig. 3). The nitrate concentrations in oxic groundwater are shown as a 5-yr mowing average curve based on groundwater measurements (nitrate analyses and CFC dating) performed from 1988–2009 representing groundwater recharged from 1952–2003 (Fig. 3). This phenomenon is elucidated in detail in Hansen et al. (2011), who report a statistically significant nitrate trend reversal in oxic groundwater around 1980. Figure 3 also shows that the increase in N surpluses in agriculture in the 1950s, 1960s and 1970s leveled out after 1980.

3.3 Nitrate trends in oxic groundwater

The nitrate trends of the time series of nitrate concentrations in the 152 oxic groundwater monitoring points were assessed by linear regression, as described in detail in Hansen et al. (2011). The slopes of each of these 152 linear regression lines represent the nitrate trend or the changes in nitrate shown in mg nitrate l^{-1} yr^{-1}. Thus, negative slopes represent downward trends, while positive slopes represent upward nitrate trends. In Fig. 4, the upward and downward nitrate trends are illustrated according to the age of the groundwater determined with the CFC method and the data are divided into three age groups.

The general national nitrate trends (Fig. 3) show a trend reversal around 1980; however, the nitrate trends from the individual monitoring points show a more complex picture as seen in Fig. 4 and Table 2.

Ninety-four of the 152 oxic groundwater monitoring points have statistically significant ($p < 0.05$) nitrate trends of which 50 are upward and 44 are downward. The remaining 58 monitoring points have non-significant nitrate trends.

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As far as the upward nitrate trends are concerned, 20 % is in the youngest groundwater (< 15 yr), while 42 % is in the oldest groundwater (25–50 yr). The reverse pattern is found for the downward nitrate trends where 54 % is in the youngest groundwater and only 7 % in the oldest. Thus, the youngest groundwater has the highest proportion of downward nitrate trends with the highest rate of changes in the nitrate concentrations ($mg\,l^{-1}\,yr^{-1}$) as seen in Fig. 4.

The rate of changes in the nitrate concentrations ($mg\,l^{-1}\,yr^{-1}$) are highest in the groundwater monitoring points with the highest concentrations, which is both reflected in the significant upward and downward nitrate trends and in the non-significant nitrate trends (see Fig. 5a). Sixty-four per cent of the downward nitrate trends have mean nitrate concentrations above $50\,mg\,l^{-1}$ as opposed to only 38 % of the upward nitrate trends. These findings are in line with the indications in Fig. 2 that the largest number of downward nitrate trends is found where the mean nitrate concentrations in groundwater are highest, i.e. in the northern and western parts (geo-regions I, II, III, IV, and V).

As far as groundwater recharge is concerned, most of the upward and downward nitrate trends (approx. 50 %) are found at an annual mean recharge level of 400–600 $mm\,yr^{-1}$, and no obvious differences can be seen between the upward and downward nitrate trends (Fig. 5b).

The N load to groundwater is calculated by multiplying the nitrate concentrations and the groundwater recharge. Twenty-five per cent of the groundwater monitoring points with downward nitrate trends have a high N load (> $75\,kg\,N\,ha^{-1}\,yr^{-1}$) to the groundwater compared with only 11 % of the upward nitrate trends. This finding seems to be due to the nitrate concentration levels rather than to the local mean level of the groundwater recharge in Denmark.

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4 Conclusions

Denmark has seen its farming sector develop a high livestock density due notably to the increase in pig production and the concentration of dairy farming in Western Denmark over the last century (Dalgaard and Kyllingsbæk, 2003). Farms with a high livestock density have accomplished larger reductions in N surplus between the years 1990 and 2008 than farms with cash crop production using synthetic fertilizers and little livestock manure (Dalgaard et al., 2012). These circumstances can explain the findings in the present study where there seems to be consistency between a high N surplus in agriculture, high concentrations of nitrate in groundwater and the most pronounced downward nitrate trends in groundwater in northern and western parts of Denmark. In absolute values, the reduction in N surplus from 1980–2007 in these regions is also highest (74–87 $kg\,N\,ha^{-1}\,yr^{-1}$); however, the relative reduction in N surplus is lower (34–37 %) than in Eastern and Southern Denmark (40–55 %).

