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Application of the Netherlands Groundwater Model, LGM, for calculating concentration of nitrate and pesticides at abstraction wells in sandy soil areas of the Netherlands

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Report and Appendices

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PREFACE

The policy of the Directorate-General for Environmental Protection (DGM) of the Ministry of Housing, Spatial Planning and the Environment (VROM) aims at safeguarding an adequate industrial and drinking-water supply. Supply of good-quality water is of eminent importance for the health and welfare of society. However, water suppliers have, for several decades, been faced with the consequences of the increasing deterioration of environmental quality. Of special concern is the degradation of the quality of groundwater and surface water, which are a source for drinking-water production.

Policy measures are now being developed for environmental quality improvement. An increase in environmental quality can have a positive impact on the industrial and drinking-water supply. The effectiveness of policy measures for improving environmental quality and, ultimately, the quality of the sources for the industrial and drinking-water supply can be evaluated by means of scenario calculations.

This report will document the application of the quasi-three-dimensional RIVM groundwater model, LGM (version 2), for calculating pathlines, travel times and concentrations at abstraction wells. One scenario was calculated for nitrate leaching and three for leaching of selected pesticides into saturated groundwater. LGM was used for the so-called sandy soil areas of the Netherlands, the area for modelling comprising the entire surface area of the Netherlands, with the exception of the western part - a strip of about 60 km along the coast - and the southernmost part of the province of Limburg. The study applied to abstractions from phreatic and semi-confined aquifers; abstractions in the vicinity of large rivers (bank-infiltration abstractions) were not considered.

The research was carried out in the framework of project no. 739106 (Infrastructure for Drinking-Water and Industrial Water Supply, RIVM Long-Term Activity Programme MAP1995) and project no. 703717 (Prognosis Drinking Water, RIVM Long-Term Activity Programme MAP1996 and MAP1997) on commission of the Directorate-General for Environmental Protection (DGM) of the Ministry of Housing, Spatial Planning and the Environment (VROM).

This study was carried out with the help of many colleagues at the RIVM. The authors wish to extend special thanks to Ir. J.H.C. Mülschlegel as the project leader of this study, and to Ir. G. van Drecht and Ir. L.J.M. Boumans. Gerard van Drecht carried out the modelling of the nitrate leaching (Chapter 4), while Leo Boumans developed the "forest model" for simulating nitrate leaching in forested areas (section 4.2.2). The authors also wish to thank their colleagues Dr.ir. A. Leijnse, Ir. A.M.A. van der Linden and Dr.ir. A.F. Bouwman. Toon Leijnse's special contribution was advice on mathematical aspects of finite-element modelling, while Ton van der Linden contributed to the pesticide leaching modelling. Lex Bouwman gave valuable advice on land use in the Netherlands.

TABLE OF CONTENTS

APPENDICES, published separately

APPENDIX A: Description of all groundwater abstractions sites for drinking-water supply.

APPENDIX B: Description of 165 phreatic and semi-confined groundwater abstraction sites for drinking-water supply, as used for calculation of concentration breakthrough curves.

APPENDIX C: Well capture zones for 165 phreatic and semi-confined abstraction sites, as used for calculation of concentration breakthrough curves.

APPENDIX D: Well capture zones and pathlines for 10 selected abstraction sites.

APPENDIX E: Control breakthrough curves for 165 phreatic and semi-confined abstraction sites.

APPENDIX F: Concentration results nitrate for 14 selected abstraction sites to shallow groundwater, scenario EC(plus).

APPENDIX G: Concentration results atrazine for 14 selected abstraction sites to shallow groundwater, scenario EC.

APPENDIX H: Concentration results bentazone for 14 selected abstraction sites to shallow groundwater, scenario EC.

APPENDIX I: Concentration results 1,2-dichloropropane for 14 selected abstraction sites to shallow groundwater, scenario EC.

ACRONYMS

GEOPESTRAS	the model for simulation of the pesticide leaching flux into saturated groundwater
LGM	the Netherlands Groundwater Model (Landelijk Grondwater Model), used at RIVM
LGMGRID	a module of LGM, for generation of finite-element grid
LGMSAT	a module of LGM, for simulation of groundwater heads and fluxes in a quasi- three-dimensional, multi-aquifer system
LGMCAL	a module of LGM, for calibration (parameter estimation) for LGMSAT
LGMFLOW	a module of LGM, for calculation of pathlines and travel times
LGMCAM	a module of LGM, for stream-tube-based calculation of solute breakthrough curves at abstraction wells
LGMCAD	a module of LGM, for random-walk-based calculation of concentration in space and time
LGN	the soil-use map of the Netherlands (landelijke grondgebruikskaart)
LMD	the Dutch national monitoring network of drinking-water quality
LMG	the Dutch national groundwater quality monitoring network
LMM	the Dutch national network for monitoring the effectiveness of animal waste control policy
NLOAD	the model for simulation of the nitrate leaching flux into saturated groundwater (at agricultural land)
REWAB	the water-quality registration system of drinking-water supply companies
RIVM	the National Institute of Public Health and the Environment

ABSTRACT

In a study aimed at assessing the impact of historical and future solute leaching into saturated groundwater, the quasi-three-dimensional RIVM groundwater model, LGM (version 2), was used for calculating pathlines, travel times and concentration breakthrough curves at 165 groundwater abstraction locations in the sandy soil areas of the Netherlands. The future concentration variations were assessed for nitrate and three pesticides (atrazine, bentazone and 1,2-dichloropropane). This study was carried out in the framework of the Dutch government's long-term planning for industrial and drinking-water supply. The assessment was carried out for 76 phreatic and 89 semi-confined abstraction sites on the basis of the 1988 maximum-permitted abstraction rates. Three economic scenarios were used for generating solute leaching into saturated groundwater for the scenario years 2000 and 2020. The nitrate leaching flux was simulated by a combination of methods, including the NLOAD model for agricultural land. The pesticide leaching flux was simulated by means of the GEOPESTRAS model. Subsequently, LGM was applied to calculate the concentrations at the abstraction wells, using the concentrations in the leaching flux as input. LGM is a numerical model (based on the finite-element method), comprising complex geohydrological system components. LGM uses spatially variable (heterogeneous) data for four aquifers, covering the entire surface area of the Netherlands. The concentration breakthrough curves were calculated with the module LGMCAM, based on forward pathline tracking in saturated groundwater. The processes used in LGMCAM were advection and full mixing in well screens. Neither denitrification nor degradation of pesticides in saturated groundwater were taken into account. The results are presented (1) as concentration maps for the 165 abstraction sites for 2020 and 2050 and (2) by means of time-based concentration bar charts (1950-2050) for the totalized abstraction rate for phreatic and semi-confined abstraction sites. For a number of abstraction sites a comparison has also been made between calculated and observed breakthrough concentrations. The calculated concentrations can be concluded to be a worst-case outcome. Finally, recommendations for future improvements of the method are given.

SUMMARY

The aim of this study was to assess the impact of historical and future leaching of nitrate and pesticides on the concentrations in abstracted groundwater. The assessment of the future development of nitrate and pesticide load on soil was based on various economic scenarios. The study was carried out in the framework of the Dutch government's long-term planning for industrial and drinking-water supply. The results will be of use in:
(1) obtaining insight into the extent of the problem;
(2) designing possible policy measures for improving environmental quality, including the quality of abstracted groundwater as a source for drinking-water production, and
(3) evaluating the effectiveness of (future) policy measures.

This report documents the application of the quasi-three-dimensional RIVM groundwater model, LGM (version 2), for calculating pathlines, travel times and concentration breakthrough curves at the abstraction sites of the water-supply companies in the sandy soil areas of the Netherlands. The modelled area covers most of the country, focusing on 165 abstraction sites, namely 76 phreatic and 89 semi-confined. The abstractions in the vicinity of large rivers (referred to as bank-infiltration or river-system abstractions) were not considered. Though the concentration variation was calculated for a longer time period, it is presented only for the period 1950-2050. Steady-state groundwater flow is simulated using the maximum-permitted abstractions rates for the year 1988.

Methods and models

Use was made of three economy-based scenarios (*Divided Europe*, *European Coordination* and *Global Competition*) for generating the load of nitrate and pesticides onto the land surface. The three scenarios were developed by the RIVM for the Environmental Outlook 1997-2020. The "historical" leaching flux into the saturated groundwater was calculated for the years 1950, 1960, 1970, 1980, 1990 and 1995, while the leaching flux for the scenarios was calculated for the years 2000 and 2020.

Three specific pesticides, namely 1,2-dichloropropane, atrazine and bentazone, were considered as the best-known examples of pesticides posing a problem for drinking-water production.

The nitrate leaching flux was simulated by combined methods, the choice of a method depending on the land use. The nitrate leaching flux for agricultural land was assessed using the NLOAD model and for forested areas the flux was defined using a regression

model based on concentrations observed in the uppermost groundwater. Finally, the nitrate leaching flux in urban areas was calculated with the help of the Dutch national groundwater quality monitoring network (LMG). The nitrate leaching flux for agricultural areas is dominant over the leaching flux for the other two land-use types. The pesticide leaching flux was simulated by means of the GEOPESTRAS model. The leaching fluxes of nitrate and pesticides are spatially variable as constant values within 500 × 500 m grid cells.

LGM, a numerical model (based on the finite-element method), comprises complex geohydrological system components. LGM uses spatially variable (heterogeneous) data for four aquifers covering the entire surface area of the Netherlands. LGM was applied in five main steps:

a) simulation of groundwater heads and fluxes, using the LGMSAT module for 15 model areas with finite-element base grid of 250 × 250 m. LGMSAT was calibrated on groundwater heads using the inverse-approach based module LGMCAL;
b) simulation of pathlines and travel times (module LGMFLOW);
c) simulation of so-called control breakthrough curves at wells (module LGMCAM);
d) simulation of concentration breakthrough curves at wells (module LGMCAM);
e) aggregation of concentration breakthrough curves at wells for phreatic and semi-confined pumping sites.

A control breakthrough curve is the concentration variation in abstracted groundwater occurring with an instantaneous, spatially constant and time-invariable solute load of 100 concentration units (at zero time) on top of the saturated system. The control breakthrough curves serve as background information for understanding the behaviour of the geohydrological system. They are not used for the assessment of breakthrough concentrations (step d).

LGMCAM uses the concentrations in the leaching fluxes of nitrate and pesticides as input. The concentration breakthrough curves calculated by LGMCAM were based on forward pathline tracking in saturated groundwater. The processes modelled in LGMCAM were advection and full mixing in well screens. In other words, dispersion, diffusion, sorption and decay were disregarded. Solute decay comprises (1) denitrification of nitrate and (2) degradation of pesticides in saturated groundwater. As was anticipated, when compared, the calculated concentrations were found to be mostly higher than the observed ones. From this it is concluded that solute decay (denitrification and degradation of pesticides) and retardation (pesticide sorption) may strongly affect the calculated concentrations. Solute decay and sorption were not taken into account because of lack of input data to define the relevant processes. It is planned to incorporate solute decay and sorption in future studies.

Results

The results for the entire model area are presented: as (1) maps of concentration for the 165 abstraction sites for 2020 and 2050, and (2) concentration bar charts. The bar charts show time-based concentration variation (1950-2050). The bar charts for aggregated (totalized) abstraction rate of 76 phreatic and 89 semi-confined abstraction sites are separate.

Calculated and observed concentration values between 1965 and 1997 were compared for: (1) nitrate and pesticides at a number of selected abstraction sites, and (2) nitrate for an aggregated volume abstracted at phreatic sites. Because the calculated concentrations are (significantly) higher than the observed ones, it is concluded that denitrification and degradation of pesticides in saturated groundwater have a greater impact on the results than the uncertainty in the concentrations of nitrate and pesticides in the leaching flux. Therefore, the modelled breakthrough concentrations can be concluded to be a worst-case outcome.

The concentration in the phreatic systems increases more rapidly (deterioration of groundwater quality) than the concentration in semi-confined systems, due to the faster geohydrological response of the former. Conversely, due to the slow response of semi-confined systems, a relatively large portion of groundwater in these systems retains its good quality for a much longer period of time than in phreatic systems. Consequently, once solute leaching into saturated groundwater terminates, the concentrations in semi-confined groundwater will also start decreasing (improvement of groundwater quality) much later than those in phreatic systems and the decrease rate in time will be smaller.

The calculation results for nitrate lead to the conclusion that especially phreatic abstractions have been affected up to now by nutrient application since 1950 and will be affected in the future. The percentage of the abstracted groundwater volume with calculated concentrations higher than the drinking-water standard (50 mg l^{-1} NO_3) is not likely to increase during the 1995-2020 period. Recalling the worst-case nature of the calculation results, about 25% of the total volume abstracted at phreatic sites in 2020 and 2050 will have concentrations higher than the drinking-water standard. According to the calculation results, an important part of groundwater-based drinking-water production in Oost-Gelderland and Noord-Limburg will continue to be threatened in the future by too high nitrate concentrations.

According to the calculation results, an important part of groundwater-based drinking-water production in Drenthe, Overijssel, Oost-Gelderland and western part of Noord-Brabant will be threatened in the future by an increase in pesticide concentrations. It is concluded that about 10% of both phreatic and semi-confined pumping sites will face atrazine concentrations exceeding the drinking-water standard (0.1 $\mu g\ l^{-1}$). From the calculation results, the number of pumping sites where the drinking-water standard is exceeded for bentazone increase relatively fast in the beginning of the 1995-2020 period. The increase continues during this time period and beyond. Finally, the number of phreatic pumping sites where the drinking-water standard is exceeded for 1,2-dichloropropane decrease after the end of the 1995-2020 period. The number of semi-confined pumping sites exceeding the drinking-water standard increase during the 1995-2020 period. It is likely that the number will stabilize after 2020.

Currently, no policy measures exist that would on short term result in a balanced nutrient application, i.e. no leaching of nitrate into saturated groundwater. As a consequence, the nitrate concentrations in abstracted groundwater will decrease (very) slowly in the future.

A special application of LGM is discussed for the model area *achterhoek*, in the eastern part of the country (Oost-Gelderland). The application illustrates the effectiveness of prospective groundwater protection measures to be taken to decrease the future nitrate concentrations in abstracted groundwater. Most abstractions in the *achterhoek* area are phreatic, i.e. with a rapid response to the changes in concentration of the leaching flux. LGM was used to assess the future concentration of nitrate for the case in which the nitrate leaching to saturated groundwater in the entire capture zones is completely terminated in 2000.

Obviously, the most effective manner to ensure that the quality of abstracted groundwater improves sufficiently and rapidly in the future is concluded to be measures leading to a complete termination of solute leaching (nitrate and pesticides) within the entire surface area of the capture zone of groundwater abstraction sites. As the complete termination is an extreme measure, a spatially optimized pattern of reduced solute leaching could be designed, taking into account, among other factors, the effect of solute decay in saturated groundwater. Possible policy measures to achieve a decrease in nitrate concentrations in the leaching flux are: (1) a decrease in the application rate of nutrients (manure and fertilizers), such as determining the fertilizer requirements for a targeted yield; (2) a change in agricultural practices, such as attuning of the timing of fertilizer application to the crop requirements, or (3) a change in land use, such as converting agricultural land into forest.

In order to ensure that leaching is decreased or terminated within the actual capture zone, the delineation of the protection zone boundaries should be carried out by applying a stochastic (Monte Carlo, uncertainty) analysis. Though this method would result in a larger surface area of capture zones than obtained by the deterministic approach used in this study, it would ensure that the entire capture zone is protected. Various methods are already operational in the Netherlands for the reliability assessment of travel-time zones.

Discussion

In this study, the reliability (uncertainty) of the calculated results was not quantified, e.g. by means of sensitivity analysis. The uncertainty in the concentrations is expected to be especially affected by the following factors, arranged in order of importance:
(a) ignoring denitrification (removal of nitrate) in saturated groundwater,
(b) underestimation of the concentration of nitrate leaching fluxes,
(c) uncertainty in the concentration of leaching fluxes of pesticides,
(d) ignoring degradation of pesticides in saturated groundwater,
(e) schematization of time-variable abstraction rates as a constant well rate since 1950,
(f) uncertainty in the groundwater recharge rate (based on the relatively wet year 1988),
(g) uncertainty in the geohydrological parameters (transmissivities, etc.), and
(h) ignoring sorption of pesticides in saturated groundwater.

Of the above-listed factors (a) through (h), the "direction" (bias) of the error is known only for factors (a), (b), (d) and (h). The factor (e) leads to too high concentrations only in the first part of the calculation period but has probably no effect on the concentrations at later times. Ignoring denitrification and degradation of pesticides results in calculated concentrations in abstracted groundwater which are too high. The validation of NLOAD results indicates that the calculated concentrations in the nitrate leaching flux in the agricultural areas are probably lower than the actually occurring concentrations. This would result in calculated nitrate concentration breakthrough values which are too low (b). As very little is known about the magnitude and a possible bias in the uncertainty in the leaching flux of pesticides (c), no conclusions can be drawn with regard to the effect of this uncertainty source on the pesticide concentrations. The effect of the schematization of time-variable abstraction rates as constant values (e), the effect of the overestimation of the groundwater recharge rate (f), the effect of the uncertainty in the geohydrological parameters (g), and the effect of pesticide sorption in saturated groundwater (h) are considered of less importance than items (a) through (d).

Recommendations

Because of the follow-up of this study in the future, recommendations are given which result in an increase in the reliability of the calculated breakthrough concentrations:

i) Incorporation of solute decay, i.e. denitrification and degradation of pesticides in saturated groundwater. Currently, solute decay can be included in LGMCAM as a spatially constant value of the half-lifetime constant. However, the spatial variability of the half-lifetime constant would have to be taken into account and/or a simple multiple-parameter reaction for the decay process introduced. For this purpose, use can be made of conceptual models available in literature.
ii) Development of a calibration (inverse) method to assess: (1) the parameters governing denitrification and degradation of pesticides and (2) the uncertainty in the concentrations in the leaching fluxes.
iii) Extending the modelling procedure in LGMSAT-LGMFLOW-LGMCAM for time-invariant groundwater abstraction rates, instead of the current single value of abstraction rates, assumed as being constant from 1950 onwards.
iv) Using the groundwater recharge rate (LGM input) for an average meteorological year. The current recharge rate is for the year 1988, which results in values about 20% higher than the average.
v) Using a denser finite-element grid in the immediate vicinity of wells.
vi) Improving the spatially variable parameterization of physical and physico-chemical processes governing transport and fate of solutes in saturated groundwater. Use can be made of data from existing national monitoring networks (LMG and LMD).

In addition, it is proposed that a modelling procedure be developed to yield not only the deterministic values of concentration breakthrough curve at abstraction wells, but also the uncertainty bounds of the concentration variation in time. The methodology would be useful for an uncertainty assessment of the effectiveness of the anticipated groundwater protection zones and for the design of groundwater-quality monitoring wells around a well-field to serve as an early warning system.

Keywords
groundwater, saturated zone, multi-aquifer, quasi-three-dimensional flow, groundwater abstractions, solute transport, advection, numerical modelling, simulation, finite-element method, pathlines, travel times, capture zones, nitrate, denitrification, pesticides, degradation, leaching, concentration, breakthrough curves, land use, scenarios.

SAMENVATTING

Het doel van deze studie is inzicht te krijgen in de gevolgen van historische en toekomstige uitspoeling van nitraat en pesticiden op de concentraties in onttrokken grondwater. De bepaling van de toekomstige belasting van de bodem door nitraat en pesticiden is gebaseerd op verschillende (economische) scenario's. De studie is uitgevoerd in het kader van de nationale lange termijn planning voor drink- en industriewatervoorziening. De resultaten kunnen gebruikt worden ten behoeve van:
(1) het verkrijgen van inzicht in de omvang van de problematiek;
(2) het formuleren van mogelijke (beleids)maatregelen ter verbetering van de milieukwaliteit, waaronder die met betrekking tot de kwaliteit van onttrokken grondwater, zijnde een bron voor de bereiding van drinkwater;
(3) de evaluatie van de effectiviteit van (toekomstige) beleidsmaatregelen.

Dit rapport beschrijft de toepassing van versie 2 van het quasi-driedimensionale RIVM grondwatermodel LGM, voor de berekening van stroombanen, verblijftijden en concentratie-doorbraakkrommen bij grondwaterwinningen van waterleidingbedrijven in de zandgebieden van Nederland. Het gemodelleerde gebied bedekt het grootste gedeelte van Nederland. De studie is toegespitst op 165 onttrekkingslocaties, namelijk 76 freatische en 89 semi-spanningslocaties. Onttrekkingen in de buurt van grote rivieren (zgn. oevergrondwater- en oeverinfiltratie-onttrekkingen) zijn buiten beschouwing gelaten. Alhoewel de berekening van concentraties voor een langere periode is uitgevoerd, zijn de concentraties alleen voor de periode 1950-2050 gepresenteerd. De grondwaterstroming is stationair gemodelleerd, gebruik makend van de maximale hoeveelheid te winnen grondwater volgens vergunning in 1988.

Methoden en modellen

Voor de berekening van belasting van de bodem door nitraat en pesticiden is gebruik gemaakt van verschillende landbouwkundige ontwikkelingen, gebaseerd op drie economische scenario's, genoemd *Divided Europe*, *European Coordination* en *Global Competition*. De ontwikkelingen zijn door het RIVM geformuleerd voor de Nationale Milieuverkenning 1997-2020. De "historische" uitspoelingsflux naar het verzadigde grondwater is berekend voor de jaren 1950, 1960, 1970, 1980, 1990 en 1995. De uitspoelingsflux voor de scenario's is berekend voor de jaren 2000 en 2020.

Drie specifieke pesticiden zijn beschouwd, namelijk 1,2-dichloropropaan, atrazin en bentazon. Dit zijn de voorbeelden van bekende pesticiden die in het ruwwater het meest worden gemeten en die een probleem vormen bij de bereiding van drinkwater.

De uitspoeling van nitraat is gesimuleerd door middel van een combinatie van methoden, afhankelijk van het landgebruik. De nitraatuitspoeling in het landbouwgebied is bepaald met behulp van het model NLOAD. De nitraatuitspoeling in bos- en natuurgebieden is bepaald door gebruikmaking van een regressiemodel gebaseerd op concentraties waargenomen in het bovenste grondwater. Tenslotte is de nitraatuitspoeling in het stedelijk gebied berekend met behulp van het Landelijk Grondwaterkwaliteits Meetnet (LGM). De nitraatuitspoeling in het landbouwgebied is dominant over de uitspoeling in de andere twee landgebruikstypen. De uitspoelingsflux van pesticiden is gesimuleerd met behulp van het model GEOPESTRAS. De uitspoelingsfluxen van nitraat en pesticiden zijn ruimtelijk variabel, als constante waarden in cellen van 500 × 500 m.

Het LGM is een numeriek model (gebaseerd op de eindige elementenmethode), dat complexe geohydrologische systeem-componenten bevat. Het maakt gebruik van ruimtelijk variabele (heterogene) gegevens voor vier watervoerende pakketten voor het gehele gebied van Nederland. De toepassing van LGM in deze studie bestond uit vijf hoofdstappen:

a) simulatie van grondwaterpotentialen en fluxen door toepassing van het module LGMSAT voor 15 modelgebieden. De dichtheid van het basisgrid was 250 × 250 m. LGMSAT was gecalibreerd op grondwaterstanden door middel van een inverse techniek (module LGMCAL);
b) simulatie van stroombanen en verblijftijden (module LGMFLOW);
c) simulatie van zgn. controle-doorbraakkrommen op pompstations (module LGMCAM);
d) simulatie van concentratie-doorbraakkrommen op pompstations (module LGMCAM);
e) aggregatie van doorbraakkrommen op pompstations naar landelijke beelden voor freatische en semi-spanningswinningen.

Een controle-doorbraakkromme is de variatie van de concentratie in het onttrokken grondwater als gevolg van een instantane, in ruimte en tijd constante belasting van 100 concentratie-eenheden aan de bovenzijde van het verzadigde systeem. De controle-doorbraakkrommen dienen als achtergrondinformatie voor het verklaren van het gedrag van het geohydrologische systeem. Ze worden niet gebruikt voor de berekening van de concentratie-doorbraakkrommen (stap d).

LGMCAM gebruikt als input de concentraties van de uitspoelingsfluxen van nitraat en pesticiden. De met LGMCAM berekende concentratie-doorbraakkrommen zijn gebaseerd op de voorwaartse particle-tracking in het verzadigde grondwater. De processen die in LGMCAM zijn gemodelleerd zijn advectie en volledige menging in putfilters. Met andere woorden, dispersie, diffusie, sorptie en stofafbraak zijn buiten beschouwing gelaten. Stofafbraak betreft (1) denitrificatie van nitraat en (2) degradatie van pesticiden in het verzadigde grondwater. Zoals werd verwacht, uit vergelijking van waargenomen en berekende concentraties (de berekende concentraties zijn meestal hoger dan de waargenomen waarden) wordt geconcludeerd dat stofafbraak (denitrificatie en degradatie van pesticiden) en retardatie (door sorptie van pesticiden) een belangrijk effect kunnen hebben op de berekende concentraties. Stofafbraak en sorptie waren niet verwerkt vanwege het gebrek aan invoergegevens voor de definitie van relevante processen. Het is gepland om in toekomstige studies aan stofafbraak en sorptie aandacht te geven.

Resultaten

De resultaten voor het gehele modelgebied zijn gepresenteerd (1) als kaarten van concentraties op de 165 pompstations voor 2020 en 2050, en (2) door middel van staafdiagrammen van concentraties. De staafdiagrammen tonen de concentratievariatie in tijd (1950-2050). Afzonderlijke staafdiagrammen zijn gemaakt voor de geagreggeerde (getotaliseerde) onttrekkingshoeveelheid van 76 freatische en 89 semi-spannings pompstations.

De berekende en waargenomen concentraties tussen 1965 en 1997 zijn vergeleken voor: (1) nitraat en pesticiden op een aantal geselecteerde pompstations, en (2) nitraat voor een geaggregeerd volume grondwater onttrokken op freatische pompstations. Omdat de berekende concentraties (aanzienlijk) hoger zijn dan de waargenomen waarden, wordt geconcludeerd dat denitrificatie en degradatie van pesticiden in het verzadigde grondwater een grotere invloed hebben op de resultaten dan de onzekerheid in de concentraties in de uitspoelingsflux van nitraat en pesticiden. Daarom kunnen de gemodelleerde doorbraak-concentraties als een worst-case resultaat worden gezien.

Vanwege hun snellere geohydrologische respons neemt de concentratie in de freatische systemen sneller toe (verslechtering van grondwaterkwaliteit) dan in semi-spanningsystemen. Vanwege hun langzamere respons zal daarentegen het grondwater in semi-spanningsystemen in vergelijking met freatische systemen gedurende een veel langere periode een betere kwaliteit behouden. Het gevolg hiervan is dat als de uitspoeling naar het verzadigde grondwater stopt, de concentratie in semi-spanningssystemen veel later zal gaan afnemen (verbetering van grondwaterkwaliteit) dan in freatische systemen en de afnamesnelheid kleiner zal zijn.