A clear indication of an effect of reduced nitrate leaching on groundwater nitrate concentrations in Denmark is seen in the age of the groundwater recharge relating to the upward and downward nitrate trends. Here 20 % of the youngest (0–15 yr old) and 42 % of the oldest (25–50 yr old) groundwater display upward nitrate trends, while the opposite pattern is seen for the downward nitrate trends where 54 % can be found in the youngest and 7 % are in the oldest groundwater.

Mean nitrate concentrations above 50 $mg\,l^{-1}$ are seen in 64 % of the downward nitrate trends in oxic groundwater, but only in 38 % of the upward nitrate trends. The N load in groundwater is the amount of nitrogen being transported by the groundwater and which might eventually flow out into groundwater-dependent ecosystems. The geographical variation in nitrate concentrations in groundwater rather than the amount of groundwater recharge influences the distribution of the groundwater N load, and 25 % of the groundwater monitoring points with downward nitrate trends have a high N load compared with only 11 % of the upward nitrate trends.

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Together with the findings in Hansen et al. (2011), this study demonstrates a clear relationship between changes in the N surplus in agriculture, both at national and regional level, and changes in the nitrate concentrations in oxic groundwater with the same synchronic oscillations and trend reversals around 1980. The change and the development in Danish agricultural management of N have been driven mainly by politically enforced regulations since 1985, but also changes in the economic and technical conditions for farming with fluctuating product prices and revisions of the European Agricultural Policies have been instrumental in accomplishing these changes (Uthes et al., 2011; Happe et al., 2011). Regulations in agriculture have a significant impact on the environment, as evidenced by the simultaneousness of the reduction in N surplus in agriculture and the reduced nitrate contamination of oxic groundwater. The experience from Denmark may provide inspiration for other countries where control of agricultural N losses is needed, as for example pointed out by Vitousek et al. (2009).

Of all the coastal waters in the world, those in Denmark are some of the most frequently exposed to hypoxia (Diaz and Rosenberg, 2008). The EU Water Framework Directive stipulates that Denmark reverses the upward trends in groundwater nitrate concentrations and complies with the groundwater quality standards of 50 mg l^{-1} in certain areas. Thus, the environmental goals in both Danish legislation and EU directives (The Nitrate Directive, 1991/696/EC; The Water Framework Directive, 2000/60/EC; the Groundwater Directive, 2006/118/EF) have not yet been fully met. There is a need for more holistic future solutions to protect both groundwater, nature and the wider environment and to meet legislative requirements for a good chemical status of groundwater, and a good ecological status of the Danish estuaries and oceans is just one of the important goals.

The latest report from the Danish agricultural monitoring sites (Grant et al., 2011) shows a small increase in modelled nitrate leaching from 2003 to 2010. The results presented in this paper are based on groundwater data sampled from 1988–2009, and they represent groundwater recharged from 1952–2003 where the oldest monitored groundwater is about 46 yr old and the youngest monitored groundwater is about 6 yr

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old. Repetition of the nitrate trend analyses at five-year intervals based on dating of the groundwater recharge in the coming years and a longer time series of the nitrate analyses will shed more light on the groundwater effect of the evolution in nitrate leaching from Danish agriculture during the past 10 yr.

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Table 1. Distribution statistics of annual agricultural N surpluses from 1950 to 2007 in 10 different geo-regions of Denmark, and in Denmark as a whole.

Georegion No.	Georegion Name	1950	Max	2007	Mean 1950–2007	Increase 1950–2007	Reduction 1980–2007
I	West Jutland	48	217	143	139	95 (198 %)	74 (34 %)
II	Mid Jutland	50	200	126	130	76 (152 %)	74 (37 %)
III	Thy	50	234	147	148	97 (194 %)	87 (37 %)
IV	North Jutland	50	212	138	137	88 (176 %)	74 (34 %)
V	Himmerland	50	213	135	137	85 (170 %)	78 (37 %)
VI	Djursland	50	177	106	117	56 (112 %)	71 (40 %)
VII	East Denmark	51	162	96	117	45 (88 %)	66 (41 %)
VIII	North Zealand	40	91	41	60	1 (3 %)	50 (55 %)
IX	South Zealand	40	101	58	68	17 (43 %)	43 (42 %)
X	Bornholm	57	183	127	125	70 (123 %)	56 (30 %)
DK	Denmark (in total)	49	187	117	122	68 (139 %)	70 (37 %)

Table 2. Amount (%) of statistically significant upward, statistically significant downward and non-significant nitrate trends in 152 oxic CFC-dated groundwater monitoring points with a 95 % confidence level. The nitrate trends are grouped according to (1) groundwater recharge age, (2) the average nitrate concentrations in groundwater, (3) the annual groundwater recharge, and (4) the N load to groundwater.