De berekeningsresultaten voor nitraat leiden tot de conclusie dat in het bijzonder de freatische systemen tot nu toe door vermesting zijn beïnvloed en dat dit ook in de toekomst het geval zal zijn. Het percentage van het opgepompte grondwater met concentraties hoger dan de drinkwaternorm (50 mg l^{-1} NO_3) zal gedurende de periode 1995-2020 waarschijnlijk niet toenemen. Onder verwijzing naar de worst-case aard van de berekeningsresultaten blijkt dat in ongeveer 25% van het totale volume onttrokken op freatische locaties, de concentraties in 2020 en 2050 hoger zullen zijn dan de drinkwaternorm. Volgens de berekeningsresultaten zal in Oost-Gelderland en Noord-Limburg een belangrijk deel van drinkwaterproductie uit grondwater ook in de toekomst worden bedreigd door te hoge nitraatconcentraties.

Volgens de berekeningsresultaten zal in Drenthe, Overijssel, Oost-Gelderland en westelijk Noord-Brabant een belangrijk deel van drinkwaterproductie uit grondwater in de toekomst bedreigd worden door een toename van pesticideconcentraties. Geconcludeerd wordt dat ongeveer 10% van zowel freatische als semi-spanningswinningen te maken zullen krijgen met atrazinconcentraties die de drinkwaternorm zullen overschrijden (0,1 µg l^{-1}). Uit de berekeningsresultaten volgt dat het aantal pompstations waar de drinkwaternorm voor bentazon wordt overschreden in het begin van de periode 1995-2020 relatief snel zal toenemen. De toename zal gedurende de rest van de periode en daarna verder doorzetten. Het aantal freatische winningen waar de drinkwaternorm voor 1,2-dichloorpropaan zal worden overschreden zal afnemen na het einde van de periode 1995-2020. Het aantal semi-spanningslocaties met overschrijding van de drinkwaternorm voor deze stof zal gedurende de periode 1995-2020 toenemen. Het aantal zal zich na het jaar 2020 waarschijnlijk stabiliseren.

Op dit moment bestaan er geen beleidsmaatregelen die op korte termijn zullen leiden tot zodanige evenwichtige toepassing van nutriënten dat geen nitraatuitspoeling naar het verzadigde grondwater plaats heeft. Het gevolg hiervan is dat nitraatconcentraties in het onttrokken grondwater in de toekomst (heel) langzaam zullen blijven dalen.

Een bijzondere toepassing van het LGM is ontwikkeld voor het modelgebied *achterhoek*, in het oosten van het land (Oost-Gelderland). De toepassing illustreert de effectiviteit van mogelijke maatregelen ter bescherming van het grondwater. De meeste onttrekkingen in dit gebied zijn freatisch, dus met een snelle respons op veranderingen in de concentratie van de uitspoelingsflux. Met LGM is berekend wat de toekomstige nitraatconcentratie zal zijn voor het geval de nitraatuitspoeling naar het verzadigde grondwater in het gehele intrekgebied vanaf het jaar 2000 zou worden stopgezet.

Het is evident dat de meest effectieve aanpak die het mogelijk zou maken dat de kwaliteit van het opgepompte grondwater binnen afzienbare termijn voldoende en snel verbetert maatregelen zijn die tot een volledige stopzetting van stofuitspoeling (nitraat en pesticiden) leiden binnen het volledige intrekgebied van grondwateronttrekkingen. Omdat de volledige stopzetting een extreme maatregel is, zou een ruimtelijk geoptimaliseerd patroon van gereduceerde stofuitspoeling kunnen worden ontworpen. Mogelijke beleidsmaatregelen om een verlaging van nitraatconcentraties in de uitspoelingsflux te bereiken zijn: (1) verlaging van de hoeveelheid toegepaste nutriënten (mest en kunstmest), bijvoorbeeld door bepaling van nutriëntbehoefte voor een gewenste gewasopbrengst; (2) een verandering in landbouwpraktijken, bijvoorbeeld door het aanpassen van toedieningstijdstippen van kunstmest aan de gewasbehoeften, en (3) verandering van het landgebruik, bijvoorbeeld door het veranderen van landbouwgrond in bos.

Om zekerheid te hebben dat de uitspoeling is gereduceerd of gestopt in het (werkelijke) intrekgebied, zou de bepaling van de rand van een beschermingsgebied door middel van een stochastische (Monte Carlo, onzekerheids) analyse moeten worden gedaan. Alhoewel deze methode tot een grotere oppervlakte van beschermingsgebieden zou leiden dan de in deze studie gebruikte deterministische methode, het zou een bescherming van het gehele intrekgebied zekerstellen. Voor het bepalen van de betrouwbaarheid van verblijftijden zijn in Nederland al diverse operationele methoden beschikbaar.

Discussie

In deze studie was de betrouwbaarheid (onzekerheid) in de berekeningsresultaten niet gekwantificeerd, bij voorbeeld door middel van een gevoeligheidsanalyse. Verwacht wordt dat de onzekerheid in de concentraties in het bijzonder beïnvloed wordt door de volgende factoren, gerangschikt in volgorde van betekenis:

a) buiten beschouwing laten van denitrificatie (omzetting van nitraat) in het verzadigde grondwater,
b) onderschatting van de concentratie van de nitraatuitspoelingsflux;
c) onzekerheid in de concentratie van de pesticidenuitspoelingsflux;
d) buiten beschouwing laten van degradatie van pesticiden in het verzadigde grondwater;
e) schematisering van in tijd variërende grondwateronttrekkingen als een constant debiet vanaf 1950;
f) onzekerheid in de grondwateraanvullingsflux (gebaseerd op het relatief natte jaar 1988);
g) onzekerheid in de geohydrologische parameters (doorlaatvermogen, etc.);
h) buiten beschouwing laten van sorptie van pesticiden in het verzadigde grondwater.

Van voornoemde factoren (a) t/m (h) is alleen voor factoren (a), (b), (d) en (h) de "richting" van de fout bekend. De factor (e) leidt tot te hoge concentraties alleen in het eerste gedeelte van de berekeningsperiode, maar heeft waarschijnlijk geen effect op de concentraties op latere tijdstippen. Het buiten beschouwing laten van denitrificatie en degradatie van pesticiden resulteert in berekende concentraties in het onttrokken grondwater die te hoog zijn. Uit validatie van de NLOAD-resultaten blijkt dat de voor landbouwgebieden berekende concentraties van de nitraatuitspoelingsflux waarschijnlijk lager zijn dan de werkelijk optredende waarden. Dit zou op zijn beurt resulteren in berekende concentratiewaarden van het ruwwater die te laag zijn (b). Omdat weinig bekend is over de grootte en de mogelijke richting van de fout in de onzekerheid van de pesticidenuitspoelingsflux (c), kunnen geen conclusies worden getrokken ten aanzien van het effect van deze bron van onzekerheid op de pesticiden-concentraties. Het effect van de schematisering van in tijd varierende grondwateronttrekkingen door constante waarden (e), het effect van overschatting van de grondwateraanvullingsflux (f), het effect van de onzekerheid in de geohydrologische parameters (g), en het effect van sorptie van pesticiden in het verzadigde grondwater (h) worden geacht minder invloed te hebben op de berekeningsresultaten dan punten (a) t/m (d).

Aanbevelingen

Mede omdat deze studie nog een vervolg heeft in de komende jaren worden aanbevelingen gedaan waardoor onder andere de betrouwbaarheid van de berekende concentratie-doorbraakkrommen toeneemt:

i) In rekening brengen van stofafbraak, dat wil zeggen denitrificatie en degradatie van pesticiden in het verzadigde grondwater. Stofabraak kan op dit moment in LGMCAM worden verwerkt door middel van een ruimtelijk constante halfwaardetijd. Het zou echter nodig zijn om met een ruimtelijk variabele halfwaardetijd rekening te houden en/of het afbraakprocess door middel van een simpele multiple-parameter reactie te beschrijven. Hierbij kan gebruik worden gemaakt van in literatuur beschikbare conceptuele beschrijvingen.

ii) Ontwikkeling van een calibratie (inverse) methode ten behoeve van de bepaling van: (1) de parameters die invloed hebben op denitrificatie en degradatie van pesticiden en (2) de onzekerheid in de concentraties van de uitspoelingsfluxen.

iii) Aanpassing van de modelleringsmethode LGMSAT-LGMFLOW-LGMCAM om rekening te kunnen houden met in tijd variërende grondwateronttrekkingen. Op dit moment wordt gewerkt met constante waarden van grondwateronttrekking vanaf 1950.

iv) Het gebruiken van grondwateraanvullingsflux (invoer voor LGM) voor een gemiddeld meteorologisch jaar. Op dit moment is de grondwateraanvulling op het jaar 1988 gebaseerd, wat tot waarden leidt die ca. 20% hoger zijn dan de gemiddelde grondwateraanvullingsflux.

v) Het gebruiken van een dichter eindige elementen grid in de naaste omgeving van onttrekkingslocaties.
vi) Het verbeteren van ruimtelijk variabele parameterisatie van fysische en fysisch-chemische processen die bepalend zijn voor het transport en gedrag van opgeloste stoffen in verzadigd grondwater. Hierbij kan gebruik worden gemaakt van gegevens uit bestaande nationale meetnetten (LMG en LMD).

Verder wordt aanbevolen dat een modelleringsmethode wordt ontwikkeld die niet alleen deterministische waarden van concentratie-doorbraakkrommen op pompstations produceert, maar ook de onzekerheidsgrenzen van de concentratievariatie in tijd. De methode zou nuttig zijn voor de bepaling van de betrouwbaarheid van de effectiviteit van de geplande grondwaterbeschermingszones en voor het ontwerpen van grondwaterkwaliteitsmeetnetten rondom winningslocaties (ten behoeve van de kwaliteitsbewaking).

Trefwoorden
grondwater, verzadigde zone, meer-lagen-aquifer, quasi-driedimensionale stroming, grondwateronttrekkingen, stoftransport, advectie, numerieke modellering, simulatie, eindige elementen methode, stroombanen, verblijftijden, intrekgebieden, nitraat, denitrificatie, pesticiden, degradatie, uitspoeling, concentratie, doorbraakkrommen, landgebruik, scenario's.

1. INTRODUCTION

Simulation models for groundwater quantity and quality are essential tools for the long-term planning of the drinking-water and industrial water supply. For this purpose, at the National Institute of Public Health and the Environment (RIVM) the Netherlands Groundwater Model (LGM-Landelijk Grondwater Model) is being used.

The LGM, version 1, has previously been documented by Pastoors (1992) and Kovar *et al.* (1992). Version 2 of the LGM also contains modules for simulation of pathlines, travel times and solute breakthrough curves at groundwater abstraction wells. The evaluation of the merits of the LGM modules (version 2) carried out by Kovar *et al.* (1996), served as basic information for the current study.

Agriculture is the dominant source of nitrate and pesticides in groundwater. Although the environmental hazard - including human-health aspects - associated with the occurrence of these solutes is not well-defined in all cases, it is generally accepted that leaching of these solutes from agricultural fields to the groundwater should be minimized. An important reason for the concern about groundwater contamination by nitrate and pesticides in the Netherlands is groundwater's important function as a source (about two-third's) of drinking-water.

This report documents results of applying LGM (version 2), to groundwater abstraction sites in a major part of the Netherlands. The aim of this study is to assess the future changes in nitrate and pesticide concentrations in abstracted groundwater. Three specific pesticides were considered, namely 1,2-dichloropropane, atrazine and bentazone. These solutes are the best-known examples of pesticides that pose a problem for drinking-water production.

The results of this study have been subsequently used for the RIVM's Environmental Outlook 1997-2020 (RIVM, 1997).

Use has been made of one scenario for generating the leaching flux of nitrate and three scenarios for generating the leaching flux of pesticides into saturated groundwater. The leaching flux for the scenarios was calculated for the years 2000 and 2020. The three economy-based scenarios were developed by the RIVM for the Environmental Outlook 1997-2020 (RIVM, 1997). The three scenarios are referred to as *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*).

The following LGM modules were used in this study:

- LGMGRID, for generation of finite-element grid;
- LGMSAT (and LGMCAL), for simulation of groundwater heads and fluxes in a quasi-three-dimensional, multi-aquifer system;
- LGMFLOW, for calculation of pathlines and travel times;
- LGMCAM, for stream-tube-based calculation of solute breakthrough curves at abstraction wells, using forward particle tracking (by LGMFLOW) from so-called top-grid starting-points. LGMCAM is based on advection and full mixing in well screens. Solute decay is in principle possible but was disregarded in this study. Solute decay could be denitrification of nitrate or degradation of pesticides. Solute decay was disregarded because of lack of input data on a national scale. Solute decay is planned for incorporation in future studies.

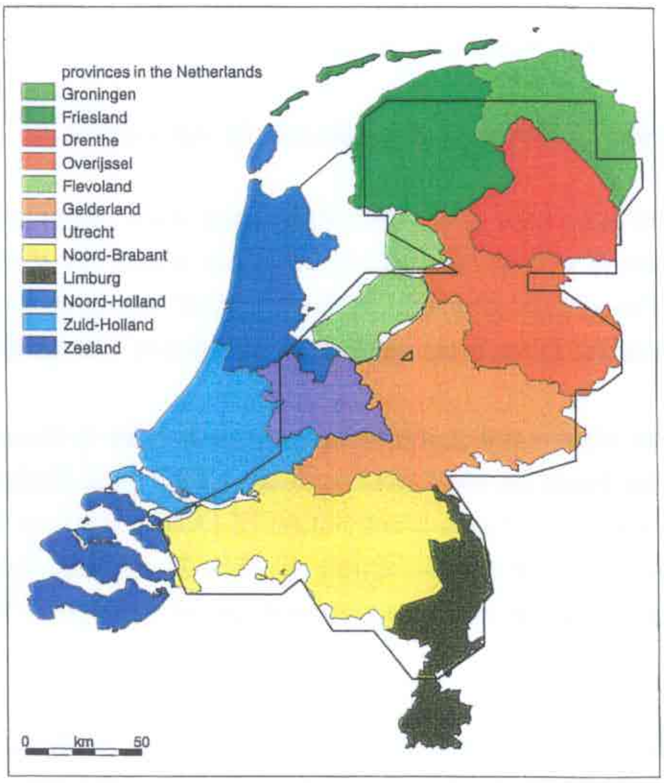


Figure 1.1 *Modelled area and the 12 provinces in the Netherlands. The bold black line is the boundary of the modelled area.*

The LGM represents a "combination" of databases, the Geographic Information System (ARC/INFO) and simulation modules. ARC/INFO is used for storage of spatially related data, a major part of pre- and post-processing and for presentation of model results. LGM is based on the numerical technique of finite elements. The elements are triangles and quadrilaterals. The grid can be locally refined, e.g. within a well capture-zone or in the vicinity of rivers. The saturated multi-aquifer geohydrological system consists of four aquifers separated by aquitards. In LGMSAT, the system is assumed to be quasi-three-dimensional, i.e. two-dimensional horizontal flow in aquifers and vertical flow in aquitards. Though the LGM-programs can also handle transient flow conditions, steady-state flow could be used in this study. LGM was applied to the entire surface area of the Netherlands, with the exception of the western part of the country - a strip of about 60 km along the coast - and the southernmost part of the province of Limburg. The study area also excluded the provinces Noord-Holland, Zuid-Holland and Zeeland. Most of the study area consists of sandy soils. The modelled area is depicted in Figure 1.1.

The calculation of nitrate and pesticide concentrations was carried out for 165 abstraction sites, namely for 76 phreatic and 89 semi-confined abstraction sites. The abstractions in the vicinity of large rivers (river system) were not considered.

Background information on potential pollution of groundwater by agriculture

Results show that the total capture zone area of the 165 abstraction sites amounted to 1703 km^2. A capture zone of an abstraction well is the surface area from where the abstraction well "collects" its groundwater. The source of water can be "on land" by recharge, or from ditches, rivers and lakes by infiltration through the surface-water bottom into groundwater.

The extent of potential leaching of nitrate and pesticides into groundwater is shown in Tables 1.1 and 1.2. Both tables are based on the LGN2 land-use map of the Netherlands (Noordman *et al.*, 1997). LGN2 was created as a combination of satellite images from 1990, 1992 and 1994. The original land-use map is stored on 25 × 25 m cells. Based on the original map, RIVM has prepared a map of dominant land use within 500 × 500 m cells.

It should be noted that the LGN2 is the most recent land-use map of the Netherlands. Because it was not available during this study, previous land-use maps were used, such as the LGN1 (Chapter 10) (Thunissen *et al.*, 1992). The differences between LGN2 and LGN1 are not significant for their purpose in Tables 1.1 and 1.2.

Table 1.1 shows the land use within the 165 capture zones. The agricultural area covers 57.1% of the capture zone area. For comparison, the agricultural area in the entire Netherlands in 1989 amounted to 49% (LEI-CBS, 1995, Table 21-d). The agricultural area forms a significant part of the total capture zone area, representing a potential threat to groundwater quality due to application of nutrients (nitrate, etc.) and pesticides under current agricultural practices. However, other land-use types can also contribute to the deterioration of groundwater quality. Examples are natural areas (forest and heather, etc.) where locally high-nitrate leaching occurs due to nitrate deposition by air and in urban areas where nitrate leaching originates from sewage systems.

Table 1.1 *LGN2-based land use within total capture zone area (1703 km^2).*

Land use	Area (km^2)	Area (%)
Agriculture	973	57.1
Natural (forest/heather, etc.)	450	26.4
Urban area	152	8.9
Other (lakes, rivers, etc.)	128	7.6

Table 1.2 provides a further insight into the potential impact of agriculture on groundwater quality within the total capture zone area. The table indicates that on 88 km^2 crops are used that in principle imply, under the current agricultural practices, the use of atrazine (maize) and bentazone (maize, seed grass, vegetables). 1,2-Dichloropropane is potentially used on 45 km^2 (flower bulbs, potatoes, sugar beet).

Table 1.2 *Agricultural land use (LGN2) as potential solute source within the total capture zone area (1703 km^2) as dominant land use within 500 × 500 m.*

Agricultural land use with potential application of	Area (km^2)
Atrazine/bentazone	88
1,2-Dichloropropane	45
Nitrate (grass)	693
Nitrate (other land)	166

The land-use map aggregation from 25 × 25 m to 500 × 500 m causes the level of resolution to decrease. This has consequences for the accuracy of assessment of land-use area for relatively small parcels. Small parcels are often used for growing maize (use of atrazine and bentazone). A sensitivity study (F.W. van Gaalen, RIVM, personal communication) has indicated that when using the original land-use map, based on 25 × 25 m cells, the surface area of the land use, with application of atrazine/bentazone, could be up to 40 km^2 larger than the surface area with dominant land use derived for 500 × 500 m cells (i.e. 128 km^2 instead of 88 km^2). However, as the map resolution has an effect on only small parcels (atrazine/bentazone), the accuracy of data in Table 1.2 is considered sufficient to provide an insight into the areal extent of the potential leaching sources. Obviously, a certain type of land use implies in general a combination of various leaching fluxes, such as atrazine and nitrate for maize. In other words, the land-use areas listed in Table 1.2 partially "overlap".

Finally, Table 1.3 gives an overview of the spatial distribution of agricultural use in the Netherlands, presented for the 12 provinces (see also Figure 1.1) and is based on LEI-CBS (1995, Table 21-d). The information is for the year 1994. The agricultural land is defined as total of arable land, grass, horticulture and fallow land. As provinces also contain surface water (lakes, etc.), the portion of agricultural land in some provinces is relatively small, e.g. 40% in Zeeland. The current study included most of the area of the nine provinces listed; it does not include the provinces Noord-Holland, Zuid-Holland and Zeeland. The right-most column gives an indication of the percentage of the provincial area that is a potential source of leaching of nitrate and pesticides into groundwater.

Table 1.3 *Outline of agricultural land use (grass and agricultural land) in the 12 provinces in the Netherlands.*

Province	Total (km^2)	Grass (km^2)	Grass/Total (%)	Agr.land (km^2)	Agr.land/Total (%)
Groningen	2968	591	20	1651	56
Friesland	5315	1931	36	2251	42
Drenthe	2680	693	26	1603	60
Overijssel	3420	1513	44	2111	62
Flevoland	1637	127	8	931	57
Gelderland	5143	1755	34	2547	50
Utrecht	1434	620	43	707	49
Noord-Brabant	5083	1177	23	2718	53
Limburg	2209	383	17	1090	49
Noord-Holland	3518	761	22	1372	39
Zuid-Holland	3334	817	25	1484	45
Zeeland	3117	137	4	1247	40
	39,858	10,505		19,712	

The concentration variation of nitrate and pesticides in abstracted groundwater was calculated starting from 1950. The primary reason for selecting 1950 as starting time was that an intensive application of nitrate had started after the Second World War, the start of the application of the three pesticides being somewhat later.

The accuracy of the calculated concentrations depends strongly on the accuracy (reliability) of the fluxes of nitrate and pesticides leaching into shallow groundwater. The amount of solute leaching depends, for example, on (1) the amount applied to the soil, (2) the percentage of agricultural land and (3) the specific crop.

Background information on groundwater abstractions

The concentration variation in abstracted groundwater depends not only on the concentration of the solute leaching into saturated groundwater, but also on the direction and magnitude of flow in saturated groundwater. The flow in the saturated system depends on geohydrological parameters and boundary conditions. An important boundary condition is formed by groundwater abstraction rates.

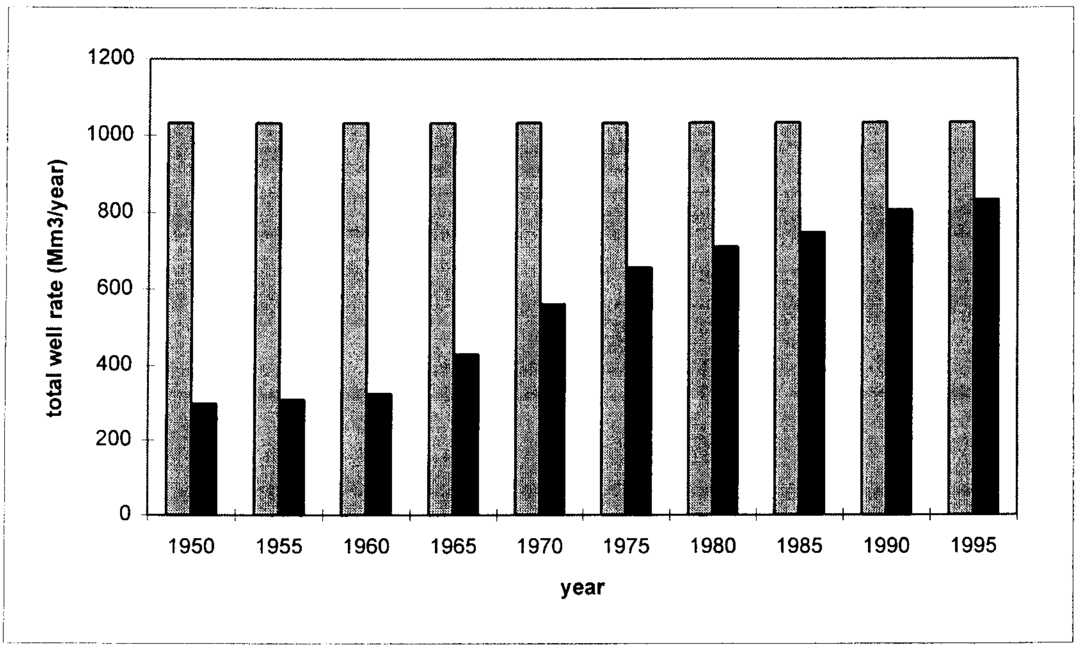


Figure 1.2 *Historical development of groundwater abstractions in the Netherlands (grey: 1988 maximum-permitted abstraction rate; black: actual total abstraction rate).*

Use is made of maximum-permitted abstraction rates for the year 1988. As steady-state groundwater flow is simulated, the applied abstraction rates are assumed to be constant in time from 1950. However, in reality, most abstractions came into being after 1950 and have gradually increased in time. Figure 1.2 shows the variation of the actual totalized groundwater abstraction rates for all abstractions in the Netherlands between 1950 and 1995. The figure was compiled at RIVM using the water-supply company statistics for the Netherlands, published yearly by VEWIN. The total volume of groundwater that can be abstracted according to the maximum-permitted abstraction rates for 1988 is depicted as a constant value. The maximum-permitted abstraction rates for 1988 were used as input for this study; they obviously differred per abstraction site. The rates are higher than the actual volume of groundwater abstracted in 1995. The consequences of the discrepancy between the assumed time-invariant maximum-permitted abstraction rates for 1988 - from 1950 onwards - and the actual time-variable rates are discussed in section 2.3.

Structure of this report

The chapters in this report are broken down as follows:

- Chapter 2 (Modelling Groundwater Potential) discusses the application of LGM in solving the groundwater potential problem, i.e. for the simulation of groundwater heads and fluxes in a saturated multi-aquifer system. Specifically, it describes the application of the module LGMSAT and the results achieved. Use is made of maximum-permitted abstraction rates for the year 1988. The results will be used as input for Chapter 7.

- Chapter 3 (Scenarios for Solute Leaching into Groundwater) gives as background information for Chapters 4 and 5 the (economic) scenarios used for the assessment of future leaching of nitrate and pesticides into saturated groundwater. Scenarios used are *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*).

- Chapter 4 (Modelling Nitrate Leaching) documents the assessment of the nitrate leaching flux (concentration) into saturated groundwater, both for the historical period (1950 to 1995) and for the scenario period (2000 and 2020). The leaching flux will be used for calculation of the nitrate concentration in abstracted groundwater (Chapter 7).

- Chapter 5 (Modelling Pesticide Leaching) deals with the assessment of the pesticide leaching flux (concentration) into saturated groundwater, both for the historical period (1950 to 1995) and for the scenario period (2000 and 2020). The leaching flux is calculated by means of the GEOPESTRAS model. The leaching flux will be used for calculation of pesticide concentration in abstracted groundwater (Chapter 7).

- Chapter 6 (Modelling Pathlines and Travel Times) describes the modelling of pathlines and travel times in saturated groundwater with the module LGMFLOW, as required for the assessment of control breakthrough curves (Appendix E) and concentration variation in abstracted groundwater (Chapter 7).

- Chapter 7 (Modelling Concentrations at Abstraction Wells) discusses the application of the module LGMCAM for the simulation of concentration breakthrough curves of nitrate and pesticides in the course of 1000 years (1950-2950) at 165 abstraction locations (76 phreatic, 89 semi-confined). Concentrations are calculated for the three selected scenarios. Calculated and observed concentrations are compared for a number of selected abstraction sites.

- Chapter 8 (Concentration Results Aggregated for Model Area) presents the concentration results from Chapter 7 by means of bar charts. The concentration bar charts show concentration variation in time (1950-2050). The bar charts are prepared separately for an aggregated abstraction rate of 76 phreatic and 89 semi-confined abstraction sites. The total well rate of 76 phreatic and 89 semi-confined abstractions is 282.10×10^6 and 457.66×10^6 m^3 $year^{-1}$, respectively.

- Chapter 9 (Effect of Termination of Nitrate Leaching) concerns a specific application of LGM. LGM was used for the model area *achterhoek* to calculate the future concentration of nitrate for the case where nitrate leaching to saturated groundwater in the entire capture zones would be completely terminated in the year 2000.

- Chapter 10 (Land-use Maps) gives an overview of land-use maps used.

- Chapter 11 deals with Groundwater Protection Measures.

- Chapter 12 lists Conclusions and Recommendations.

2. MODELLING GROUNDWATER POTENTIAL

2.1 Introduction

This chapter discusses how LGM is used to find a solution to the groundwater potential problem, i.e. for the simulation of groundwater heads and fluxes in a saturated multi-aquifer system. Specifically, it describes the application of the module LGMSAT and its results, which will be used as input for modelling concentration breakthrough curves at abstraction wells (Chapter 7).

LGMGRID (this chapter)
generation of finite-element grid

↓

LGMSAT (and LGMCAL) (this chapter)
simulation of groundwater heads
and fluxes in multi-aquifer system

↓

LGMFLOW (Chapter 6)
simulation of pathlines and travel times
in multi-aquifer system

→ **LGMCAM** (Appendix E and Chapter 7)
stream-tube-based calculation of concentration breakthrough curves

→ **LGMCAD** (not used in this study)
random-walk-based calculation of concentration distributions and breakthrough curves

Figure 2.1 *Interrelationships of LGM modules.*

The functionality and interrelationships between the various computer programs (modules) within LGM are overviewed, with special attention given to the functionality of the module LGMSAT for modelling groundwater flow. Figure 2.1 shows the sequence in which the LGM modules have to be used.

Version 2 of the RIVM Netherlands Groundwater Model (LGM) consists of the modules LGMGRID, LGMSAT, LGMCAL, LGMFLOW, LGMCAM and LGMCAD. The abbreviation LGM stands for Dutch "Landelijk Grondwater Model". The database for LGM will eventually cover the entire area of the Netherlands. The storage of spatially related data, most of post-processing and presentation of model results, is done using the Geographic Information System (ARC/INFO). The concept of LGM is described by Pastoors (1992) and Kovar *et al.* (1992).

LGM is based on the numerical technique of finite elements. The elements are triangles and quadrilaterals. The combination of triangles and quadrilaterals in one grid is possible. The model area is a polygon of arbitrary shape (convex or concave). Strictly speaking, only LGMGRID, LGMSAT, LGMCAL and LGMFLOW actually use the finite-element method grid. The programs LGMCAM and LGMCAD are post-processors of LGMFLOW. LGMCAM does not explicitly use the finite-element method. LGMCAD uses the finite-element grid for interpolation of groundwater velocities.

Though the LGM modules can also handle transient conditions, the groundwater flow in this study was assumed steady-state. However, evidently, the solute transport is transient, i.e. concentrations in abstracted groundwater vary in time.

The sequence of module application within LGM is as follows. First, the module LGMGRID is used to generate the finite-element grid. LGMGRID distinguishes two main steps: (a) generation of the so-called base grid (with user-specified grid density), and (b) local adaptions of this base grid (e.g. local refinements). LGMGRID also carries out the discretization of the spatially variable data for the nodes of the finite-element grid. Second, the packages LGMSAT, LGMCAL and LGMFLOW are used to perform the actual modelling analysis of the geohydrological system:

- LGMSAT module (previously the programme package AQ-**EP**, Kovar & Leijnse, 1989a): calculation of groundwater heads in aquifers and fluxes across aquitards, in a saturated multi-aquifer system, i.e. the quantitative aspects. This module is discussed by Pastoors (1992) and Kovar *et al.* (1992). The output of LGMSAT are groundwater heads in aquifers, the flux across the aquitards, the flux between the top aquifer and rivers, and the flux between the top aquifer and the top system (dense system of ditches).
- LGMCAL module (Leijnse & Pastoors, 1996): automated calibration (inverse modelling) of parameters, by minimizing of objective function (total of squared residuals between observed and calculated heads, and co-weighting the best estimates of the parameter values).

- Module LGMFLOW (previously the programme package AQ-**EF**, Kovar & Leijnse, 1989b): calculation of pathlines and travel times in the saturated multi-aquifer system.

Finally, once LGMFLOW has been applied, it is possible to use LGMCAM or LGMCAD to calculate solute breakthrough curves at abstraction wells:
- LGMCAM module: stream-tube-based calculation of concentration, applying forward particle tracking from so-called top-grid starting-points. The functionality of LGMCAM is discussed in Kovar *et al.* (1996, section 2.1.1). LGMCAM is based on advection and solute decay of particles along pathlines. Only advection is used in this study.
- LGMCAD module: random-walk-based calculation of concentration, applying the groundwater velocity field. LGMCAD is based on advection, dispersion, solute decay and sorption. The particle-tracking routine in LGMCAD is based on the Random Walk Method (Uffink, 1990; Kinzelbach & Uffink, 1991). The particles in LGMCAD represent a mass of the solute. The module LGMCAD was <u>not</u> used in this study.

2.2 Geohydrological System

Figure 2.2 depicts schematically the geohydrological system used in LGM. The saturated multi-aquifer geohydrological system consists of four aquifers separated by aquitards. The flow in the system is assumed to be quasi-three-dimensional.

The groundwater head is specified along the model periphery in each aquifer.

The top aquifer can be replenished by a spatially variable groundwater recharge. The procedure for calculating the groundwater recharge rate is explained in Kovar *et al.* (1992, section 6.5).

The second component affecting the top aquifer is the so-called top-system flux relation. The top-system flux is also spatially variable. The top-system flux relation is discussed in Kovar *et al.* (1992, sections 5.5, 6.7 and 6.8).

The top aquifer is also hydraulically connected with rivers and canals. The aquifer recharge from rivers and canals depends on river-water level, groundwater head and hydraulic resistance of river bottom. Details of the river concept can be found in Kovar *et al.* (1992, section 6.6).

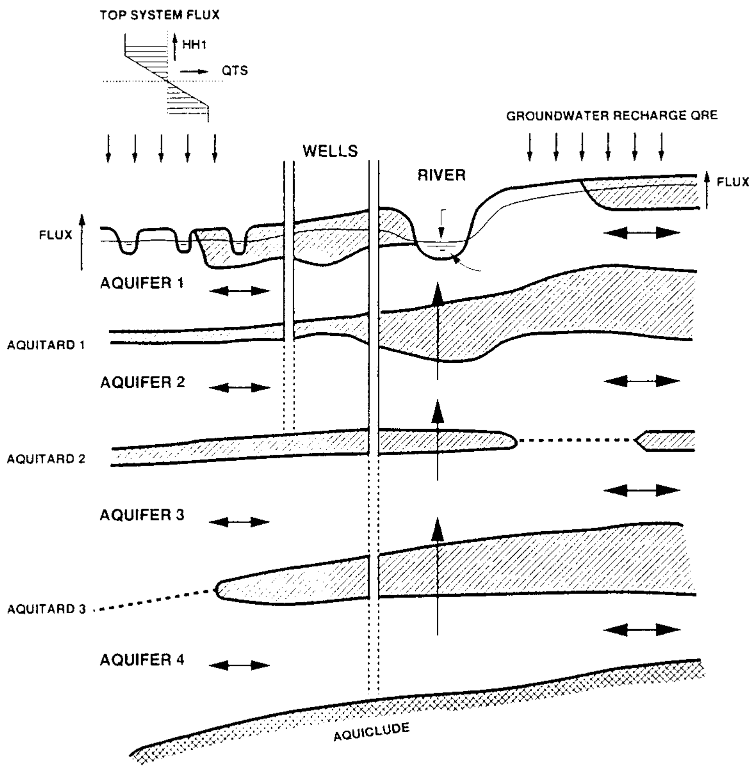


Figure 2.2 *Geohydrological system for LGM.*

The remaining system elements are wells. The well screens are assumed to fully penetrate aquifer thickness. As the actual well screens, in general, cut through a number of layers (aquifers and aquitards), they are transformed into a number of well screens per aquifer, each with its own well rate. The procedure for calculating the model well rates per aquifer is described in Kovar *et al.* (1992, section 6.4 and Chapter 13). The input data for well abstractions used in this study are discussed in section 2.3.

2.3 Well Abstractions

The groundwater abstractions consist of (a) abstractions for drinking-water supply and (b) abstractions for industrial purposes. Figure 2.3 depicts the location of abstractions for drinking-water supply. The blue, green and red colours indicate that the abstraction occurs in phreatic, semi-confined and river systems, respectively. A *phreatic* groundwater system is a system without a confining layer overlying the well screen or layer(s) with relatively low hydraulic resistance. A *semi-confined* groundwater system is a system with a considerable hydraulic resistance of layers overlying the well screen. A *river system* is a system where groundwater abstraction takes place in the vicinity of a large river (induced surface-water recharge).

Use was made of an existing identification system for *phreatic*, *semi-confined* and *river-system* abstractions (Beugelink & Mülschlegel, 1989). This identification system was developed in the framework of a study regarding the well-head protection zones of groundwater abstractions.

The calculated control breakthrough curves (Appendix E) reflect in some cases a type different from that in the current identification system, e.g. a breakthrough curve indicates that an abstraction is phreatic, while that abstraction has been ranked as semi-confined. In this respect, the results of this study could be used in the future for adjusting the selection into categories of *phreatic*, *semi-confined* and *river-system* abstractions.

Labelling of abstraction sites as *phreatic*, *semi-confined* or *river system* has neither influence on the calculations of groundwater flow (this chapter), nor on the calculation of pathlines and travel times (Chapter 6), nor on the calculation of concentration breakthrough curves (Chapter 7). The labelling is only used for presentation of the aggregated results (Chapter 8).

Figure 2.3 pertains to the total area modelled in this study, i.e. the entire surface area of the Netherlands, with the exception of the western part of the country along the coast and southern-most part of the province of Limburg.

The basic and refined models are discussed in sections 2.5 and 2.6. From Figure 2.3 it follows that two abstraction wells will be located in the area covered by the three basic models, but not included in any of the 15 refined models.

It is important to note that the LGM calculations were carried out for a single ideal well location for each abstraction site. In reality, the abstraction takes place in most cases by means of a series of wells spread over a certain area. A maximum distance between the wells within a well field of 500 m or more is not unusual. The ideal well location in LGM is supposed to be at the "centre of gravity" of the real well locations, the well rate in LGM being the total of all well rates.

For each groundwater abstraction site in the Netherlands, the maximum allowable abstraction rate (on a yearly basis) is restricted by a permit. It was decided to use the maximum-permitted abstraction rates, as in effect for the year 1988, because at the time of the study, the abstractions for 1988 were the most recent data available for the entire country. The relevant abstraction rates were drawn from VEWIN (1989), the medium-term planning document of the Dutch association of water-supply companies in the Netherlands.

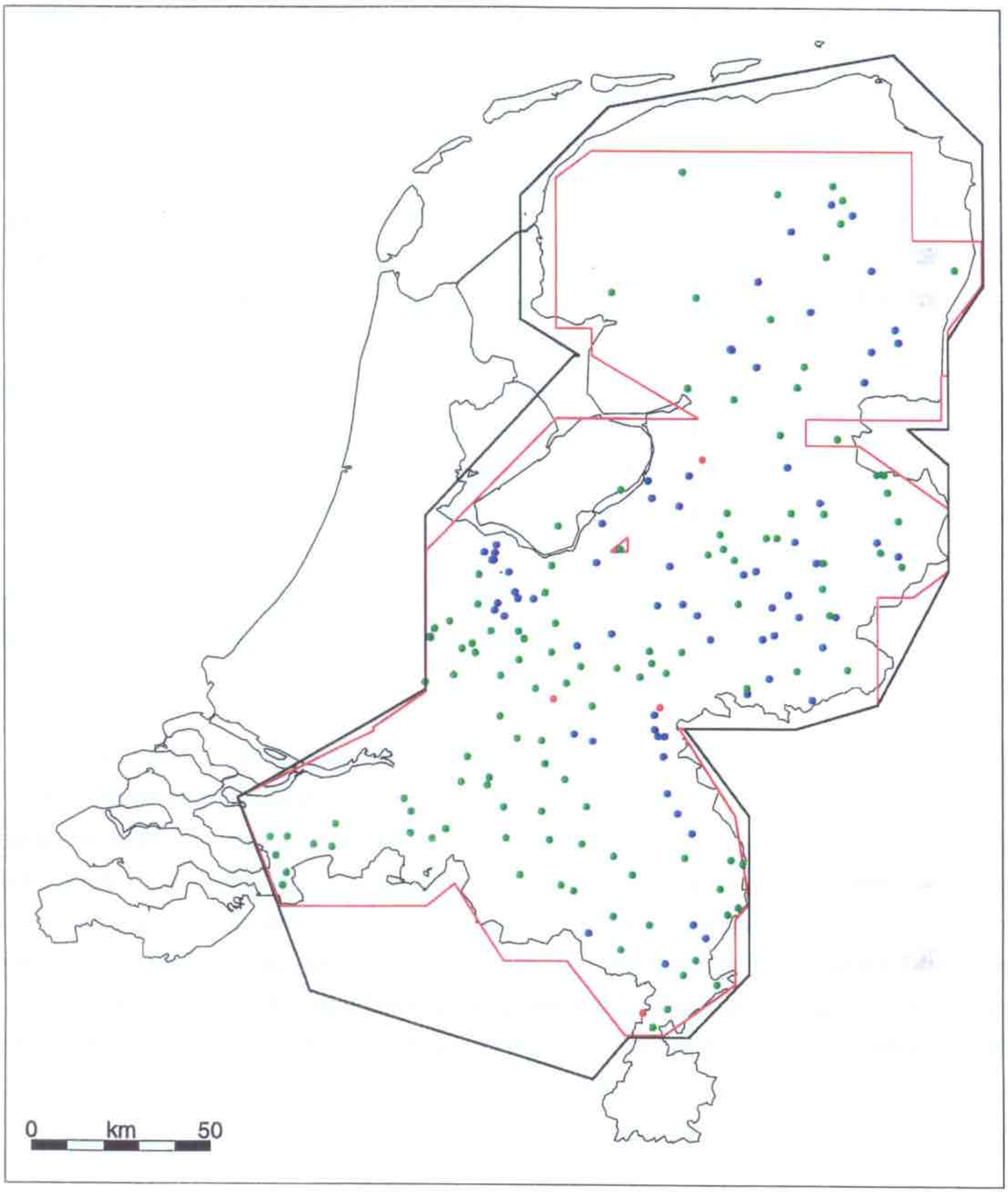


Figure 2.3 *Location of all abstractions for the drinking-water supply (blue:phreatic, green:semi-confined, red:river system), where the solid black line represents the boundary of the joint area of three basic models and the solid red line the boundary of the joint area of 15 refined models.*

The concentration variation of nitrate and pesticides in abstracted groundwater has been calculated starting from 1950. In addition to the leaching fluxes (concentrations) of nitrate and pesticides into groundwater, the second major important factor influencing the concentration variation are groundwater abstractions. As steady-state groundwater flow is simulated, the applied abstraction rates are assumed to be constant in time from 1950. However, in reality, many abstraction sites were put into operation after 1950 (mostly between 1960 and 1970) and/or the abstraction rates have changed - mostly continously increased - with time. This was illustrated in Figure 1.2 (Chapter 1). The abstracted volume (Figure 1.2) concerns all abstractions in the Netherlands, i.e. also outside the area modelled in this study, such as in dunes along the west coast. The total volume of groundwater that can be abstracted according to the 1988 maximum-permitted abstraction rates is 1033×10^6 m^3 per year. The actual volume of groundwater abstracted in 1995 was about 880×10^6 m^3. The difference between the last two volumes has consequences for the calculated concentration variation of nitrate and pesticides in abstracted groundwater. In general, this will lead to (1) an earlier occurrence of a certain concentration level, and (2) a deviation (positive or negative) in the final long-term concentrations:

- The earlier occurrence is caused by faster "depletion" of the "old" resident groundwater (infiltrated before 1950) which is mostly of better quality than current groundwater recharge.
- The deviation is caused by the fact that a higher well rate implies a greater capture zone area and, consequently, a greater effect of solute leaching in that extra portion of the capture zone.

The graph in Figure 1.2 (Chapter 1) shows the total abstracted volume. The actual abstraction "history" is different for each abstraction location. Some abstractions have gradually increased since 1950. At other locations the abstraction rate has been maintained constant or increased stepwise from the onset of abstraction (e.g. 1960) up to now. The discrepancy between the constant 1988 maximum-permitted abstraction rates and the actual temporal variation of abstraction rate per specific site has an effect on the accuracy of the calculated solute concentration in abstracted groundwater. In general, when compared to reality, this would lead to an earlier occurrence of a certain concentration level. Theoretically, the deviation between the 1988 maximum-permitted well rate and the actual well rate also affects the calculated concentrations much later.

Consequently, when interpreting the concentration results (Appendix E and Chapter 7), one should also take into account the difference between the constant 1988 maximum-permitted abstraction rates assumed in this study and the actual well rates. The actual well rates seldom exceed the 1988 maximum-permitted abstraction values.

Appendix A provides information on all abstractions for the drinking-water supply, i.e. phreatic, semi-confined and river system. The information was compiled using the data available at RIVM. In many cases, the data was provided by the Dutch provinces. Appendix A contains, for example, (1) the so-called LGM-number (LGM-no.) (this is the sequence number of an abstraction in the LGM database), and (2) the so-called LAC-number (LAC-no.) (this is an identification codename (number) for an abstraction location). The LAC-numbers were assigned to abstraction locations in 1970s and 1980s by the RIVM Laboratory for Inorganic Chemistry (LAC), throughout the execution of chemical analyses of abstracted groundwater. Table 2.1 summarizes the data on groundwater abstractions, as listed in Appendix A.

Table 2.1 *Summary of basic data on drinking-water abstractions (permit 1988).*

	number	total rate (m^3 $year^{-1}$)
phreatic abstractions	83	295.45×10^6
semi-confined abstractions	109	518.95×10^6
river-system abstractions	4	13.00×10^6

2.4 Groundwater Recharge Rate

In addition to groundwater abstraction, the calculated concentration of abstracted groundwater is also sigificantly affected by the groundwater recharge rate. The groundwater recharge rate is the net flux from the root zone into the saturated groundwater, i.e. the flux across the phreatic water table. Important are both the magnitude and the spatial distribution. The average groundwater recharge rate in sandy soil areas of the Netherlands is about 1.0 mm day^{-1} for grass, 0.9 mm day^{-1} for decidious forest, and 0.8 mm day^{-1} for coniferous forest.

The procedure followed for the calculation of groundwater recharge rate is described in Pastoors (1992, section 4.3). In LGMSAT, the recharge rate is calculated as the difference between precipitation and actual evapotranspiration. The actual evapotranspiration depends highly on the land use. The land-use map applied to calculating groundwater recharge is described in section 9.1.

According to Pastoors (personal communication), the reliability of the assessed groundwater recharge rate is hampered by the following two aspects, listed in order of importance:

Table 2.2 *Statistics of the LGMSAT groundwater recharge rate (mm day^{-1}) and 1961-1990 average groundwater recharge rate for five refined model areas.*

	LGMSAT (nodes)		1961-1990 (500 × 500 m)	
	MV	Var	MV	Var
veluwe	1.13	0.38	0.89	0.17
achterhoek	0.95	0.44	0.84	0.14
twente	1.04	0.41	0.83	0.14
ruhrdalslenk	1.04	0.42	0.76	0.18
eindhoven	1.13	0.35	0.76	0.16

2.5 Basic Finite-Element Models

Three models were developed as a first step in groundwater flow modelling, namely:
- a model for the central-eastern Netherlands (model code *o2*);
- a model for the northern Netherlands (model code *n2*); and
- a model for the southern Netherlands (model code *z2*).

These three models form the basis of the current application of LGM, version 2. The location of the three models is depicted in Figure 2.4.

The finite-element grid is generated by the LGM module LGMGRID. The density of the base grid used for the three basic models was 1 × 1 km. In some models, the grid is locally refined in (user-specified) polygon-shaped areas. Figure 2.5. gives an example of the resulting finite-element grid for the model area for the central-eastern Netherlands (model code *o2*). It can be seen that element boundaries follow the river courses.

Once the finite-element grid is generated and input data is prepared, the module LGMSAT can be used to solve the groundwater potential problem, resulting in groundwater heads in aquifers, fluxes across the aquitards, etc.

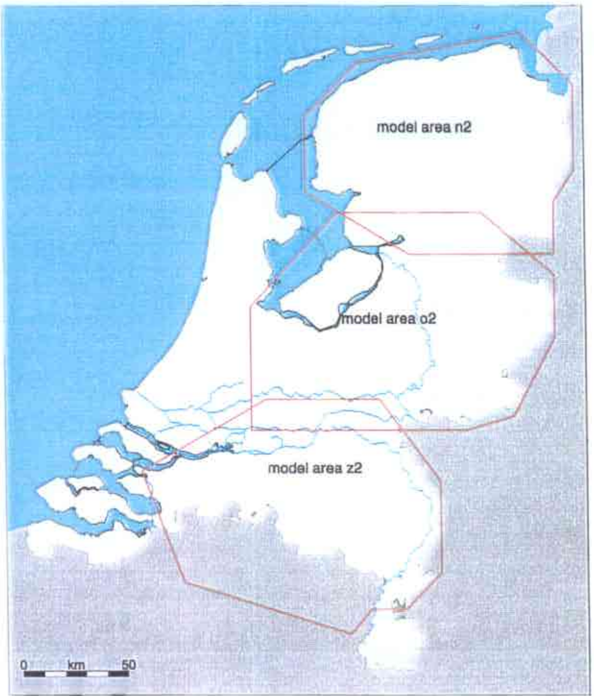


Figure 2.4 *Location of three basic model areas.*

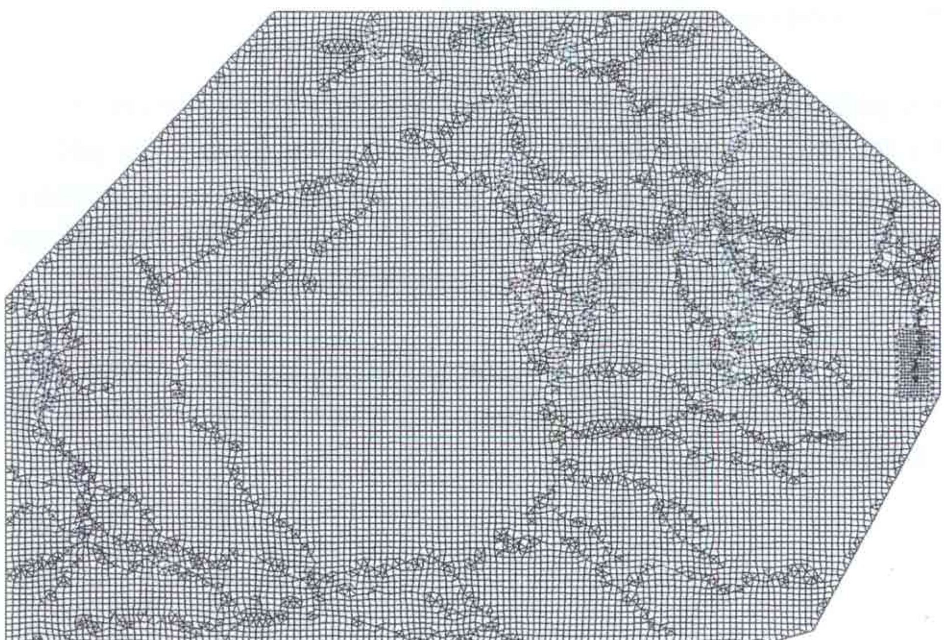


Figure 2.5 *Example of a finite-element grid, for model area o2.*

The three basic model areas (*o2*, *n2* and *z2*) overlap. The reason for this is twofold:

a) to minimize the effect of possible errors in groundwater heads specified along the model boundaries. Once the three models have been run separately, the combined maps of output variables (groundwater heads and fluxes) are produced by ARC/INFO within the conjunctive model area. In the overlapping areas, the output variables are calculated as a weighted average of the variables calculated in two model areas, weighting conforming to the distance of a point to the model boundaries.

b) to make it possible to use the models for the scenario calculations where the effects of scenario changes, e.g. increase of groundwater abstractions, can extend close to a model boundary. Theoretically, a scenario input should be such that its effect does not extend beyond a model boundary, or is only negligible at that distance. By applying overlapping model boundaries, the user has an opportunity to choose for the scenario calculation that model area in which the impact of the scenario at the model boundary is minimal.

The three models have been calibrated by means of the module LGMCAL, using as calibration variables the observed groundwater heads. The calibration was done for the drainage resistance of the top system, the hydraulic resistance of aquitards and the transmissivity of aquifers.

The three basic models were used for generating input boundary conditions (groundwater heads) for the 15 refined finite-element models.

2.6 Refined Finite-Element Models

The final aim of this study is to calculate the concentration variation in time in abstracting wells. Referring to Figure 2.1, the output of LGMSAT (groundwater heads and fluxes) is used for calculating pathlines and travel times (Chapter 6) and breakthrough concentrations (Appendix E and Chapter 7).

In the study carried out by Kovar *et al.* (1996) it has been established that when using LGM for calculating pathlines, travel times and breakthrough curves for phreatic and semi-confined abstractions, one must use the base grid element size of 250 × 250 m in the entire capture zone. Obviously, the grid density of the three basic models (1 × 1 km) is too coarse. In order to meet the requirements set by Kovar *et al.* (1996), 15 so-called *refined finite-element models* have been developed. The density of the base grid used for these refined models was 250 × 250 m.