		Upward	Downward	Non significant	Total
Total		50 (100 %)	44 (100 %)	58 (100 %)	152 (100 %)
Recharge age	< 15	10 (20 %)	24 (54 %)	21 (36 %)	55 (36 %)
(yr)	15–25	19 (38 %)	17 (39 %)	28 (48 %)	64 (42 %)
	25–50	21 (42 %)	3 (7 %)	9 (16 %)	33 (22 %)
Nitrate in groundwater	1–10	6 (12 %)	0 (0 %)	5 (9 %)	11 (7 %)
($mg l^{-1}$)	10–50	25 (50 %)	16 (36 %)	27 (47 %)	68 (45 %)
	≥ 50	19 (38 %)	28 (64 %)	26 (44 %)	73 (48 %)
Groundwater recharge	< 400	19 (38 %)	15 (34 %)	23 (40 %)	57 (38 %)
($mm yr^{-1}$)	400–600	24 (48 %)	23 (52 %)	21 (37 %)	68 (45 %)
	600–750	7 (14 %)	6 (14 %)	13 (23 %)	26 (17 %)
N load to groundwater	< 25	18 (36 %)	15 (34 %)	21 (37 %)	54 (36 %)
($kg N ha^{-1} yr^{-1}$)	25–75	25 (50 %)	18 (41 %)	23 (40 %)	66 (44 %)
	≥ 75	7 (14 %)	11 (25 %)	13 (23 %)	31 (20 %)

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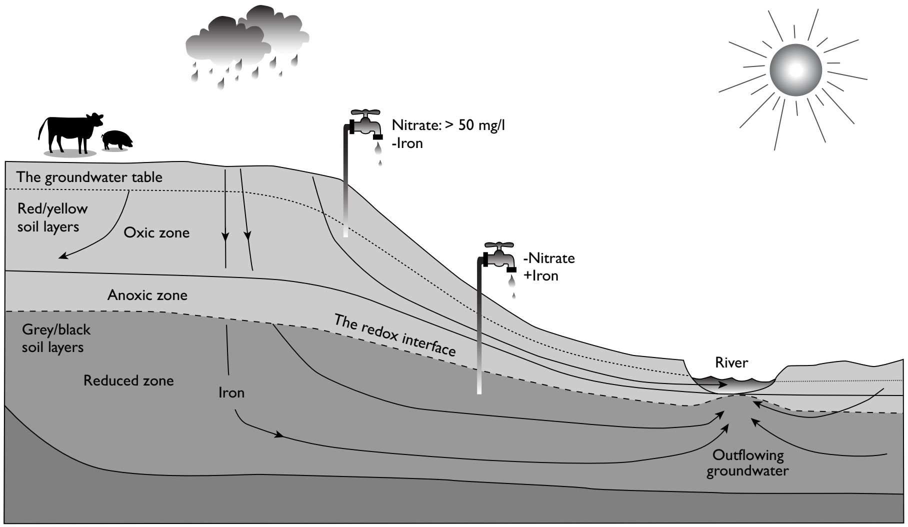

Fig. 1. Conceptual model of the typical groundwater redox environment in Denmark with oxic zone, anoxic zone with nitrate reduction and reduced zone. The interface between the anoxic zone containing nitrate and the reduced nitrate-free zone is called the nitrate interface and can also be determined based on the colours of the sediments. Above the nitrate interface, the soil layers have red and yellow colours and below the nitrate interface the soil layers have grey and black colours.