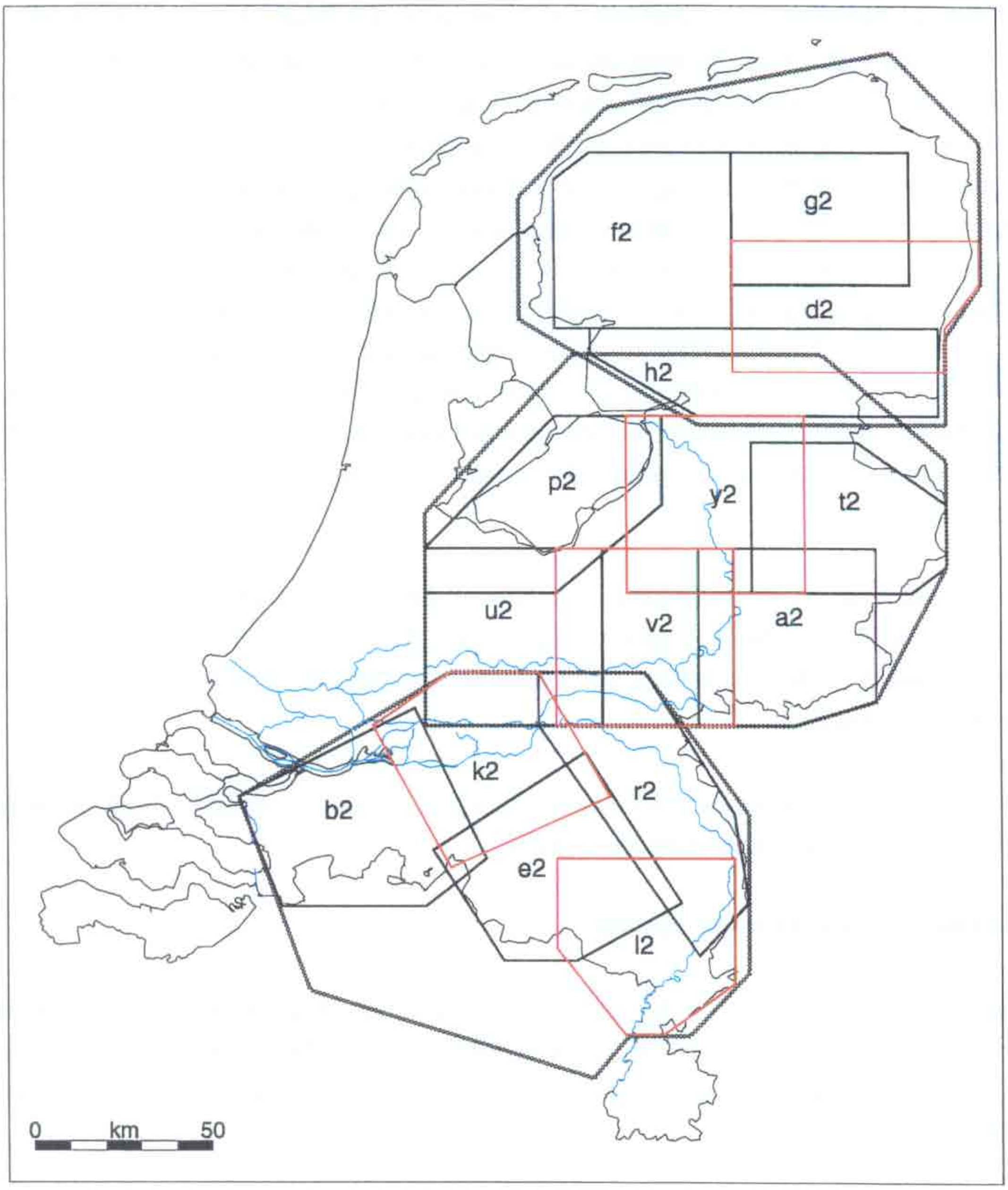


Figure 2.6 *Location of 15 refined model areas, with several model boundaries in red.*

The location of the 15 models is shown in Figure 2.6, wit several model boundaries drawn in red for clarity. The figure also contains the boundary of the three basic models.

The model location was selected such that each refined model is fully located with either of the three basic finite-element models. The following refined models were developed:

- within basic model *o2* (the central-eastern Netherlands):
  - refined model *utrecht* (model code *u2*);
  - refined model *veluwe* (model code *v2*);
  - refined model *achterhoek* (model code *a2*);
  - refined model *polder* (model code *p2*);
  - refined model *yssel* (model code *y2*);
  - refined model *twente* (model code *t2*);
- within basic model *n2* (the northern Netherlands):
  - refined model *hoogeveen* (model code *h2*);
  - refined model *drenthe* (model code *d2*);
  - refined model *groningen* (model code *g2*);
  - refined model *friesland* (model code *f2*);
- within basic model *z2* (the southern Netherlands):
  - refined model *ruhrdalslenk* (model code *r2*);
  - refined model *limburg* (model code *l2*);
  - refined model *brabantwest* (model code *b2*);
  - refined model *kaatsheuvel* (model code *k2*);
  - refined model *eindhoven* (model code *e2*).

With the exception of model *f2*, which does not overlap with other models, the model areas overlap along their boundaries. The considerations for introducing an overlap are analogous to those for the basic finite-element models (section 2.5). Model *f2* has no overlap with other models because no major groundwater abstractions are located on either side of its boundary (Figure 2.3).

It can be seen from Figure 2.6 that the 15 refined models do not fully cover the area of the three basic models. This is especially the case for basic model *n2* (the northern Netherlands) and for basic model *z2* (the southern Netherlands). The fringe area in the north of the model *n2* was not included in a refined model because of negligible effect of groundwater abstractions in that area. The Belgian area in the south of the model *z2* was not included in a refined model because this study focuses only on the territory of the Netherlands.

Unintentionally, while selecting the model boundaries, a small triangular area at the intersection of the models *v2*, *p2* and *y2* was not included in any model. The only consequence is that the abstraction (Speuld) that happens to be located in that area cannot be used for calculation of concentration breakthrough curves.

The refined finite-element models were not calibrated. It was asssumed that the calibration carried out by LGMCAL for the three basic models is sufficient for the purposes of this study.

All further LGM-modelling work carried out in the framework of this study and reported hereafter is based on the 15 refined finite-element models.

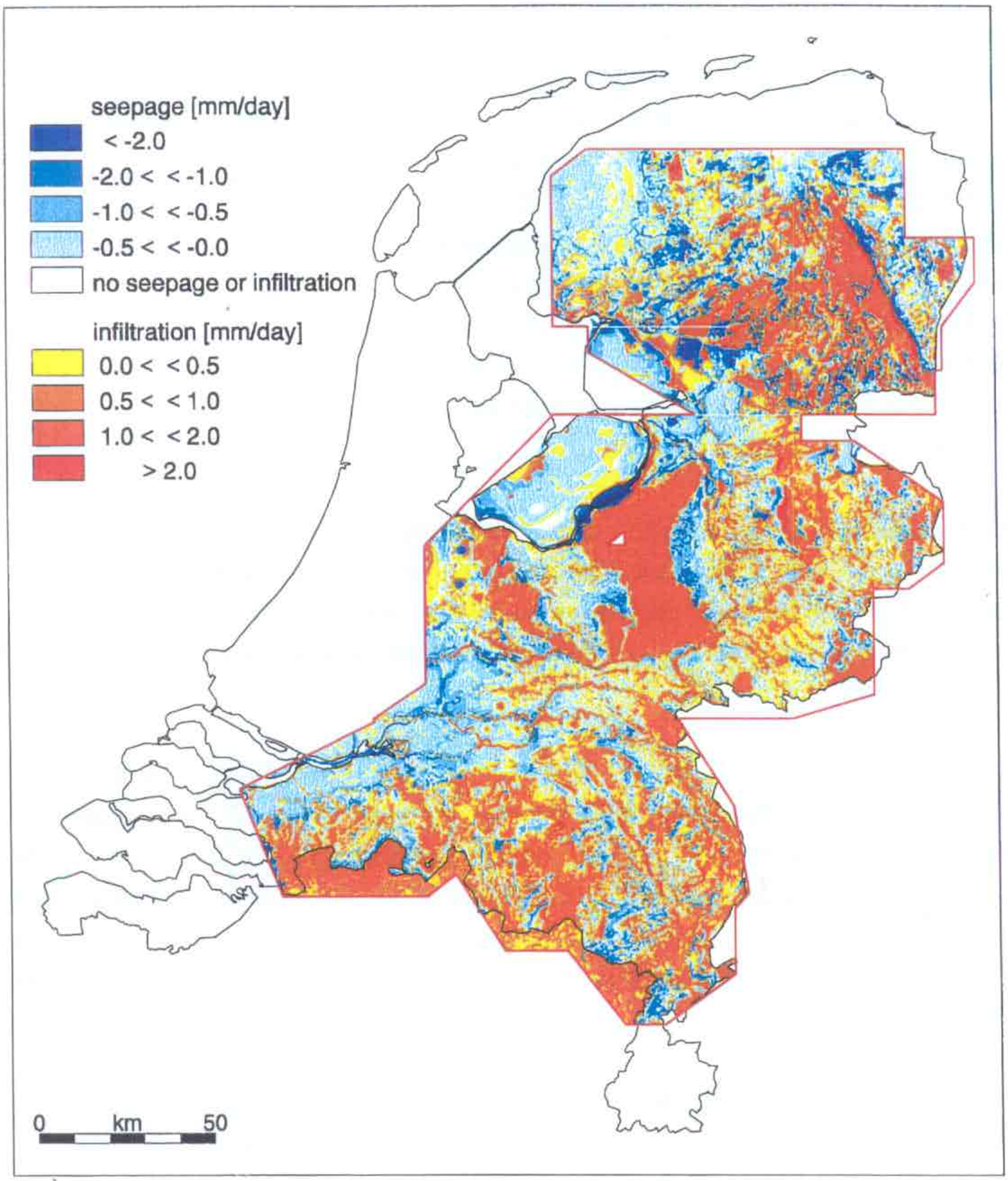


Figure 2.7 *Sum of groundwater recharge rate and top-system flux (mm day^{-1}) presented in cells 250 × 250 m within 15 refined model areas.*

1) the assessment is based on the meteorological data for the year 1988 (precipitation and grass reference evapotranspiration). However, as the year 1988 was relatively wet, it is not representative for the average situation. Therefore, the resulting groundwater recharge rate may locally be up to 20% higher than it would be for an "average" meteorological year. The year 1988 was used for maintaining consistency with groundwater abstraction data, which are also specified for that year;
2) use is made of the meteorological data at the 15 main meteo-stations of KNMI (Royal Netherlands Meteorological Institute). Precipitation and Makking reference (grass) evapotranspiration is available for each main meteo-station. In addition to the 15 main meteo-stations, there is a higher-resolution observation network for precipitation observations. From small-scale meteo observations it is known that precipitation shows great variability within short distances. The network of the 15 main meteo-stations is too coarse to adequately represent the spatial small-scale weather variability. The higher-resolution observation network was not used because the locations of the obervation points were not available.

In summary, the calculated groundwater recharge rate would be better represented if (1) an average meteo year were used instead of 1988, and (2) if in addition to the 15 main meteo-stations of KNMI, also the higher resolution meteo observation network were used.

The groundwater recharge rate discussed here was used as input flux for the top aquifer in LGMSAT, i.e. for the solution of the potential problem. Consequently, the LGMSAT groundwater recharge rate also affects pathlines and travel times (LGMFLOW). As is discussed in Chapters 4 and 5, the concentrations of the nitrate and pesticides leaching flux were calculated by applying the long-term annual average of net groundwater recharge rate between 1961 and 1990 (Van Drecht & Scheper, 1998). Unlike the LGMSAT groundwater recharge rate which was calculated at each finite-element node, the 1961-1990 annual average recharge rate was calculated as spatially-averaged values for the 500 × 500 grid- cells.

Table 2.2 shows the statistics of the groundwater recharge rate calculated by LGMSAT and the 1961-1990 average rate. *MV* and *Var* are the mean value (mm day^{-1}) and variance, respectively. The results are presented for five selected refined model areas (section 2.6). Table 2.2 indicates that the LGMSAT results are about 20% higher than the 1961-1990 results. The variance of the 1961-1990 results is smaller because spatially averaged values are generated for the 500 × 500 m grid cells. The variance of the LGMSAT results is higher because of the greater variability in groundwater recharge rate within the refined models. There are about four finite-element nodes within each 500 × 500 m grid cell.

2.7 Results Modelling Groundwater Flow

The procedure followed for modelling groundwater flow by LGMSAT follows:
a) Apply a basic finite-element model (section 2.5) to calculate groundwater heads in aquifers. The sole purpose of applying a basic model is to derive groundwater-head boundary conditions for the refined models;
b) For the given basic model, subsequently apply all refined finite-element models at hand residing in the basic model (section 2.6):
   - the input data for a refined model is drawn from the same database as the data for a basic model, i.e. groundwater abstractions are also identical in both cases;
   - with respect to the spatially distributed data, as the finite-element grid in a refined model is about 16 times finer than in a basic model, due to this grid-resolution difference the input values of parameters can be locally different;
   - the groundwater head at the boundary of a refined model is calculated by spatial interpolation (in ARC/INFO) of the groundwater head calculated by the basic model.

The final result of the LGMSAT module needed for subsequent calculation of pathlines and travel times (Chapter 6) is stored in an output file. This so-called "track file" contains, among other data, groundwater heads in aquifers, flux across aquitards, groundwater recharge rate, top-system drainage flux, and the flux between rivers and the top aquifer. Figure 2.7 depicts the sum of groundwater recharge rate and the top-system flux to illustrate this.

The assessment of the leaching flux of nitrate (Chapter 4) and pesticides (Chapter 5) is done under the assumption of a shallow groundwater table, i.e. the water table within a few metres below land surface. Table 2.3 provides information to evaluate the appropriateness of this assumption. The table concerns the depth of the groundwater head in aquifer 1, calculated by means of the 15 refined models. The model area is the conjunction of the 15 refined models (see model periphery line in Figure 2.7). The surface area of the refined models on the Netherlands' territory is 24,262 km^2. In Table 2.3, parts of the model territory located in Germany and Belgium had to be omitted. The 24,262 km^2 represents 91.4% of the total refined model area (in the Netherlands, Germany and Belgium).

Within this model area (24,262 km^2), nitrate and pesticides are used only on agricultural land. The total agricultural land area where infiltration (downward) flux occurs, i.e. including leaching into saturated groundwater, amounts to 9160 km^2. Again, the latter figure relates to the Netherlands' territory only. It is stressed that leaching of nitrate and pesticides can potentially take place within the 9160 km^2, depending on the crop. In reality, the leaching of nitrate and pesticides will occur on only a fraction of the agricultural land.

Table 2.3 *Statistics on depth of calculated groundwater table (GWT) for 15 refined model areas in the Netherlands. Model area is 24,262 km^2, agricultural area with leaching flux is 9160 km^2.*

depth of GWT below land surface	percentage of model area on NL territory	percentage of agricultural area with leaching flux within model area on NL territory
0-1 m	50.3	44.5
1-2 m	28.0	33.3
2-3 m	8.0	10.4
3-4 m	3.3	3.5
4-5 m	2.1	1.9
>5 m	8.3	6.4

The last column in Table 2.3 indicates the assumption of a shallow groundwater table to be justified for most of the agricultural area in this study. The percentage of agricultural land where leaching into saturated groundwater occurs, and where the groundwater table is within 3 m below land surface amounts to 88.2%. For the groundwater table within 5 m below land surface, this percentage is 93.6%. The 6.4% of the agricultural land with leaching flux into saturated groundwater is located primarily on high sandy soil, such as the Veluwe Hills in the centre of the Netherlands and the Hondsrug Hills in the province of Drenthe.

3. SCENARIOS FOR SOLUTE LEACHING INTO GROUNDWATER

3.1 Introduction

This chapter discusses as background information for Chapters 4 and 5 the (economic) scenarios that were used for the assessment of future leaching of nitrate and pesticides into saturated groundwater.

The calculation of pathlines, travel times and solute breakthrough curves at abstraction wells was done for three scenarios for leaching of (a) nitrate and (b) three selected pesticides (atrazine, bentazone, 1,2-dichloropropane) into saturated groundwater. The calculation of concentration in abstracted groundwater was carried out for a period of 1000 years, from 1950.

The input concentration of nitrate and the three pesticides was specified for the following eight time instants:

1) for *historical solute load* (six time instants): 1950, 1960, 1970, 1980, 1990, 1995;
2) for *scenario (future) solute load* (two time instants): 2000, 2020.

Use is made of three scenarios for generating the future solute load of nitrate and pesticides into saturated groundwater. The leaching flux for the scenarios was calculated for the years 2000 and 2020. The three economy-based scenarios were developed by the RIVM for the Environmental Outlook 1997-2020 (RIVM, 1997, section 2.2), on the basis of the three so-called *reference scenarios* developed by the Netherlands Bureau for Economic Policy Analysis (CPB, 1996).

An assessment of the developments within the next 25 years (1995 to 2020) is based on the estimate of changes in a number of factors. The influencing factors are associated with great uncertainty. The three scenarios were used to account for this uncertainty, each leading to a possible future response. The differences between the scenarios are related to global economic development, economic and political development in Europe, and some socio-cultural and technological developments.

The three scenarios are referred to as *Divided Europe (DE)*, *European Coordination (EC)* and *Global Competition (GC)*. The procedures applied for calculating historical loads and scenario values for nitrate and the three pesticides are discussed in Chapters 4 and 5, respectively.

3.2 Reference Scenarios (DE, EC, GC)

The three scenarios are called *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*). These are three plausible long-term scenarios for the Netherlands for the period 1995-2020. International economic and political developments, demographic, social and cultural factors, and technological and economic trends are the key determinants of the scenarios.

Global Competition is the scenario featuring the most rapid economic growth, both internationally and nationally. Market forces play a central role. Technology develops rapidly.

In *European Coordination*, economic growth in the Netherlands is somewhat lower than in Global Competition. Population growth is, of all three scenarios, the highest here. Compared to their counterpart in Global Competition, market forces play a less prominent role. Government intervention is important and policy cordination plays a greater role than in the other scenarios, especially on the European level.

Divided Europe envisions a Europe characterized by antagonism and nationalist sentiments. Economic problems are not properly addressed. The upshoot for Europe and therefore the Netherlands is weak economic growth.

The *European Coordination* scenario is considered to be the most probable one.

3.3 Implications of Scenarios for Nitrate and Pesticides

In general, there is a relationship between economic activity and the resulting impact (stresses) on the environment. The nature of this coupling can be changed by environmental policy measures, by changes in technology or changes in the structure of economic activities. Both consumption and production are important factors in this respect. The merit of this study is to provide an insight into the effect of various scenarios on the impact on the subsurface environment in terms of quality of groundwater abstracted for drinking-water purposes.

In the framework of the fourth national environmental survey study (Environmental Outlook 1997-2020) carried out by RIVM (RIVM, 1997), each of the three "economic" scenarios was "translated" in terms of its effect on the environment during the next 25 years (1995-2020). This "translation" is done for each (economic) interest group (agriculture, industry, traffic, nature, etc.), taking into account a set of anticipated environmental policy measures. An example of an environmental policy measure is reduction of emissions by agriculture.

Evidently, the most important factor associated with nitrate and pesticides is agriculture (RIVM, 1997, section 2.4.4). An additional, relatively minor, source of nitrate pollution forms emissions from traffic and industry.

The amount of nitrogen for fertilizers and manure to be applied by agriculture in the Netherlands in 2000 and 2020 according to the three scenarios is listed in RIVM (1997, Table 2.4.21). Unlike for pesticides, where all three scenarios (*DE*, *EC*, *GC*) were considered, only one scenario was applied for nitrate, namely *EC*. The *EC* scenario only was applied because (1) it is more probable than scenarios *DE* and *GC*, and (2) the nitrogen load is similar for all three scenarios.

The scenario for nitrate is referred to as *EC(plus)* instead of *EC*. The suffix "*plus*" has a historical background. It was introduced in the course of the environmental survey study and should not be attributed any specific meaning here.

The final result of scenario calculations are nationwide maps of the concentration of nitrate and pesticide leaching into saturated groundwater. The resolution of the maps is 500 × 500 m. Two maps are provided for each solute, for the years 2000 and 2020. The procedures applied for calculating scenario values for leaching of nitrate and the three pesticides are discussed in detail in Chapters 4 and 5, respectively.

4. MODELLING NITRATE LEACHING

4.1 Introduction

This chapter documents the assessment of nitrate leaching flux into saturated groundwater, leaching flux used for calculation of nitrate concentration in abstracted groundwater (Chapter 7).

Fraters & Boumans (1997) mention that eutrophication of soil and groundwater is caused primarily by atmospheric deposition of nitrogen and excessive application of nitrogen in agriculture, excessive in the sense that the applied amount of nutrients is higher than crop uptake. Atmospheric deposition of nitrogen is caused by emissions from industry, transport and agriculture. In forested areas, especially along the forest fringes, deposition is the main source of nitrogen load. According to various Dutch national studies (c.f. RIVM, 1996), the average nitrogen load on Dutch agricultural land in 1994 amounted 500 kg ha^{-1}. About 50 kg ha^{-1} of this amount resulted from atmospheric deposition.

It is important to stress that the procedure followed here for the assessment of the nitrate leaching flux is, in principle, applicable only to a relatively shallow groundwater table. Section 2.7 (Table 2.3) showns that the assumption of the shallow groundwater table is met in most of the agricultural area in this study. The percentage of agricultural land where leaching into saturated groundwater occurs, and where the groundwater table is between 3 and 5 m below land surface, amounts to 88.2% and 93.6% respectively.

The procedure described in this chapter yields <u>concentrations of nitrate in the leaching flux into saturated groundwater</u> as average values within 500 × 500 m grid cells. The 500 × 500 m spatial resolution is considered appropriate. This is because the spatial resolution of the leaching flux input should be more-or-less the same as the resolution used for the solution of the "hydraulic" problem, i.e. pathlines and travel times. The density of the finite-element grid was about 250 × 250 m, with local refinements. This grid density makes it possible to adequatly represent a well field by a single well in the "centre of gravity" of the well field. The pathline tracking was done from a regular system of square-shaped cells sized 250 × 250 m (section 6.2). Theoretically, the specification of nitrate leaching flux at the 250 × 250 m resolution would have resulted in a higher accuracy of the simulated concentration breakthrough curves. However, the improvement in accuracy due to the refinement from 500 × 500 m to 250 × 250 m was expected to be relatively small. For this reason, the 500 × 500 m spatial resolution is considered sufficiently accurate.

4.2 Method

Nitrate leaching was calculated for the 1950-2020 period. The years 1950, 1960, 1970, 1980, 1990 and 1995 represent so-called historical leaching years, 2000 and 2020 are future (scenario) years. Data on land use, soil type, groundwater depth class, precipitation, evapotranspiration, manuring data and atmospheric N-deposition are available for each grid cell. Soil type and groundwater depth class are derived from the digitized soil map of the Netherlands (prepared by DLO-Staring Centre, Wageningen). Use is made of the land-use map LGN1 (Thunnissen *et al.*, 1992), as described in Chapter 10. The original LGN1 data are available on 25 × 25 m scale. For study purposes, the original data were upscaled for 500 × 500 m grid cells, containing a certain percentage of specific land use with each 500 × 500 m grid cell. The upscaled LGN1 map was applied to all years, though land use changed over the 1950-1995 period. Precipitation excess (net groundwater recharge) was calculated from 30 years (1961-1990) average precipitation and reference crop evaporation (Van Drecht & Scheper, 1998). For each combination of land use and soil type in a municipality, mean N-application rates from manure and fertilizer are available (RIVM, 1996). Maps of atmospheric N-deposition are available from the OPS and DEADM models developed at RIVM (Bleeker & Erisman, 1997).

The assessment of nitrate leaching to shallow groundwater is carried out through a combination of three methods, depending on the land use:

- steady-state model, NLOAD, describing nitrate leaching from agricultural soils,
- regression model, describing nitrate concentration in shallow groundwater in forested areas, and
- mean nitrate concentration in groundwater at a depth of about 10 m in urban areas, using data from the Dutch national groundwater quality monitoring network (LMG).

Referring to Table 1.1 (Chapter 1), the percentages of agricultural area, forested area and urban area are 57.1%, 26.4% and 8.9%, respectively, within the surface area of the capture zones. As the agricultural area is dominant over other land-use types and as the major nitrate load occurs on agricultural land, the calculation of nitrate leaching flux on agricultural land (section 4.2.1) will be discussed in more detail.

Inherent to the difference in the model concept, the three methods used for calculating the nitrate leaching flux have different prediction reliability, i.e. the ability to mimic the "actual" nitrate leaching flux as an average within 500 × 500 m grid cells. The fact that the predictive ability is different for each method, does not hamper the approach used.

The mean nitrate_N leaching rate (kg ha^{-1} $year^{-1}$) in a grid cell is calculated as an area-weighted average of the nitrate load of different types of land use in that grid cell.

The nitrate concentration (mg l^{-1}) in infiltrating groundwater was calculated by dividing the mean nitrate_N leaching rate (kg ha^{-1} $year^{-1}$) by the rate of net groundwater recharge (m $year^{-1}$). Use was made of the long-term annual average of net groundwater recharge rate (1961-1990) calculated by Van Drecht & Scheper (1998). Consequently, no use was made of the net groundwater recharge as it was calculated by LGMSAT for the meteorological year 1988 (Figure 2.7). The use of the 1961-1990 annual average recharge for calculating the nitrate concentration reflects the long-term hydrological reality better than the 1988 recharge rate. If the 1988 groundwater recharge rate were used, which is about 20% higher (section 2.4), the nitrate concentrations would be about 20% smaller. The nitrate concentration (mg l^{-1}) served as input for LGMCAM (section 7.2).

4.2.1 Nitrate Leaching in Agricultural Areas

The model NLOAD (Van Drecht *et al.*, 1991; Van Drecht, 1993) employs a straightforward and empirical approach, describing the steady-state Nitrate_N leaching from agricultural soils on annual basis. NLOAD is used regularly at RIVM to assess the effects of environmental policy on nitrate leaching and exceedance of the EC standard for drinking water (50 mg l^{-1} NO_3). During the recent five years, the model was regularly updated. The current version of the model is documented in Van Drecht & Scheper (1998). A PC-version of NLOAD (used by G. van Drecht) has as input the land-use map LGN1 (Thunnissen *et al.*, 1992).

Model concept

Nitrate_N leaching of well-drained agricultural land is calculated as the sum of background leaching, leaching due to N-fertilization, additional leaching from animal manure and leaching from N supplied by grazing cattle on grassland. The background leaching is low (3 to 5 kg ha^{-1} $year^{-1}$). The atmospheric N-deposition is added to the fertilizer-N application rate. Total N-fertilization is the sum of the fertilizer-N application rate, atmospheric N-deposition, and plant-available nitrogen in manure. Plant-available nitrogen in manure is about 60-80% of the total applied manure-N (after substracting ammonia emissions). The leaching due to N-fertilization is calculated as a portion of N-fertilization. This portion (expressed as a leaching coefficient) increases with increasing N-fertilization. The urine-N excretion of grazing cattle is also a nonlinear function of N-fertilization level. About 50% of urine-N in urine spots is available for plants. Because of the high N_fertilization of urine spots, leaching is dealt with separately. Additional leaching of manure-N is a portion of the remaining N after having subtracted ammonia volatization and plant-available N. The most probable reason why the remainder "gets lost" is denitrification. As NLOAD is a steady-state model, accumulation of nitrogen in soil profile is not taken into account.

Denitrification is highly dependent on the depth of the groundwater table. After having calculated nitrate leaching of well-drained land by NLOAD in a separate post-processing step, an empirical correction for denitrification was applied (Table 4.1). The denitrification relation is based on nitrate observations at 10 experimental farms. The smaller the value in the right-most column in Table 4.1, the larger the relative effect of denitrification. Table 4.1 should not be misinterpreted: the value 1.00 for GTC = VII* does not imply that denitrification cannot take place at that depth.

Table 4.1 *Relative nitrate leaching as a function of the groundwater table class (GTC), mean lowest groundwater level in Summer (MLW) and mean highest groundwater level in Winter (MHL). Levels are in cm below land surface (LS); an asterix indicates a drier subclass (after Boumans et al., 1989).*

GTC class	MLW cm -LS	MHW cm -LS	relative leaching
II	50-80	<25	0.05
III	80-120	<25	0.08
III*	80-120	25-40	0.31
IV	80-120	40-80	0.43
V	120-160	<25	0.50
V*	120-160	25-40	0.48
VI	120-160	40-80	0.65
VII	160-220	80-140	0.83
VII*	220-280	140-220	1.00

Model validation

According to Van Drecht & Scheper (1998), the NLOAD model gives an acceptable prediction of the leaching observed at the experimental fields (parcel, ca. 1 ha). However, the model underestimates the leaching at very high fertilization levels. The model yields realistic results (predictions) for grassland on sandy soils, but still is not reliable for other combinations of agricultural land use and soil types.