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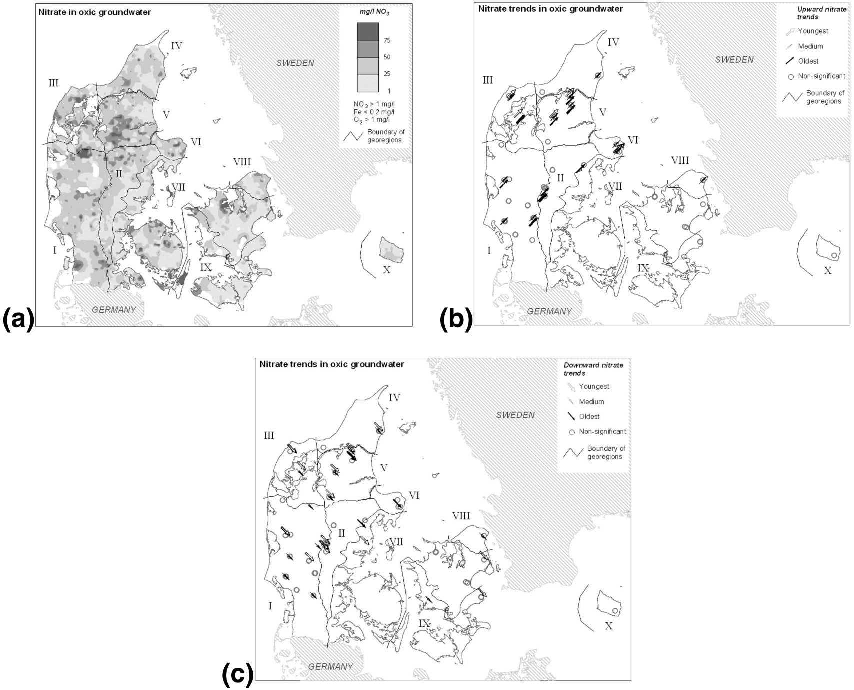

Fig. 2. (a) Interpolated geographic distribution of nitrate concentrations in oxic groundwater in Denmark based on 3757 analyses performed from 1890 to 2010. Average values from each measuring point are used in the interpolation. Shown are also 10 different geo-regions. **(b, c)** Geographic distribution of nitrate trends determined in 152 oxic CFC-dated groundwater monitoring points sampled from 1988 to 2009. A downward nitrate trend represents a negative slope of the linear regression line of nitrate versus sampling year for each groundwater monitoring point, while an upward trend shows a positive slope. The upward and downward nitrate trends are statistically significant with a 95 % confidence level. Statistically non-significant nitrate trends are also shown.

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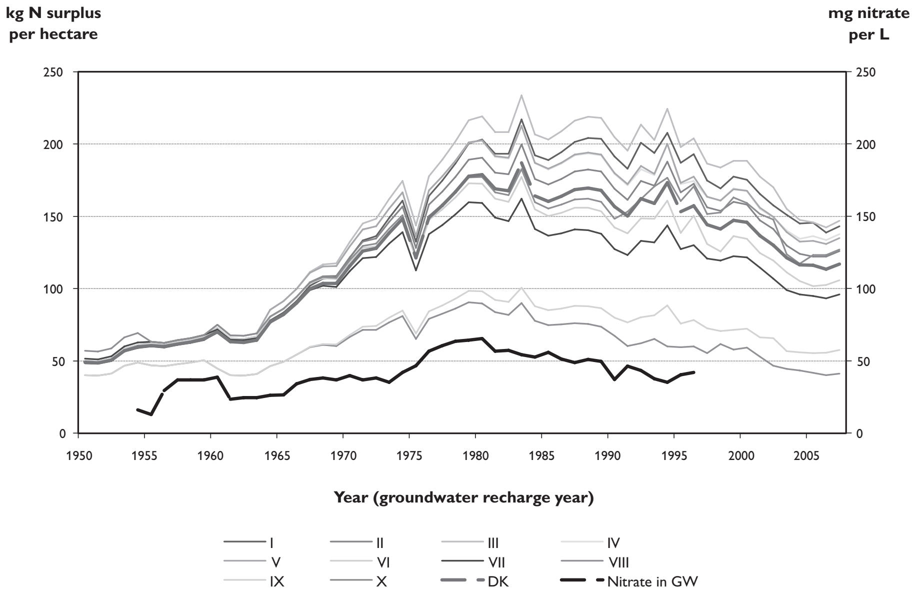

Fig. 3. Time series of agricultural N surplus in 10 different geo-regions of Denmark, and nitrate in oxic groundwater versus recharge age (CFC-age) at an annual mean level. The nitrate concentrations in oxic groundwater are shown as a 5-yr mowing average curve. The location of the 10 geo-regions is seen in Fig. 2a.