Another validation of NLOAD is reported by Boumans & Van Drecht (1998, section 3.2, Table 2). Contrary to the experimental-field-based validation carried out by Van Drecht & Scheper (1998), this validation was done for the grid blocks of 500 × 500 m, i.e. for 25 ha. A major conclusion is that NLOAD underestimates the actual leaching fluxes into saturated groundwater, especially at shallow groundwater tables ("shallow" according to the GTC data from the soil map). The actual concentration in leaching fluxes can be 1.5 to 10 times higher than the NLOAD-calculated concentration. The difference between actual and calculated concentrations depends, among other things, on the depth of the groundwater table, the error decreasing with increasing depth of the groundwater table. Boumans & Van Drecht (1998) conclude that this discrepancy is probably caused by the fact that the groundwater table class (GTC) - according to the soil map - does not match the GTC observed in the field.

Another reason for the discrepancy between observed and NLOAD-calculated values within the 500 × 500 m grid-blocks could be lack of knowledge about spatial distribution of nitrate load (input for NLOAD). The information on the nitrate load from agricultural application is available as a constant value within each municipality. As the relationship between the nitrate load and nitrate leaching flux is nonlinear (for each combination of crop and soil type), disregarding the spatial variability in nitrate load within municipalities may "filter out" spatially localized peaks of nitrate leaching, thus disproportionally decreasing the average leaching flux within the 500 × 500 m grid-blocks.

Data processing

To enable application of NLOAD on a national scale, digitized maps of municipality boundaries, land use, soil type, groundwater depth class, precipitation and evaporation are needed. All maps are available in a grid-cell format (dominant category per grid cell of 500 × 500 m). Application rates of nitrogen in manure and fertilizer as well as nitrogen supplied by grazing cattle are calculated from agricultural production statistics (RIVM, 1996). Application rates are available for each crop, soil type and municipality. For the years before 1985, application rates are estimated using national trends in manure production (Table 4.2). The moment of manure application has changed over the years 1980-1995 from Winter to Spring (Table 4.3). This trend will be assumed to continue after 2000.

Table 4.2 *Trends in manure application rates and atmospheric N-deposition for the reference year, 1985 = 100.*

year	N-animal	N-fertilizer agriculture	atm. N-deposition	atm. NHx-deposition forest areas
1945				30
1950	36	32	37	35
1955	45	39	47	41
1960	50	44	51	48
1965	54	49	56	53
1970	68	76	70	66
1975	78	86	81	77
1980	90	95	93	90
1985	100	100	100	100
1990	104	82	91	93
1995	112	75	71	66
2000	101	65	60	53
2020	86	43	54	40

Table 4.3 *Percentage of Spring application of animal manure.*

land use	soil type	1950-1990	1991	1992	1996	2000	2020
grassland	sand/clay/peat	50	60	70	70	80	95
arable land	sand	50	60	60	60	80	95
arable land	clay/peat	0	0	0	0	10	50

The nitrate_N leaching rate was calculated for each crop, soil type, and municipality and year. A nitrate leaching map was made for each crop. Afterwards, the weighted mean nitrate leaching rate of agricultural land use was calculated using the surface area of each crop within the grid cell of 500 × 500 m.

4.2.2 Nitrate Leaching in Forested Areas

Shallow groundwater in forest and heather fields in sandy soil regions of the Netherlands was sampled and analysed in 1989 and 1990 (Boumans & Beltman, 1991). This field survey resulted in 155 mean nitrate concentrations in 155 grid cells (500 × 500 m). Nitrate concentration is dependent on soil type, groundwater depth, fraction of coniferous forest, surface roughness and atmospheric N-deposition. These "geographical" variables are available from geographical grid cell-based databases. The data was statistically analysed by Leeters *et al.* (1994), Boumans (1994) and Boumans & Van Drecht (1998), resulting in a number of regression models. The models describe nitrate concentrations as a function of available geographical data on a grid-cell scale. For the purposes of this study, the following "forest model" was derived for grid cells of 500 × 500 m:

$$[NO3]^{0.35} = rc_0 + rc_1 \times th \times cf \times NHx + rc_2 \times forest$$

where:

[NO3]	=	nitrate concentration normalized by the drinking-water quality standard (50 mg l^{-1}) (-),
rc_0	=	soil-type-dependent regression coefficient (-):

soil no.	description	value
1	dry & rich (enkeerdgronden)	1.0352
2	wet & poor (humuspodzolgronden)	0.7300
3	wet, organic (moerige eerdgrond)	0.5800

rc_1	=	regression coefficient: 0.00975 (ha year $kmol^{-1}$ m^{-1}),
th	=	mean tree height (m) of the forest stands in a grid cell,
cf	=	surface area coniferous forest of the forest stands in a grid cell (fraction),
NHx	=	dry atmospheric deposition of ammonia_N (kmol ha^{-1} $year^{-1}$), (Bleeker & Erisman, 1997),
rc_2	=	regression coefficient: 0.3327 (-),
forest	=	surface area of forest and natural areas within 3×3 grid cells, i.e. grid cell and its eight neighbours derived from LGN1 (fraction) (Thunnissen *et al.*, 1992).

Note that the product *th* × *cf* × *NHx* represents the interception.

The model estimates mean nitrate concentrations in groundwater in grid cells under forest for the years 1989/1990. It was not developed with the aim to predict concentrations for future years. For this reason, systematic errors can occur when the model is applied to scenario studies. An example of a systematic error is the effect of changing NO_x deposition as part of a scenario, which is not accounted for in the regression model. Another problem is that future change of surface area and properties of the forest stands are not dealt with. The assumption of model parameters being constant in time is considered acceptable for sensitivity analysis of the effect of changing atmospheric ammonia-N deposition rates.

The *percentage variance accounted for* (*adjusted R^2 statistics*) in the forest model was 42.5%. Recently, the model was updated using new data on the atmospheric ammonia deposition (Boumans & Van Drecht, 1998). They report an explained variance of 35%. In a previous study, Leeters ***et al.*** (1994) reported a percentage variance accounted of 33%. This gives an idea of the accuracy of model predictions on the grid scale of 500 × 500 m.

The reliability of the method can be further explained by discussing two application modes. As the grid scale is fixed (500 × 500 m), both application modes listed below are hypothetical and serve as illustration only:

a) If the above regression approach were applied to grid cells smaller than 500 × 500 m, the reliability of model predictions would gradually decrease with decreasing grid-cell size. Then the model could even produce systematic errors. For details refer to Boumans & Van Drecht (1998).
b) On the other hand, if the grid-cell size increased, the reliability of the leachate concentration for that cell would also increase. From Boumans (1994, Table 5) it follows that for the concentration between 25 and 50 mg l^{-1} NO_3, the model prediction error on the national scale (i.e. a single "average" value) would be about 5% around the predicted value.

Regional dry NHx deposition maps are available for the years between 1980 and 2020 from transport models (Bleeker & Erisman, 1997). These maps contain the mean dry atmospheric NHx-deposition on the land surface within grid cells of 5000 × 5000 m. Differences in surface roughness between agricultural crops, forest and urban areas are taken into account. Maps of the mean atmospheric deposition in forested areas are not available. Lack of this data could cause systematic errors on a local scale (e.g. 50 × 50 m), however, not on the scale of 500 × 500 m, such as in this study.

The NHx deposition map of the year 1985 is used to calibrate the forest model for the year 1990. The national trend in atmospheric deposition is retrieved from the available regional deposition maps (years between 1980 and 2020). For the years before 1980, national environmental statistics data are used. To apply the model to the year 1995, atmospheric deposition from 1990 (93% of 1985) is calculated. A mean time lag of five years is appropriate for atmospheric deposition to affect the nitrate concentration in groundwater in forest areas.

The forest model predicts the mean nitrate concentration in shallow groundwater of forest in a grid cell. Nitrate_N load at the top of the phreatic aquifer is calculated with the mean precipitation excess of the 1961-1990 period (Van Drecht & Scheper, 1998).

4.2.3 Nitrate Leaching in Urban Areas

Observed nitrate concentrations in groundwater at a depth of 10 m in urban areas are available from the wells of the Dutch national groundwater quality monitoring network (LMG) (Van Drecht *et al.*, 1996). The mean concentration in the 1984-1995 period has not changed significantly. The mean value of 5.6 mg l^{-1} NO_3-N is used to estimate the mean nitrate_N load of the phreatic aquifer, assuming a mean precipitation excess of 300 mm $year^{-1}$ in urban areas.

4.3 Historical Concentrations (1950, 1960, 1970, 1980, 1990, 1995)

The resulting maps of historical leaching fluxes of nitrate are presented in Figures 4.1 through 4.6. The data is generated for grid cells of 500 × 500 m.

4.4 Future (Scenario) Concentrations (2000, 2020)

The maps of nitrate leaching fluxes for the scenario years 2000 and 2020 are depicted in Figures 4.7 and 4.8.

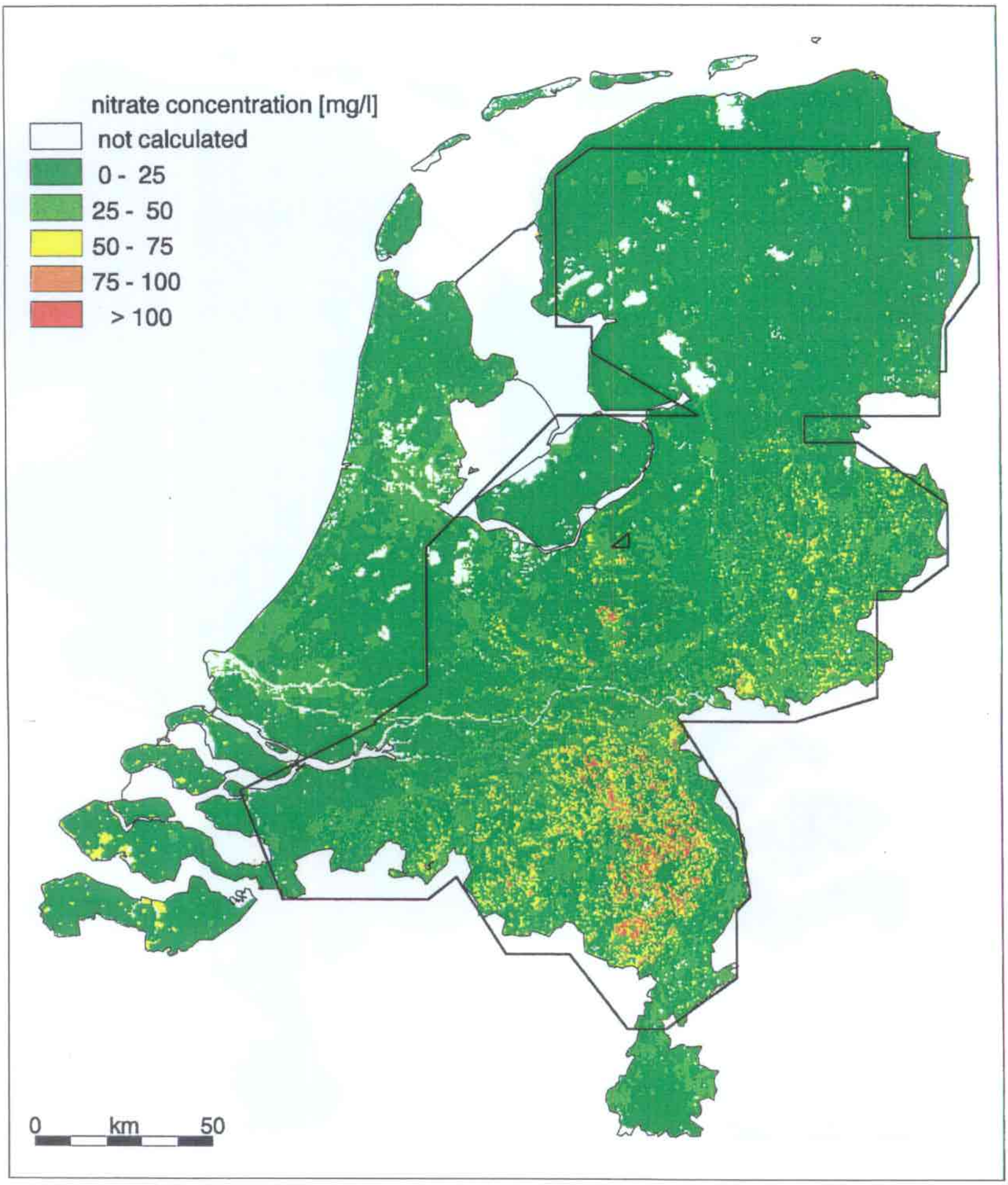


Figure 4.1 *Leaching of nitrate to shallow groundwater, historical, 1950.*

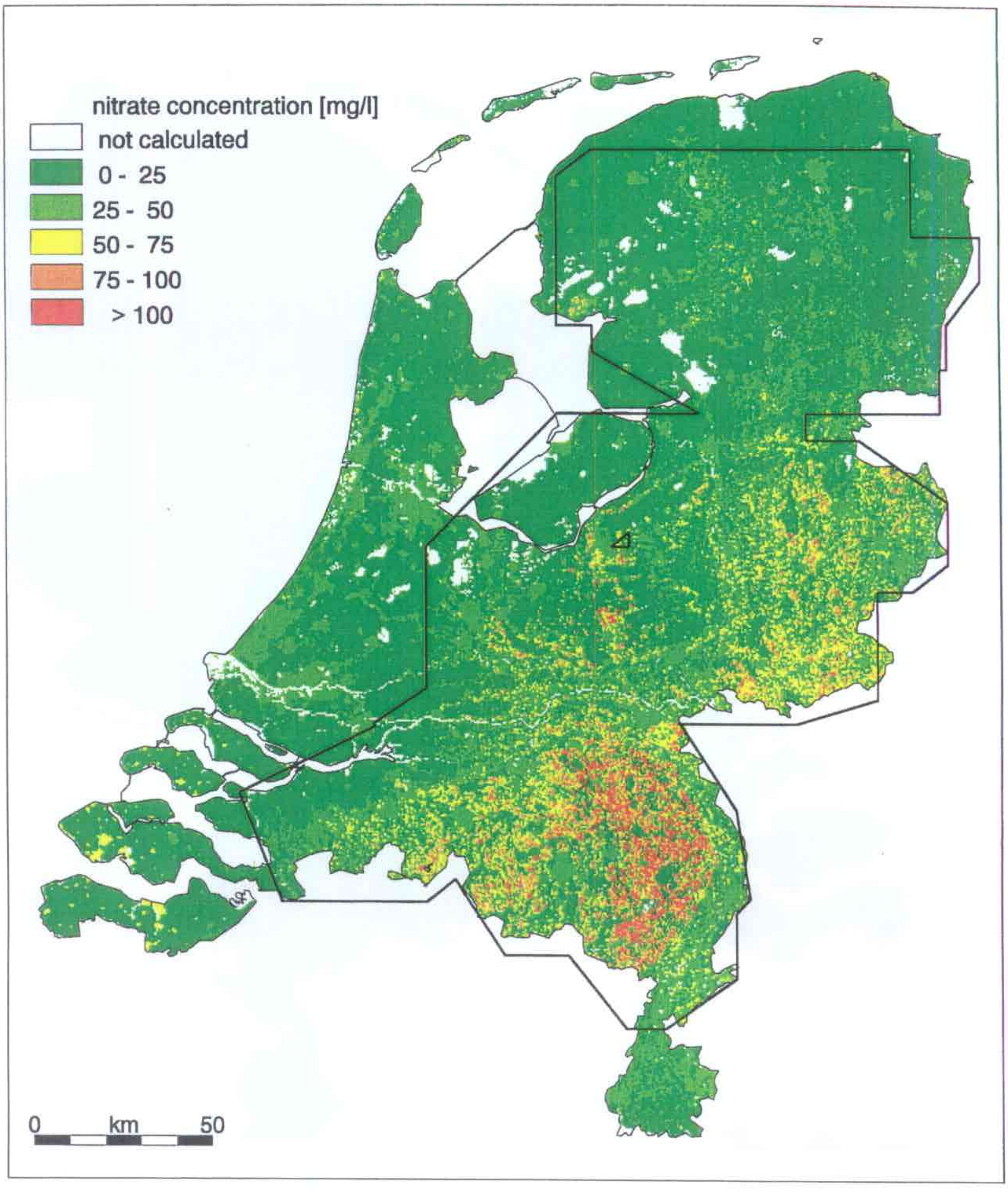


Figure 4.2 *Leaching of nitrate to shallow groundwater, historical, 1960.*

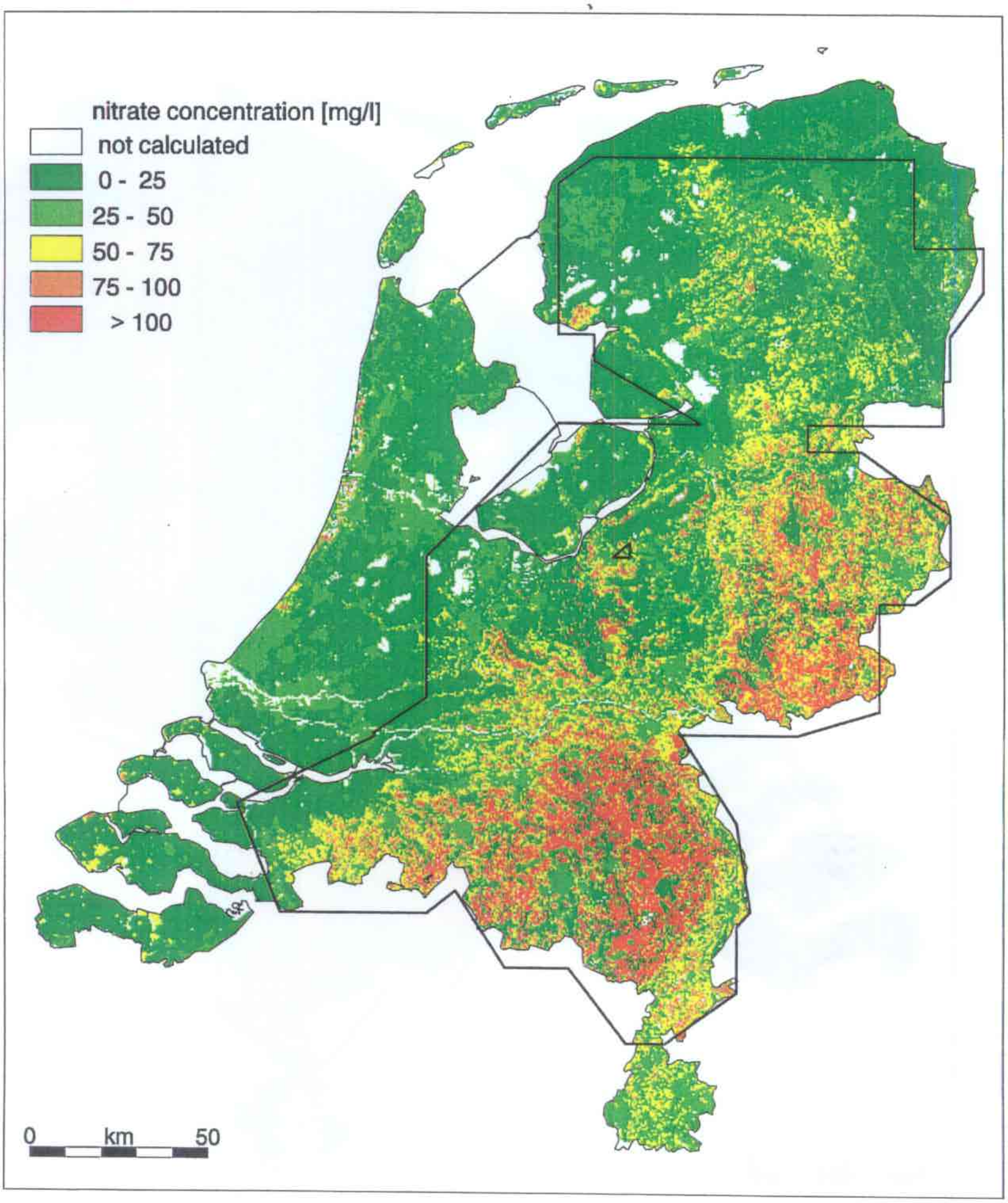


Figure 4.3 *Leaching of nitrate to shallow groundwater, historical, 1970.*

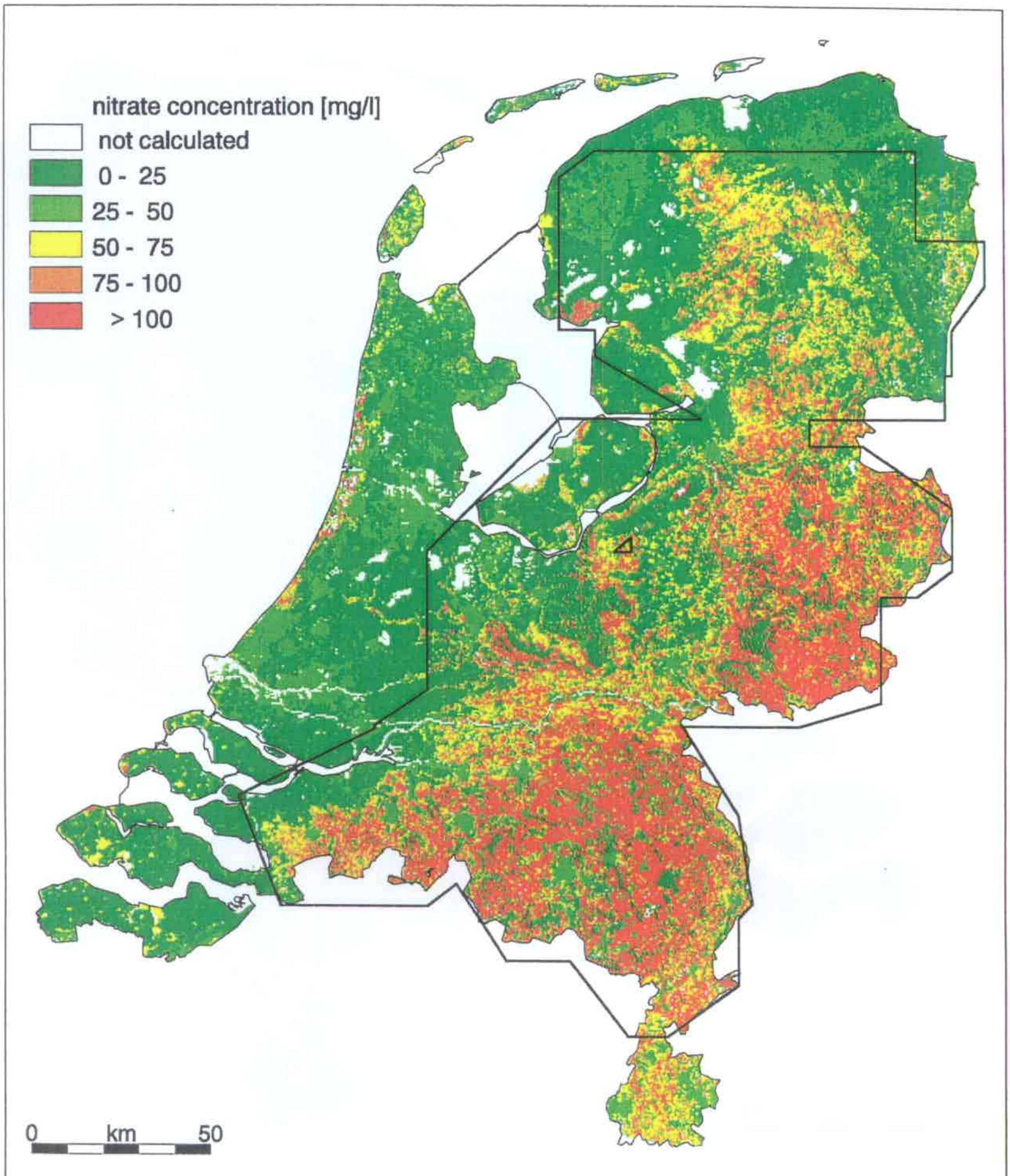


Figure 4.4 *Leaching of nitrate to shallow groundwater, historical, 1980.*

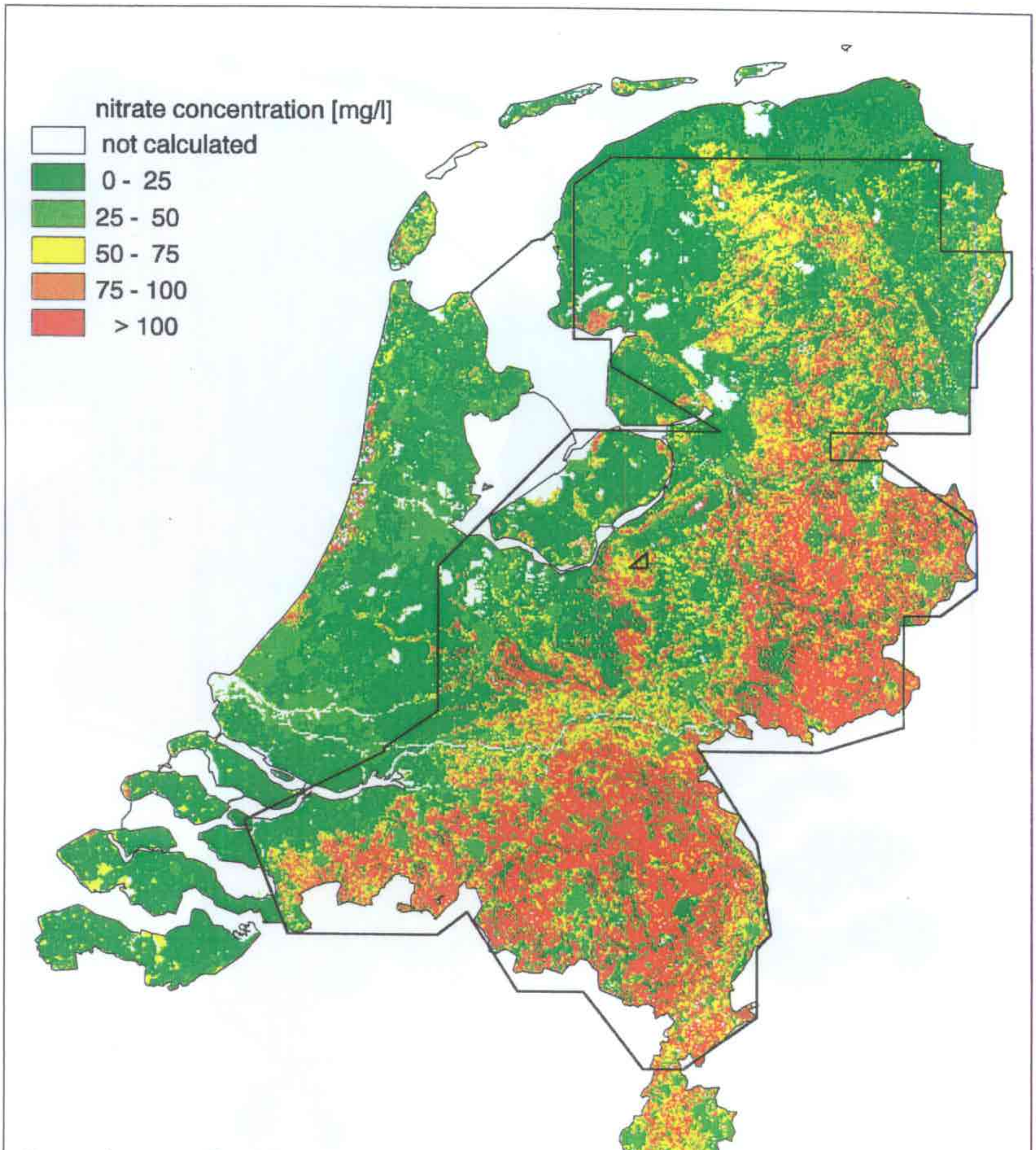


Figure 4.5 *Leaching of nitrate to shallow groundwater, historical, 1990.*

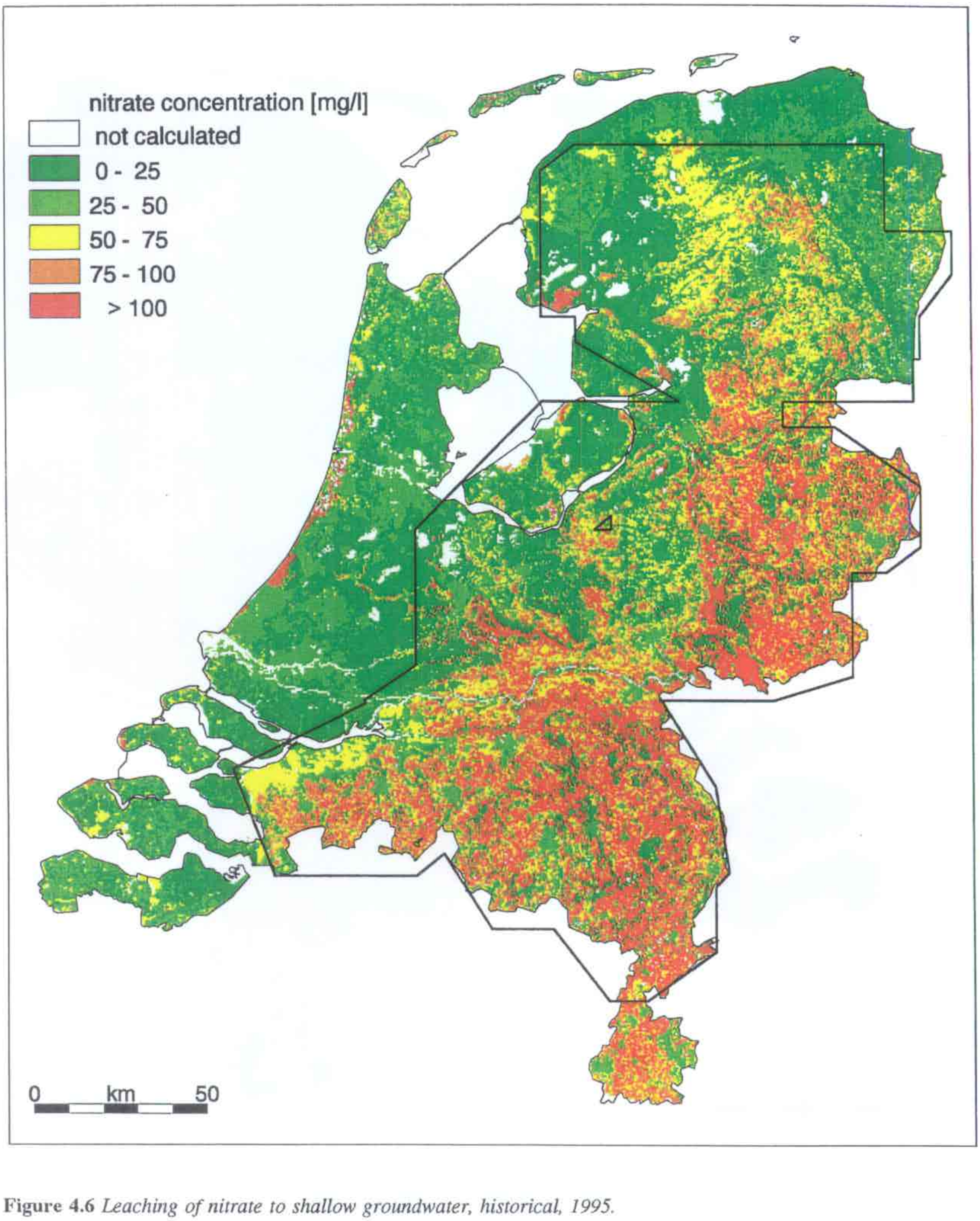


Figure 4.6 *Leaching of nitrate to shallow groundwater, historical, 1995.*

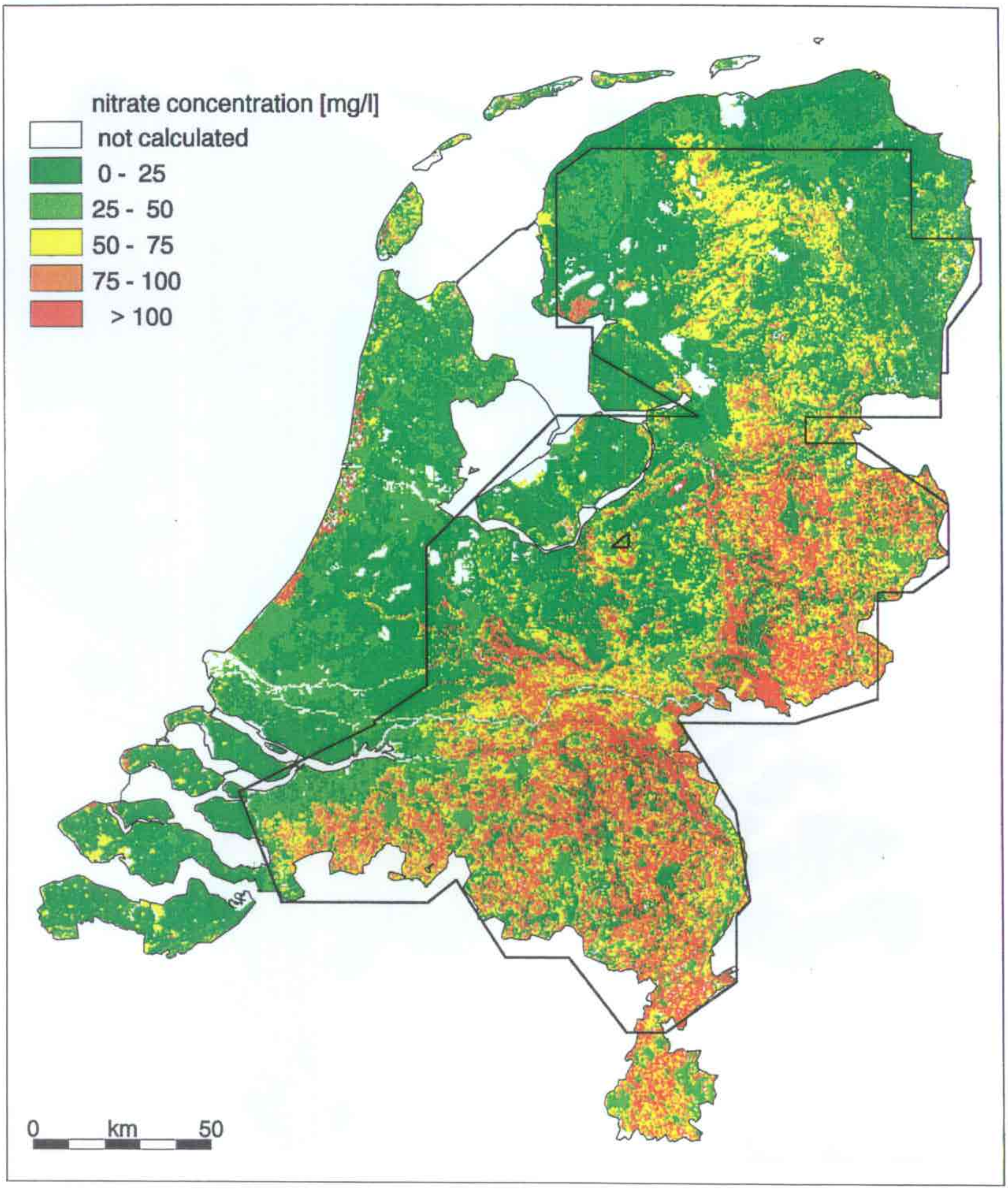


Figure 4.7 *Leaching of nitrate to shallow groundwater, scenario EC(plus), 2000.*

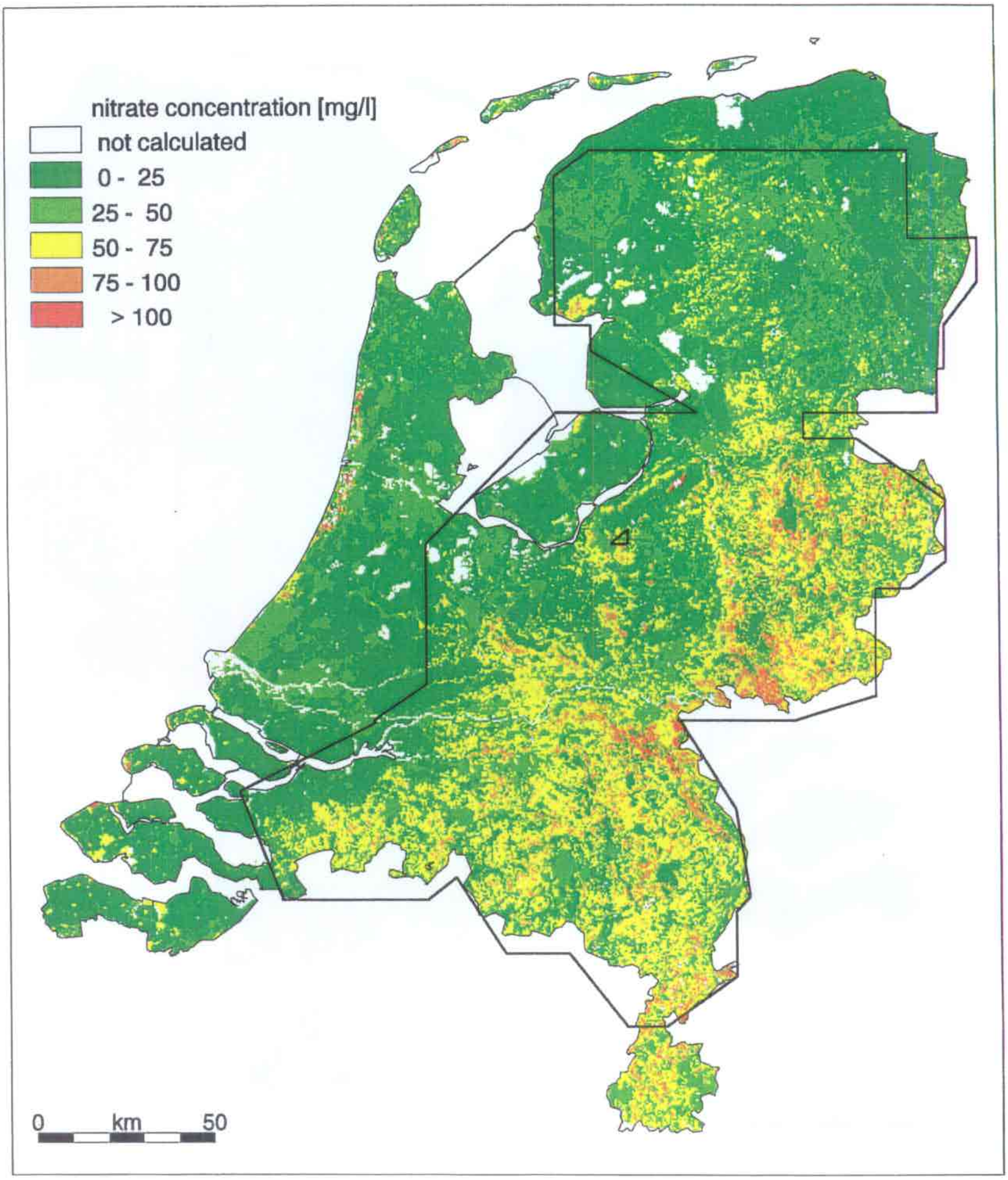


Figure 4.8 *Leaching of nitrate to shallow groundwater, scenario EC(plus), 2020.*

5. MODELLING PESTICIDE LEACHING

5.1 Introduction

This chapter deals with the assessment of pesticide leaching fluxes into saturated groundwater by using the model GEOPESTRAS. The leaching flux is to be used for calculation of pesticide concentration in abstracted groundwater (Chapter 7).

Agriculture is the dominant source of pesticide residues in groundwater. Although the environmental hazard associated with the observed chemical concentrations is poorly defined, it is generally accepted that leaching losses from agricultural fields to the groundwater should be minimized. An important reason for concern about groundwater contamination by pesticides in the Netherlands is that groundwater is an important source supplying about two-third of the drinking-water production.

In the Netherlands, 337 pesticides were officially registered at the time of the study (Merkelbach *et al.*, 1998). In 1995, the total estimated leaching of pesticides to the shallow groundwater was 43,000 kg. It turned out that 17 pesticides covered 80% of the total amount that leached to the shallow groundwater (Tiktak *et al.*, 1996). The best-known examples of pesticides and related products that pose a problem for drinking-water production are 1,2-dichloropropane, atrazine and bentazone. Especially 1,2-dichloropropane is found in abstracted groundwater.

The European Union's principle is that drinking water should not contain any pesticides. The EU drinking-water standard (maximum admissible concentration) is 0.5 $\mu g\ l^{-1}$ for the total amount of pesticide residues, as specified in Council Directive 80/778/EEC. In the Netherlands, the principle of pesticide-free drinking water was implemented in 1986 by adopting 0.1 $\mu g\ l^{-1}$ as the maximum admissible concentration for a single pesticide and 0.5 $\mu g\ l^{-1}$ for the total of pesticides and their residues (Netherlands Water Supply Companies Directive). At that time, 0.1 $\mu g\ l^{-1}$ was the analysis detection limit. If groundwater is not processed to remove pesticides, then the limit value of 0.1 $\mu g\ l^{-1}$ applies directly to the quality of abstracted groundwater. Currently, the concentration of pesticides exceeds the drinking-water standard in abstracted groundwater at some water-supply companies (Versteegh *et al.*, 1995).

As this study is restricted to phreatic and semi-confined groundwater as source for drinking-water production, it does not consider specific pesticide-related problems faced at groundwater abstractions of artificially recharged surface water. These are abstractions in the vicinity of large rivers (e.g. the Rhine River) and abstraction in dunes where treated surface water (e.g. from the Rhine River) is recharged into the ground by ponds and canals. Atrazine and bentazone are encountered most often in this type of drinking-water production.

The final product of the process described in this chapter are concentrations of pesticide in the leaching flux into saturated groundwater, as constant values within 500 × 500 m grid cells. Considered appropriate is the 500 × 500 m spatial resolution analogous to nitrate. For discussion of the selection of the 500 × 500 m resolution refer to section 4.1.

5.2 Method

The calculation of the amount of pesticide leaching to shallow groundwater for the "basic" year 1995 was carried out with the GEOPESTRAS model (Tiktak *et al.*, 1996). This model consists of two major modules. The physically-based pesticide leaching model PESTRAS (Tiktak *et al.*, 1994; Freijer *et al.*, 1996) is run in a first step to calculate the leaching of pesticides for unique combinations of soil type, groundwater depth class, crop type and climate, using one standard dose of 1 kg active ingredient per hectare. Apart from soil fumigants such as (cis)1,3-dichloropropene, this is a realistic dose. An important input of PESTRAS is the sorption constant (K_{om}) and the half-lifetime (DT_{50}). Results of PESTRAS include (1) the maximum concentration of pesticide in shallow groundwater, and (2) the fraction of pesticide leached. In a second step, these two model outputs are combined with results from the Information System on Pesticide Use, ISBEST (Merkelbach *et al.*, 1998). ISBEST provides information on the actual total pesticide use per municipality, the area on which the pesticide has been applied, and the average dose on those parcels where the pesticide has actually been used. Multiplication of the fraction leached by the total pesticide use yields the total amount (mass) of pesticide leached.

The data on the sorption constant (K_{om}) and the half-lifetime (DT_{50}) are averaged values drawn from the RIVM database TOXIS, containing confidential information provided by the industry. The information in TOXIS is essentially based on results from experiments performed to fulfill the requirements of the product registration on the market.

The input for LGM is the pesticide concentration in groundwater infiltrating from the unsaturated zone into the saturated zone, in $\mu g\ l^{-1}$. GEOPESTRAS generates the total mass of pesticide per unit of area ($kg\ m^{-2}\ year^{-1}$) that has leached into saturated groundwater between the 1-2 m depth below ground level during the so-called "basic" year 1995. The pesticide concentration ($\mu g\ l^{-1}$) in infiltrating groundwater was calculated by dividing the pesticide mass (M L^{-2} T^{-1}) by the rate of net groundwater recharge (L T^{-1}). In order to ensure compatibility with the method for the calculation of the nitrate leaching flux (Chapter 4), use was made of the long-term annual average of net groundwater recharge rate (1961-1990) calculated for nitrate leaching (Van Drecht & Scheper, 1998). The resulting leachate concentration (µg l^{-1}) served as input for LGMCAM (section 7.3).

The leaching fluxes for the years 1950-1990, preceding the "basic" year 1995, and future (scenario) leaching fluxes (2000 and 2020) were obtained by multiplying the leaching fluxes for the "basic" year 1995 with a spatially-constant factor, which was primarily based on expert judgement of past and future pesticide use.

As mentioned before, the calculation of the leaching flux by GEOPESTRAS was carried out for shallow groundwater, i.e. for the groundwater table within a few metres below land surface. It is expected that the pesticide leaching flux generated by GEOPESTRAS can be used as input for LGMCAM without causing noticeable errors for groundwater table depths up to 3 m below land surface. The maximum depth of the groundwater table acceptable without introducing major errors is arbitrarily assumed to be 5 m below land surface. From Table 2.3 (section 2.7) it follows that the assumption of shallow groundwater table below agricultural land is satisfied for most of the agricultural area in this study. The percentage of agricultural land where leaching into saturated groundwater occurs, and where the groundwater table is within 3 and 5 m below land surface, amounts to 88.2% and 93.6%, respectively.

Validation of GEOPESTRAS

So far, GEOPESTRAS results have not been validated on a regionalized scale (500 × 500 m), i.e. by comparing the leaching values calculated for a 500 × 500 m grid-block with (observed) values relevant at that spatial scale. The validation has not yet been carried out, mainly due to the high costs associated with pesticide analyses. Depending on the pesticide, the analysis is done either by gas chromatography, GC, or high performance liquid chromatograph, HPLC. Another reason why validation has not yet been conducted - obviously related to the first-mentioned reason - is the large number of pesticides admitted for application.

Van der Linden & Boesten (1989) carried out a sensitivity analysis of the PESTRAS results on the uncertainty in sorption and decay (degradation, transformation) of pesticides in a "column setup". The uncertainty in sorption and decay is related to the variability of soil profile properties, such as the content of organic matter, variability in the organic matter and variability in the soil microbial biomass. The results in Van der Linden & Boesten (1989, Chapter 4, Figure 4.3) suggest that the modelled leaching rate is, in general, very sensitive to the variation of the sorption constant (K_{om}) and the half-lifetime (DT_{50}). Taking into account the uncertainty in K_{om} and DT_{50}, the actual leaching values can vary roughly between 1/100 and 100 times the calculated value. However, as the study by Van der Linden & Boesten (1989) concerned a sensitivity analysis carried out for a hypothetical "column model", no conclusions can be drawn with respect to the modelling capability of GEOPESTRAS on a regional scale, such as 500 × 500 m.

The regional-scale model validation is also hampered by the fact that pesticide leaching is affected by short-term temporal variation in a number of processes, such as in water flow, temperature and volatilization. To account for the transient processes, monitoring data required for testing the model should be available at a high temporal resolution. As a second-best alternative to regional-scale validation, <u>PESTRAS</u> has been applied in a number of <u>field studies</u>. Boekhold *et al.* (1993) describe an experiment on a 1-ha plot. Bentazone concentrations were observed at a number of time instants in a dense network of observation points within the experimental field. Tiktak *et al.* (1998) describe an experiment in a 0.5-ha field, also with a dense network of pesticide observation points, for bentazone and ethoprophos. In both experiments, the PESTRAS input data for sorption (K_{om}) and decay (DT_{50}) were derived from soil samples from the field (batch expriments). Note again that the GEOPESTRAS application discussed in this report is based on averaged values, rather than on data obtained for actual locations within the Netherlands.

An important reason for inappropriateness of straighforward "extrapolation" of the experimental-field results to a regional scale, such as 500 × 500 m, is that the field experiments do not necessarily cover the full range of conditions encountered by the regional scale model. In these cases, processes important at part of the area to be simulated can easily be overlooked. For example, the two above-mentioned field experiments were carried out in sandy soils, where preferential flow was not an important process at the time of the experiment. Moreover, model tests for a field experiment on a limited spatial scale (e.g. 1 ha) do not necessarily properly represent the effects of model parameterization on a regional scale (here 500 × 500 m). Obviously, pesticide properties obtained from a batch sample at one field plot cannot, in general, be considered as representative input values for a 500 × 500 m grid block. As suggested by Loague & Corwin (1996), this problem can be handled by calibrating the model using simultaneously the data observed at several sites, instead of employing - as spatial aggregation - the results of stand-alone field experiments. The methodology proposed by Loague & Corwin (1996) which relies on parameter optimization is promising for the future.

The above-mentioned considerations imply that regional-scale assessments of pesticide leaching carried out in this study are associated with a considerable degree of uncertainty. In the previously mentioned field experiment with bentazone, Boekhold *et al.* (1993) found:

$$0.5 < \textit{observed concentration / calculated concentration} < 2.0$$

to be statistically significant. This uncertainty range in the bentazone leaching flux is also supported by findings of Tiktak *et al.* (1998).

When upscaling the local information, e.g. from an experimental field to a regionalized level, an important factor is the variability of data in space. Tiktak *et al.* (1996) carried out a simple min-max analysis with the GEOPESTRAS model to account for spatial variability of input data (soil physical and chemical parameters) within soil-map units, and showed that the maximum concentration of atrazine in the shallow groundwater could vary by one order of magnitude. The information available so far stresses the need for uncertainty assessment when modelling the pesticide leaching in future, especially on the regional scale of 500 × 500 m.

The overall conclusion drawn from the available information is that the uncertainty in the regionalized pesticide leaching values assessed by GEOPESTRAS can be considerable. Most probably, the actual leaching values (concentrations), as "averages" per 500 × 500 m block, can vary at least between half and double of the calculated leaching value.

No information is available about the distribution of the possible uncertainty in space. Neither is it known if the calculated leaching fluxes are systematically larger or smaller than the actual values.

5.3 Pesticide Description and Usage

In this section, the features of the three pesticides will be shortly described, including the field of their application.

Atrazine

Atrazine is a symmetric triazine used as a post-emergence herbicide in the cultivation of maize. The compound is usually mixed with other herbicides and applied at dosage rates of approximately 700 g ha^{-1}. As maize cultivation is an almost continuous process, application takes place on a yearly basis. In the unsaturated soil the compound is transformed into a number of metabolites, of which desethylatrazine and desisopropylatrazine are important in view of leaching to groundwater. The half-lifetime of atrazine in soil from the plough layer is approximately 50 days; its soil organic matter sorption coefficient is about 70 l kg^{-1}. The relevant metabolites have comparable half-lifetimes, but are somewhat more mobile in the soil. As atrazine is applied only to maize, the compound and its metabolites may be found in groundwater in areas where maize is grown; especially the sandy soil areas of the Netherlands.

Bentazone

Bentazone is a post-emergence herbicide in the cultivation of many crops, especially maize, seed grass and vegetables. The compound is used either alone (in, for example, seed grass and vegetables) in amounts of ca. 1.4 kg ha^{-1}, or in combination with other compounds (e.g. maize) in amounts of ca. 700 g ha^{-1}. Dependent on the crop type, bentazone might be used on a yearly basis. In the unsaturated zone bentazone is transformed relatively fast (half-lifetime 10-20 days). The sorption of the compound is almost negligible (K_{om} = 0.4 l kg^{-1}) and dependent on the amount of precipitation excess the compound may leach to groundwater. When the precipitation excess is high, shortly after application, there is not enough time for transformation and the compound may leach. As far as it is known, there are no relevant metabolites with respect to leaching. Bentazone is used in all areas of the Netherlands, but with some concentration in the sandy soil areas because of the maize cultivation.

1,2-Dichloropropane

1,2-Dichloropropane is a major contaminant in formulations of the soil disinfectant 1,3-dichloropropene. Treatment of the soil with fumigants takes place in intensive cultivation of flower bulbs, potatoes and sugar beet. Contaminant concentrations have not been constant; until approximately 1985, 1,2-dichloropropane constituted up to 30% of the formulation, thereafter (after recognizing negative effects of the compound) the concentration diminished to approximately 0.1%. Further clean-up of the formulation is hardly possible because of very similar physical properties of both compounds. Application rates of cis-1,3-dichloropropene are around 100 kg ha^{-1}. Taking into account contamination levels, the amounts of 1,2-dichloropropane reaching the soil diminished from ca. 100 kg ha^{-1} before 1985, to ca. 10 and 1 kg ha^{-1} (1985-1990) and, finally, to ca. 100 g ha^{-1} after 1990.

Liquid 1,3-dichloropropene formulations are injected in the soil, but the components volatilize very rapidly. The active ingredient is transformed quite readily through both chemical and biological processes. Some leaching of this compound to the groundwater may occur, but the compound is unstable in the saturated zone and therefore further transport to deeper groundwater is not very likely. In contrast, 1,2-dichloropropane is either not or only very slowly transformed in the unsaturated zone and sorption to the organic matter of the soil is weak (K_{om} is 15 l kg^{-1}). Leaching of the compound to the groundwater may occur. Areas in which starch potatoes are grown are shown to be the most important areas for 1,2-dichloropropane appearing in groundwater.

5.4 Historical Concentrations (1950, 1960, 1970, 1980, 1990, 1995)

The use of chemical plant protection products came into focus in the Netherlands shortly after the Second World War. Both the fast growing industry and the growing population required efficient use of labour and land, and high yields. The requirements led to a growing use of chemical plant protection products until approximately 1990, when the government introduced reduction plans for these agents. The following text describes globally the use of atrazine, bentazone and 1,3-dichloropropene in the Netherlands since the Second World War. Exact figures were not available until 1984.

As indicated in section 5.3, atrazine and bentazone are used as herbicides and for both products maize is the most important crop. The use of these compounds, therefore, reflects to a large extent the area cultivated to maize. 1,3-Dichloropropene is used as a soil fumigant, mainly in bulbs, potato and sugar-beet cultivation. The use of this compound reflects the narrowing of crop rotation schemes and the higher pest pressure going along with it. The contaminant 1,2-dichloropropane in formulations of 1,3-dichloropropene does not follow the sales rates of 1,3-dichloropropene because of changing impurity percentages in the course of time (see section 5.3).