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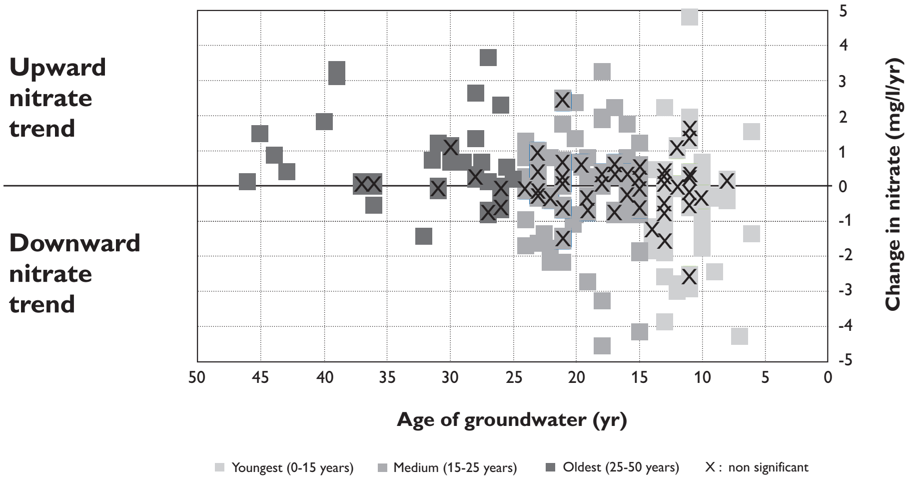

Fig. 4. Three age groups of upward and downward nitrate trends in oxic groundwater at a monitoring point level for 152 oxic CFC-dated groundwater measuring points sampled from 1988 to 2009. The change in nitrate ($mg\,l^{-1}\,yr^{-1}$) is equivalent to the slope of the linear regression lines of nitrate versus sampling year for each groundwater monitoring point. The age of the groundwater is determined with the CFC method.

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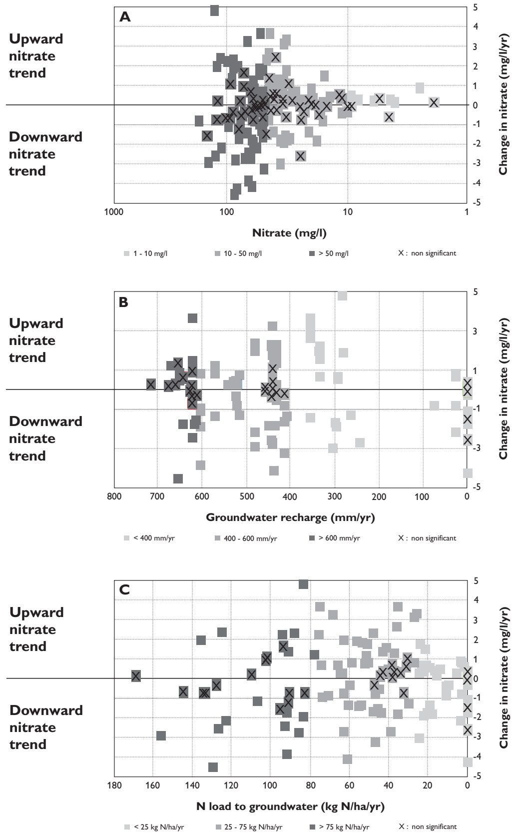

Fig. 5. Upward and downward nitrate trends in 152 oxic CFC-dated groundwater monitoring points sampled from 1988 to 2009 according to: **(A)** The mean nitrate concentration ($mg\,l^{-1}$) in each groundwater monitoring point, **(B)** the annual groundwater recharge ($mm\,yr^{-1}$) in the $500 \times 500\,m$ cell from the groundwater model where the well is situated, **(C)** the N load ($kg\,N\,ha^{-1}\,yr^{-1}$) to groundwater based on data shown in **(A)** and **(B)**.