Atrazine

Atrazine was introduced in the Netherlands around 1960, but its use was fairly low until the booming of maize grown as cattle fodder (end sixties, early seventies). The use grew steadily until approximately 1985, when the maximum level was reached, becoming approximately constant after this, despite a growth in the area cultivated for maize. This is mainly due to using mixtures of herbicides and substituting atrazine with other herbicides.

Bentazone

Bentazone was introduced in the Netherlands around 1970. Although its use is not restricted to maize, the growth of the market share of the compound correlates quite well with the area dedicated to the production of silage maize. From 1970 onwards an increase was observed until a maximum was reached around 1985. Since then the use has been fairly constant, with 1987 being an exception with a use of approximately 130% of the maximum. As also observed for atrazine, the use of bentazone in mixtures with other herbicides and substitution by other pesticides contribute to the stable market share despite of the growing area dedicated to the production of maize.

1,2-Dichloropropane

1,2-Dichloropropane, a major contaminant of 1,3-dichloropropene, was introduced in the Netherlands around 1960, shortly after the introduction of metam-sodium. In a very short time metam-sodium and 1,3-dichloropropene competed for the leading position on the market (the third soil fumigant, methylbromide, being used only in greenhouses). The choice between the two compounds depends merely on weather conditions in autumn. Around 1970 sales approached maximum levels and stayed constant until 1990. From then onwards the use of 1,3-dichloropropene declined because of a policy of restricted use of the compound and the substitution of the isomeric mixture of the compounds by the active cis-component. Meanwhile also 1,2-dichloropropane levels in the formulation products of 1,3-dichloropropene declined (see section 5.3). In 1996, the amount of 1,3-dichloropropene used was about 20% of its maximum. In 1996, a total ban on cis-1,3-dichloropropene was announced, but so far this ban is not operational.

Method used for leaching assessment

As mentioned in section 5.1, the assessment of the leaching of pesticides to the shallow groundwater in the years 1950, 1960, 1970, 1980 and 1990 was carried out assuming linear relationships in the output of the GEOPESTRAS model with respect to the "basic" year 1995. This means that the output for a certain year, e.g. 1950, was calculated as product of the output for the year 1995 and a spatially-constant multiplication factor. The factors are summarized in Table 5.1.

Table 5.1 *Summary of multiplication factors for historical leaching of pesticides. The numbers in parentheses refer to the basic year, 1995.*

year	atrazine	bentazone	1,2-dichloropropane
1950	0.00	0.00	0.00
1960	0.05	0.00	158.00
1970	0.62	0.09	863.00
1980	0.95	0.81	651.00
1990	0.98	0.98	2.50
(1995)	(1.00)	(1.00)	(1.00)

5.5 Future (Scenario) Concentrations (2000, 2020)

The simulations of pesticide variation in abstracted groundwater were carried out for three scenarios (Chapter 3), namely *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*). Similarly to the procedure for the assessment of historical leaching, the assessment of pesticide leaching for 2000 and 2020 was done by multiplying the output for the year 1995 by a spatially constant factor. The multiplication factors are summarized in Tables 5.2 and 5.3.

Table 5.2 *Summary of multiplication factors for leaching of atrazine and bentazone.*

year	scenario *DE*	scenario *EC*	scenario *GC*
(1995)	(1.00)	(1.00)	(1.00)
2000	0.85	0.82	0.80
2020	0.69	0.69	0.58

Table 5.3 *Summary of multiplication factors for leaching of 1,2-dichloropropane.*

year	scenario *DE*	scenario *EC*	scenario *GC*
(1995)	(1.00)	(1.00)	(1.00)
2000	0.00	0.00	0.00
2020	0.00	0.00	0.00

The multiplication factors for 1,2-dichloropropane are zero because in 1996 a total ban on cis-1,3-dichloropropene was announced. So far, this ban is not operational.

For practical reasons, only the results of pesticide leaching for the *EC* scenario are presented in Figures 5.1 through 5.18. The maps are not shown for the years when pesticide leaching not occurred.

Previewing the results discussed in section 6.2, the total calculated area of the 165 capture zones is 1703 km^2, a figure comparable with the area of 9160 km^2, being the total agricultural area in the 15 refined models (Table 2.3, section 2.7). The areal extent of the potential pollution by pesticides within the capture zones of 165 abstraction sites (1703 km^2) was previously indicated in Table 1.2 (Chapter 1), namely that atrazine/bentazone and 1,2-dichloropropane is potentially applied on 88 and 45 km^2 of agricultural land, respectively.

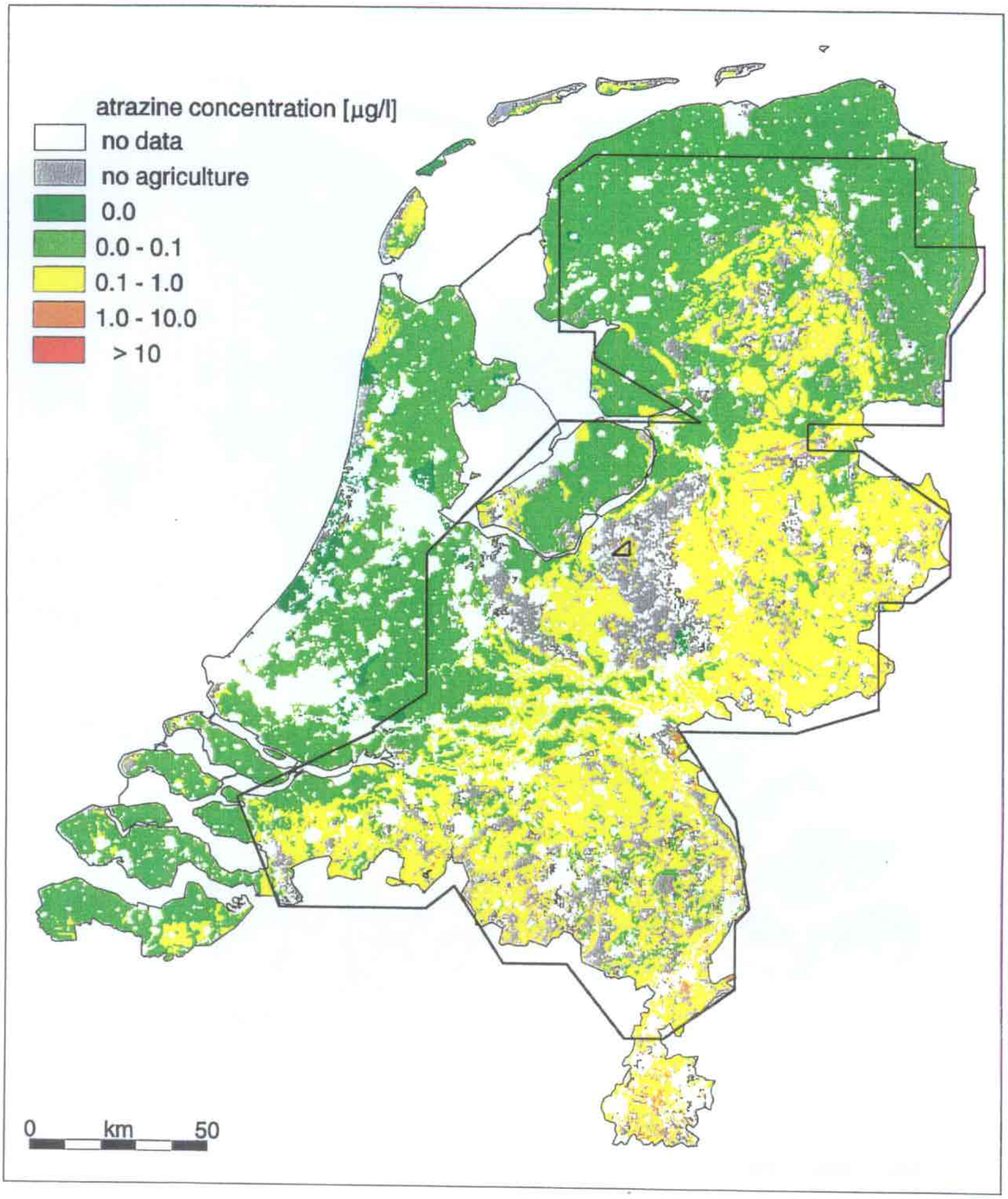


Figure 5.1 *Leaching of atrazine to shallow groundwater, historical, 1970.*

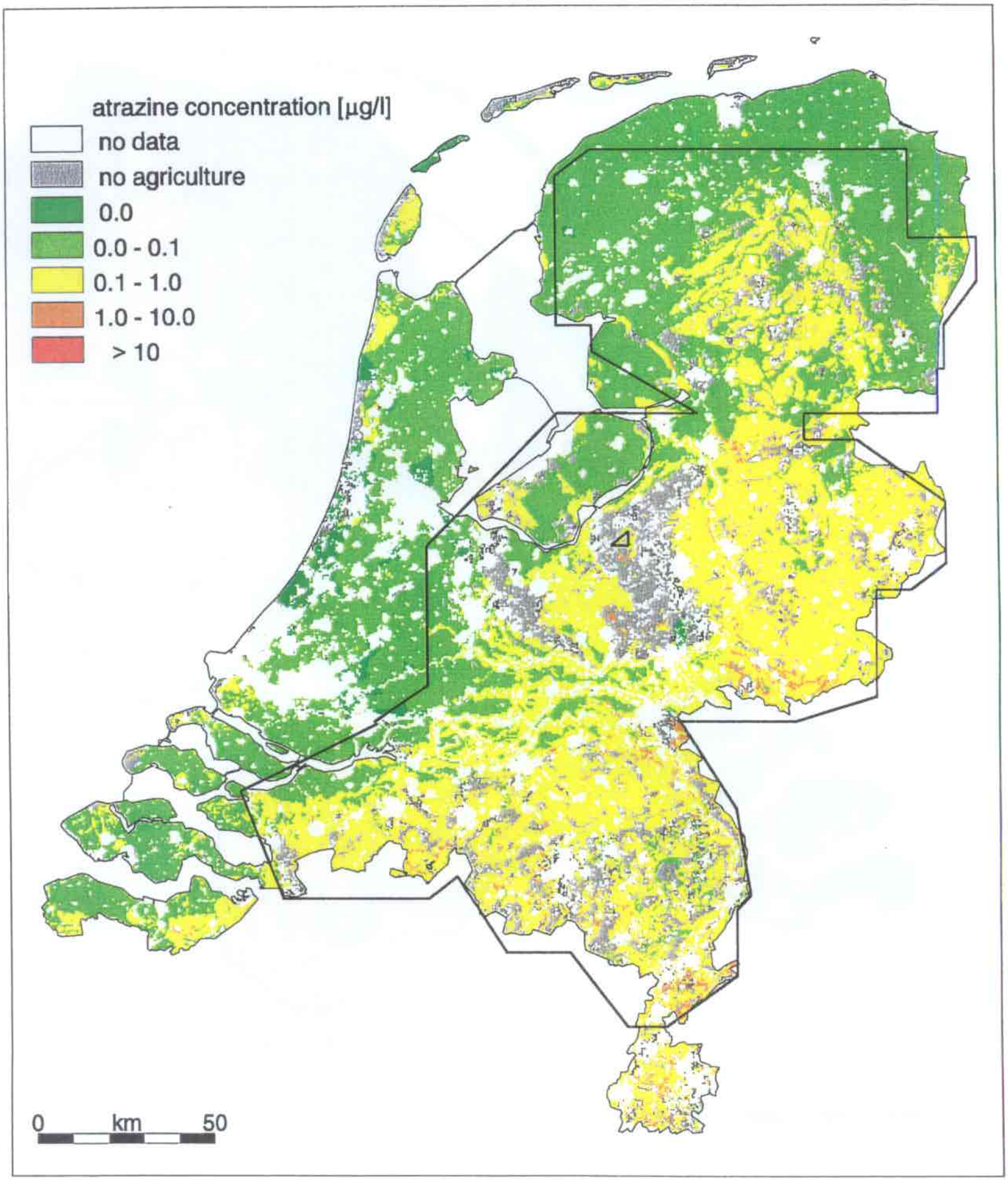


Figure 5.2 *Leaching of atrazine to shallow groundwater, historical, 1980.*

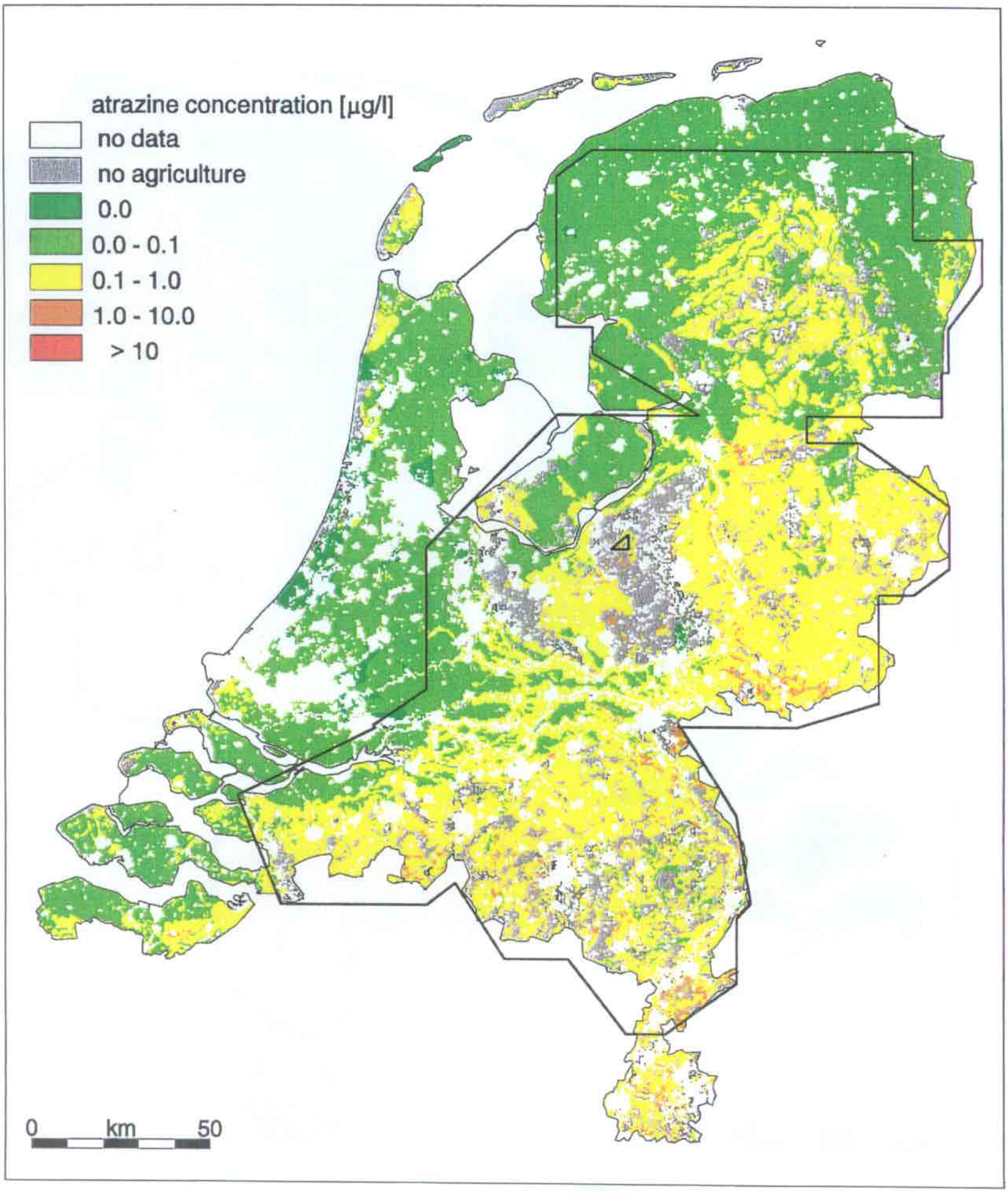


Figure 5.3 *Leaching of atrazine to shallow groundwater, historical, 1990.*

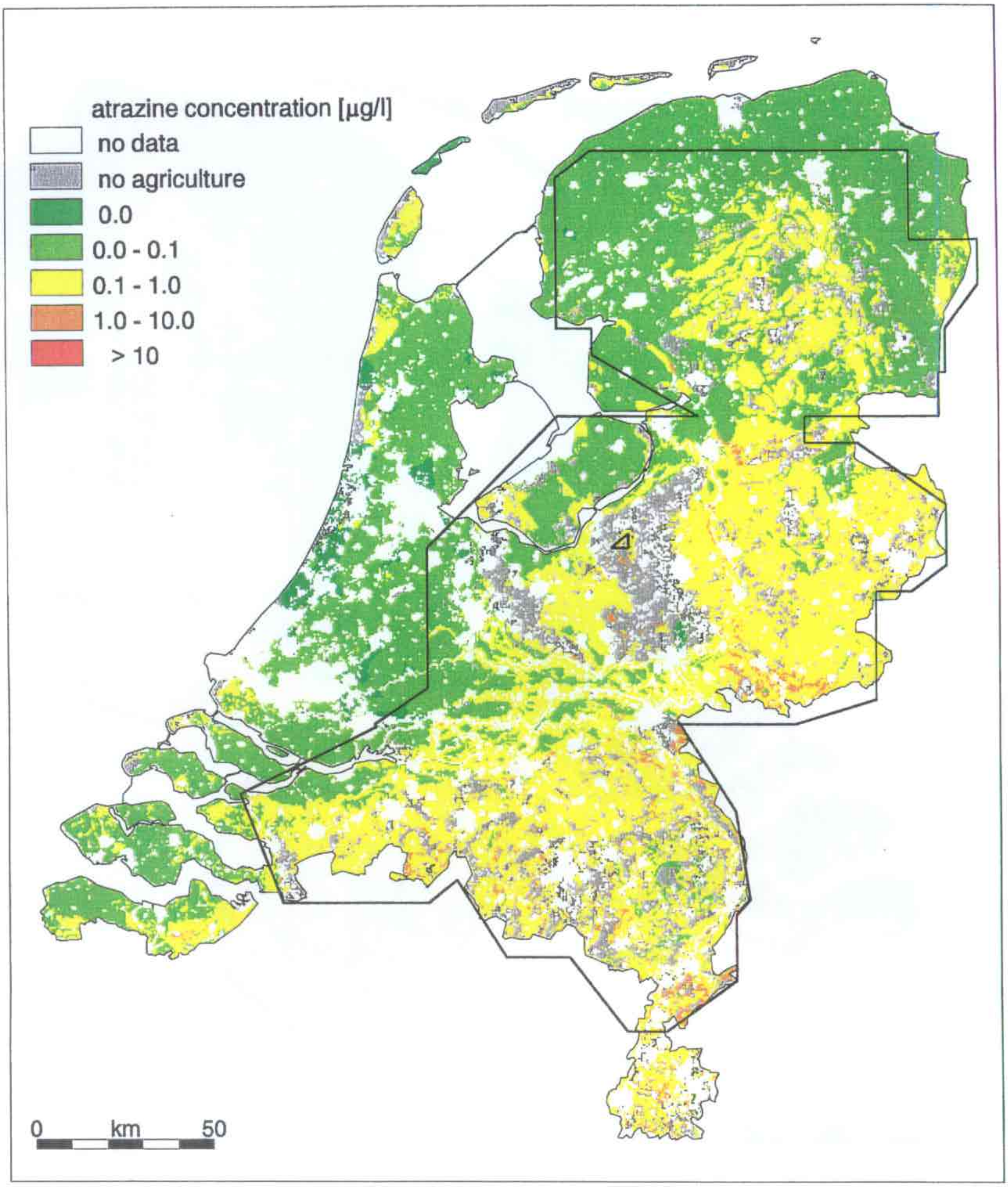


Figure 5.4 *Leaching of atrazine to shallow groundwater, historical, 1995.*

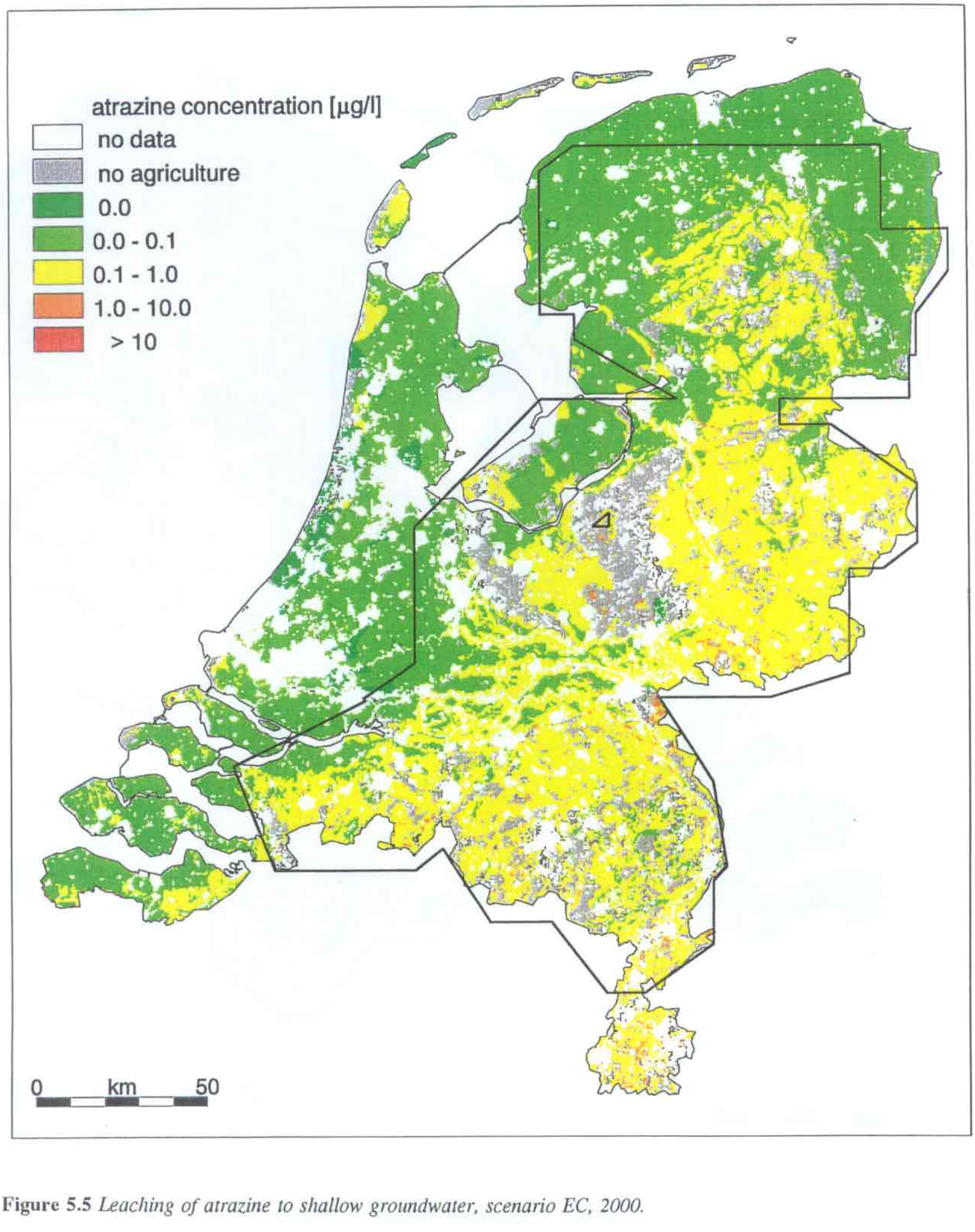


Figure 5.5 *Leaching of atrazine to shallow groundwater, scenario EC, 2000.*

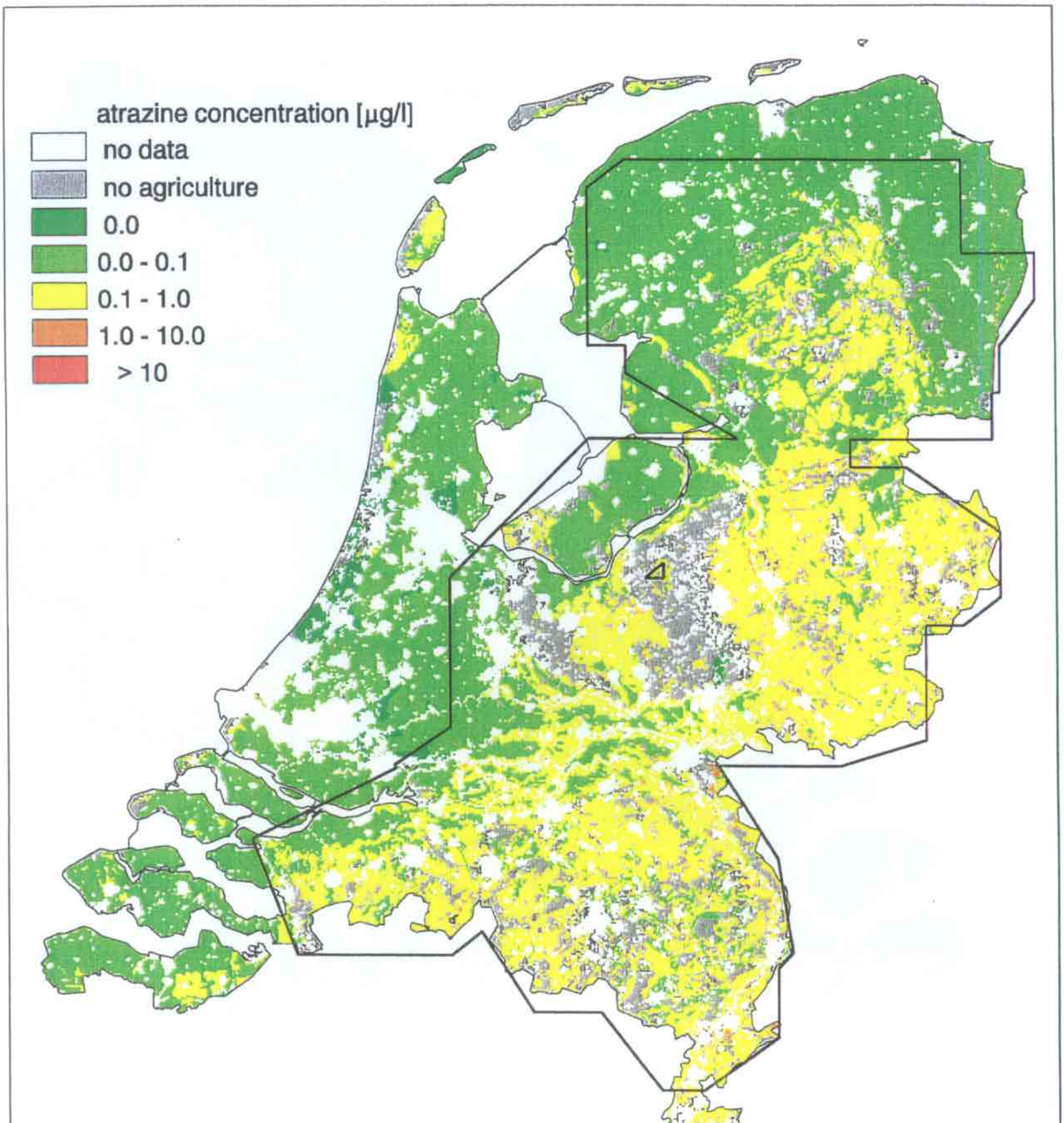


Figure 5.6 *Leaching of atrazine to shallow groundwater, scenario EC, 2020.*

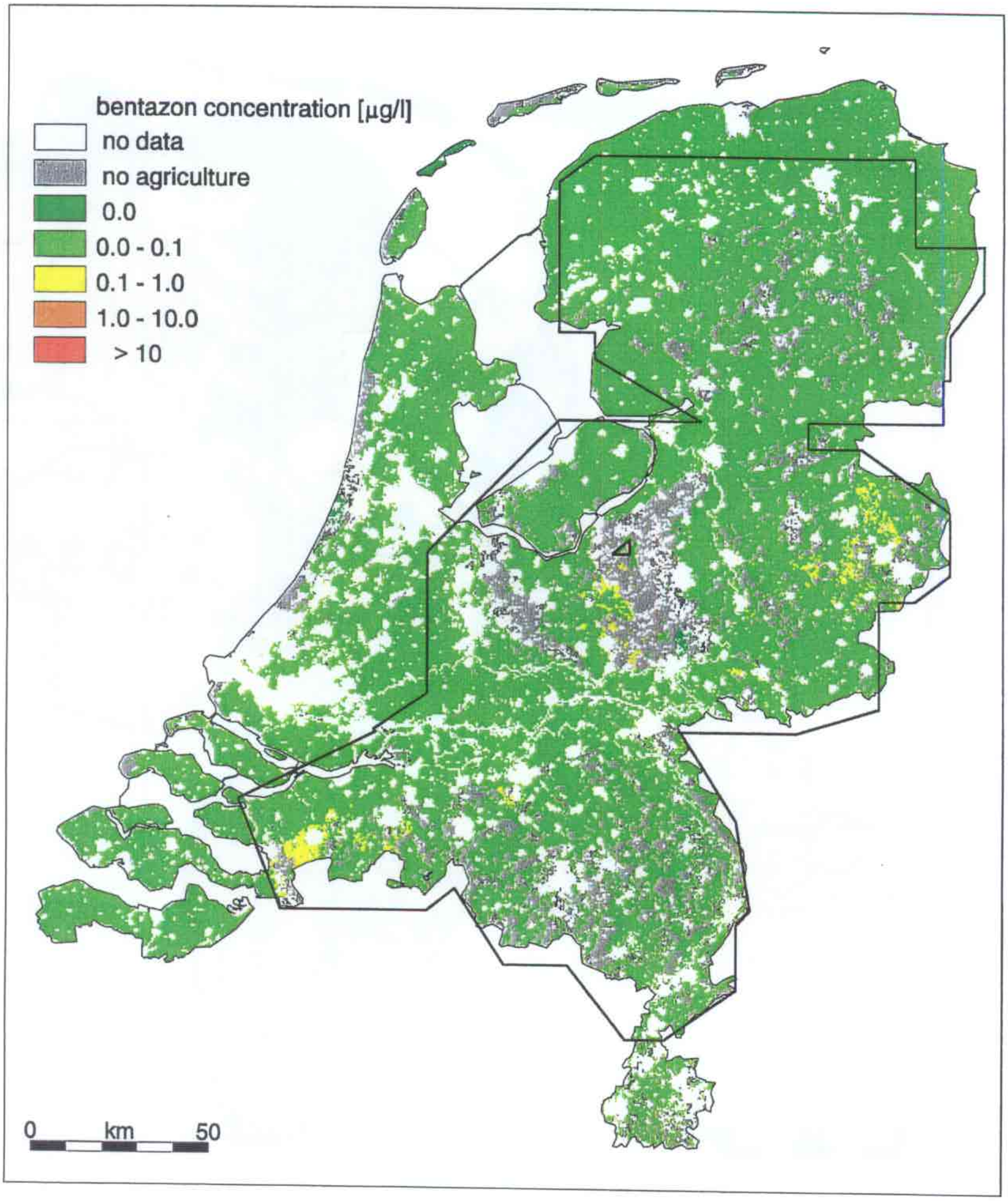


Figure 5.7 *Leaching of bentazone to shallow groundwater, historical, 1970.*

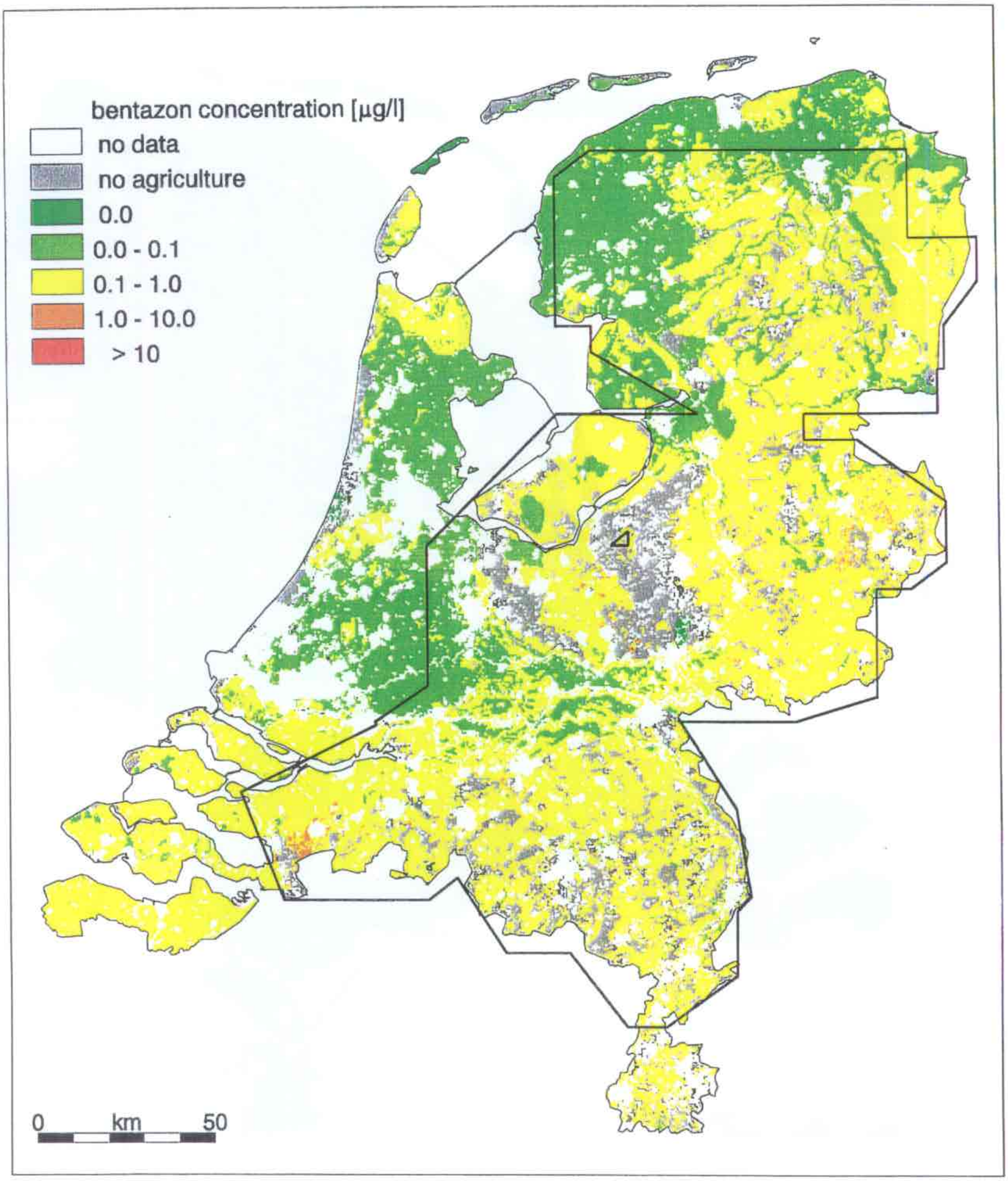


Figure 5.8 *Leaching of bentazone to shallow groundwater, historical, 1980.*

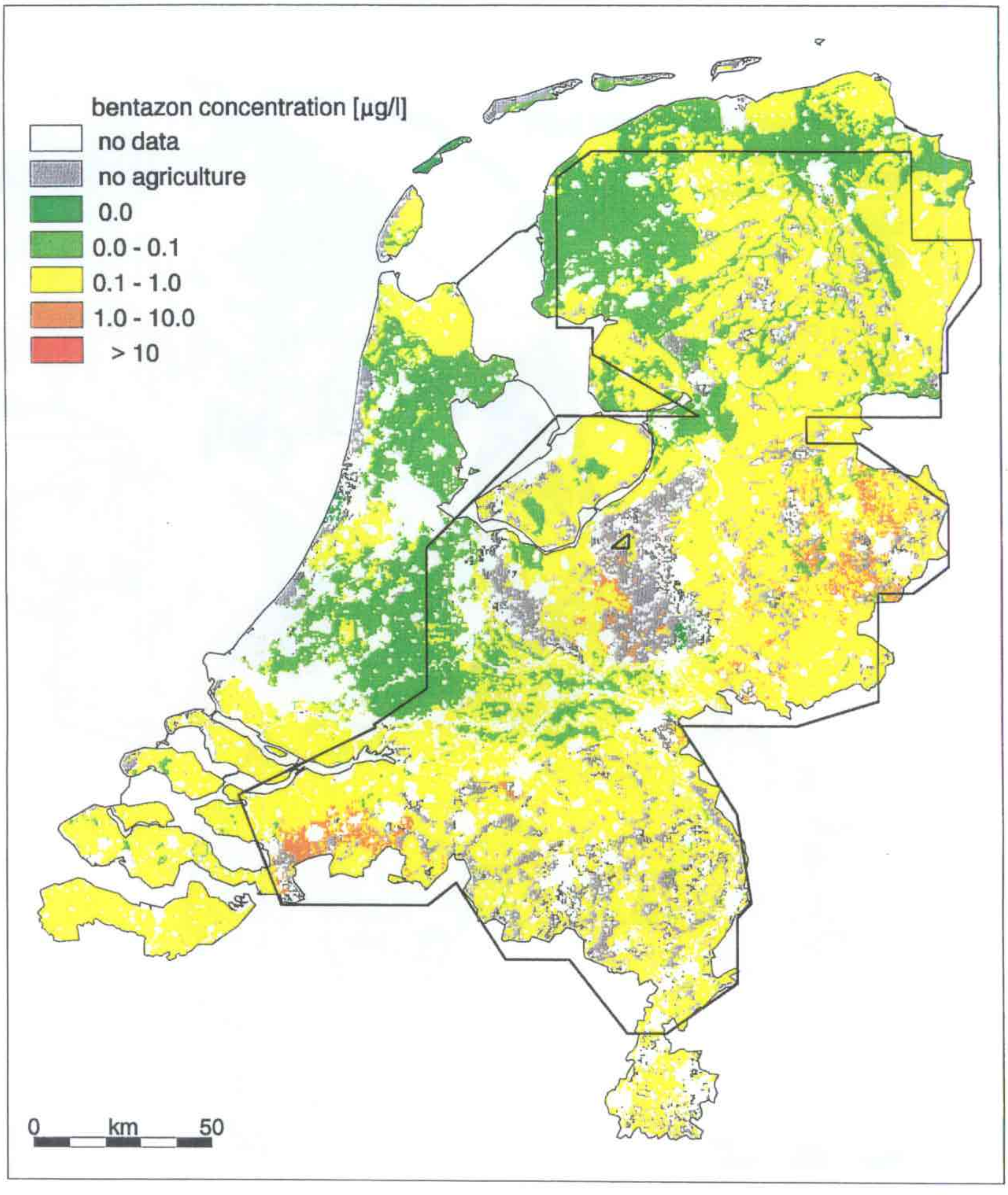


Figure 5.9 *Leaching of bentazone to shallow groundwater, historical, 1990.*

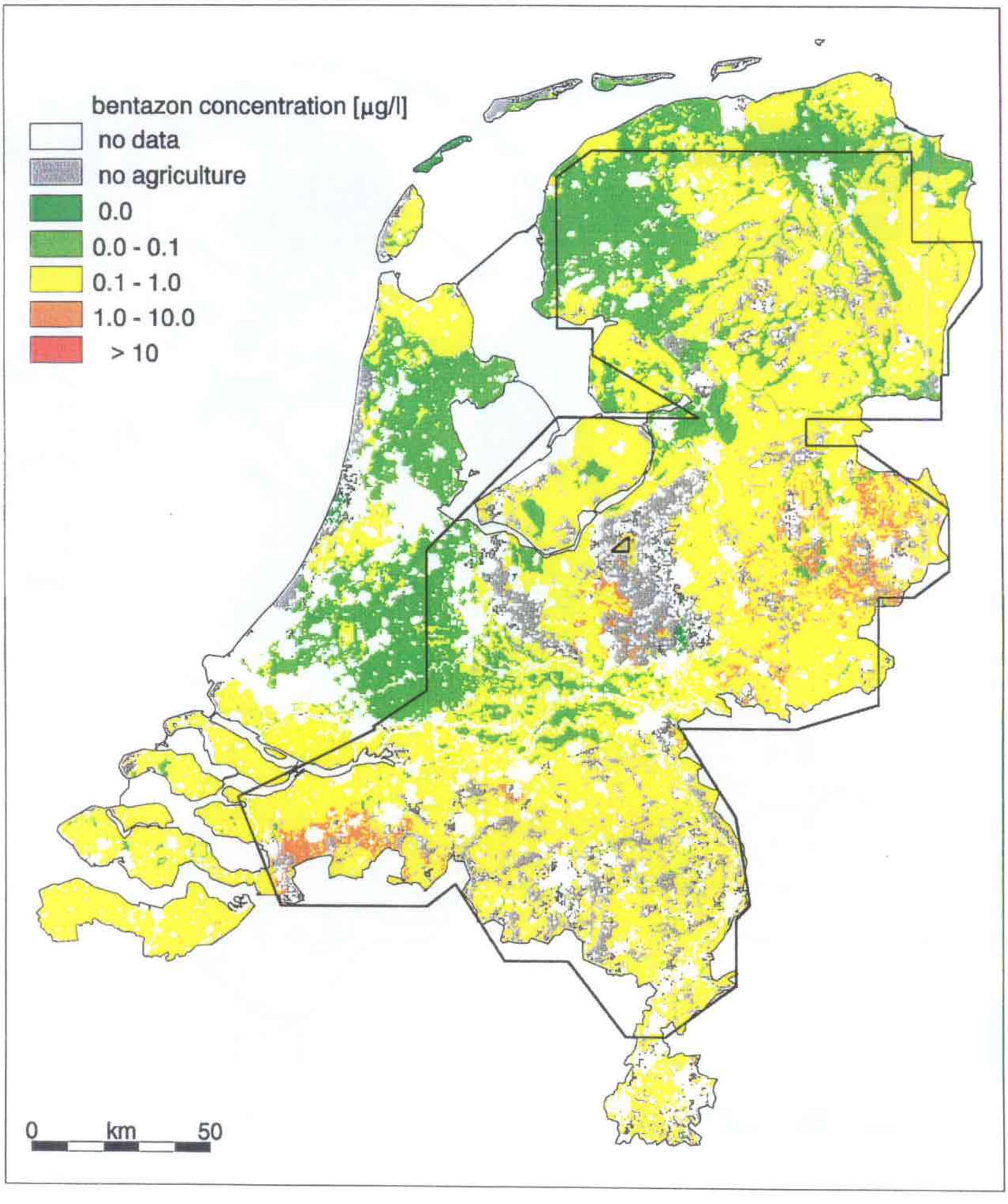


Figure 5.10 *Leaching of bentazone to shallow groundwater, historical, 1995.*

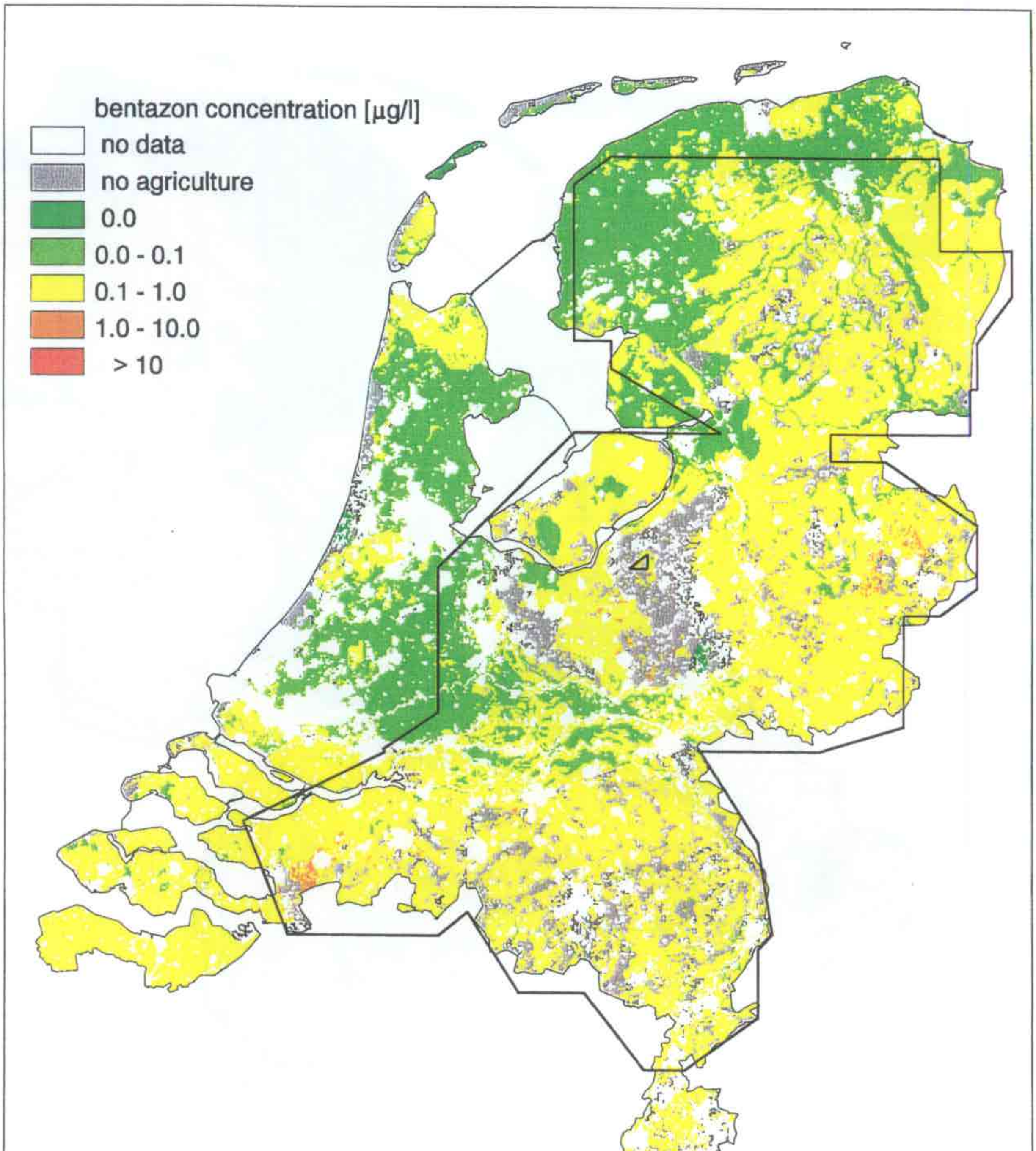


Figure 5.11 *Leaching of bentazone to shallow groundwater, scenario EC, 2000.*

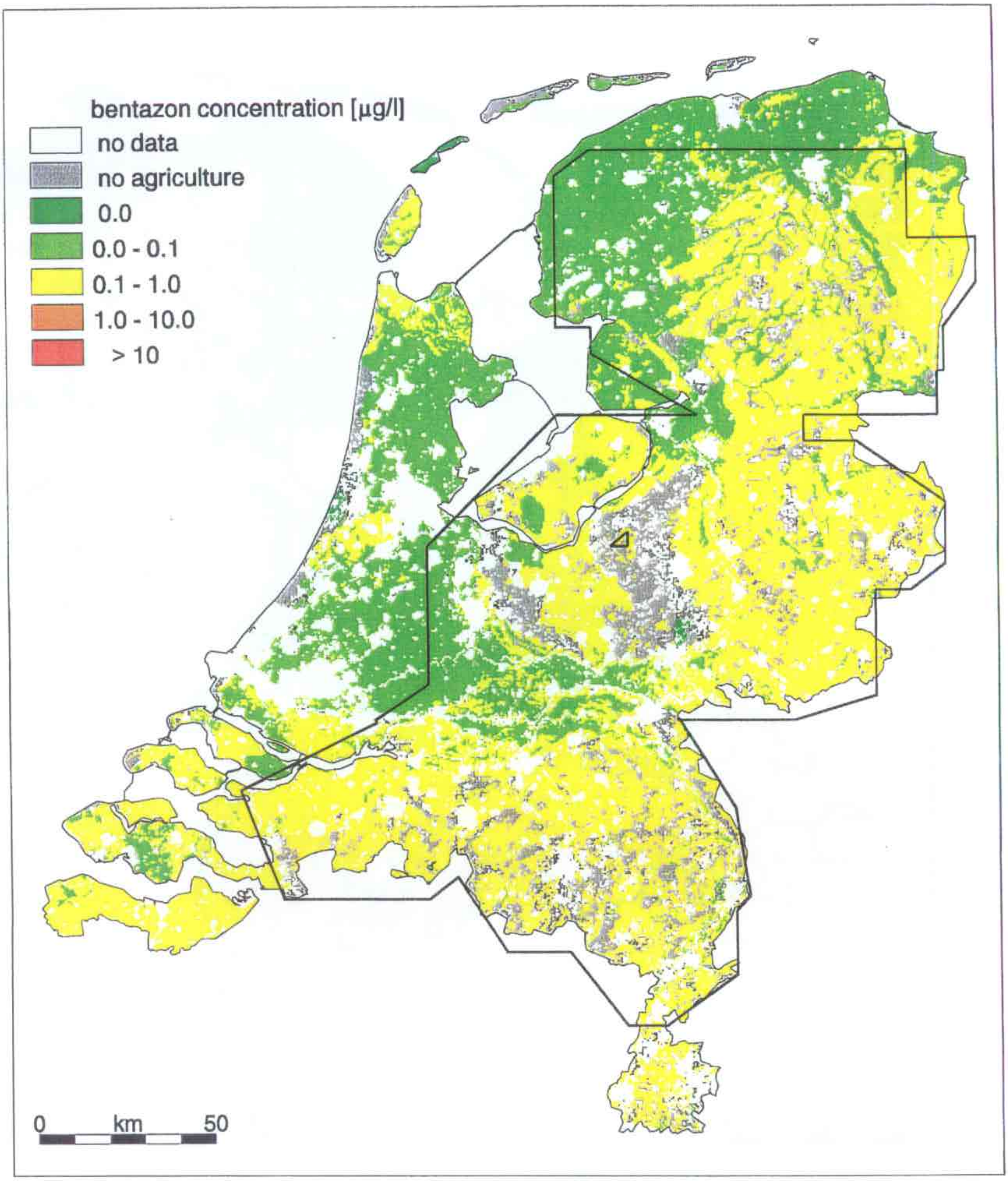


Figure 5.12 *Leaching of bentazone to shallow groundwater, scenario EC, 2020.*

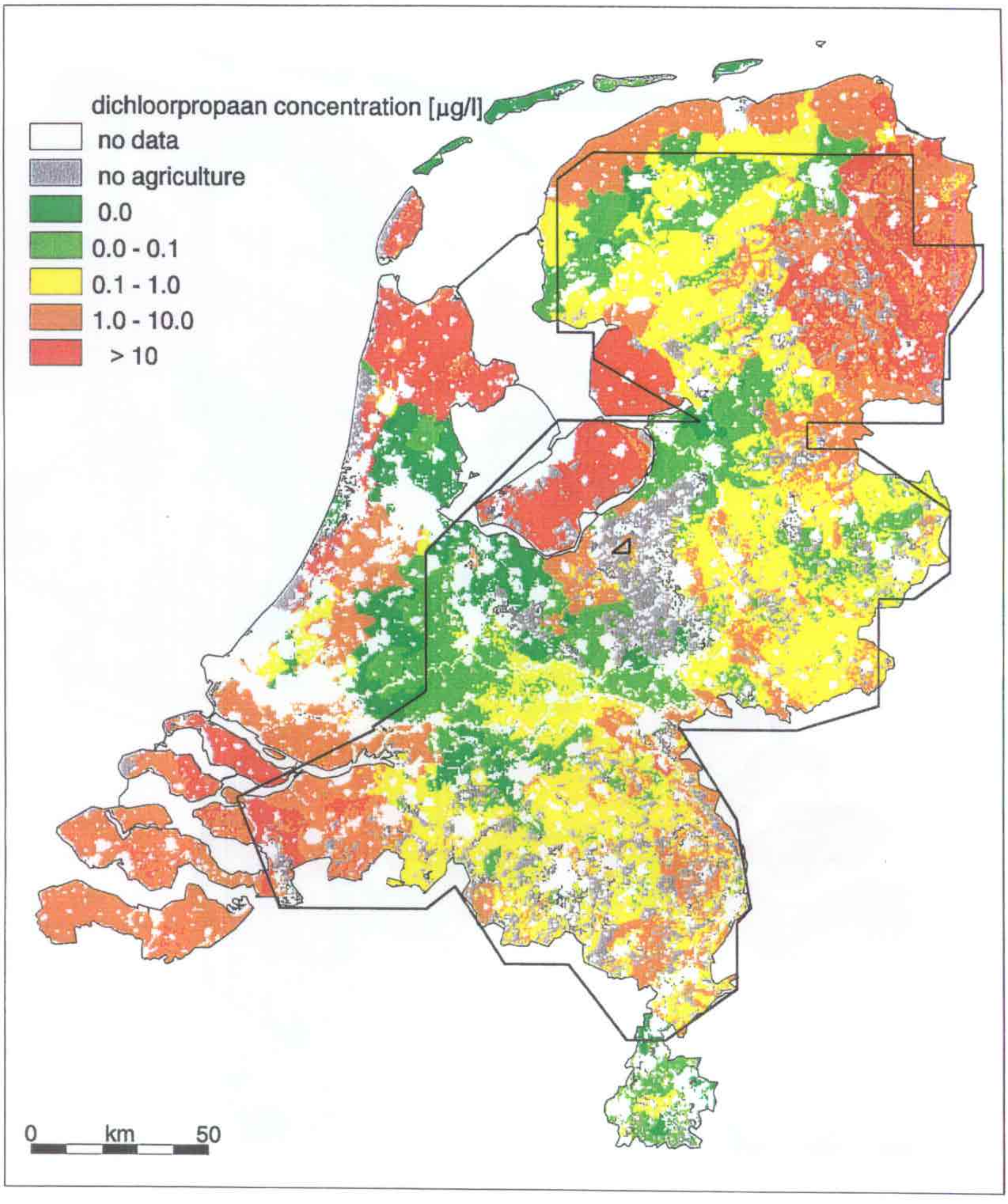


Figure 5.13 *Leaching of 1,2-dichloropropane to shallow groundwater, historical, 1960.*

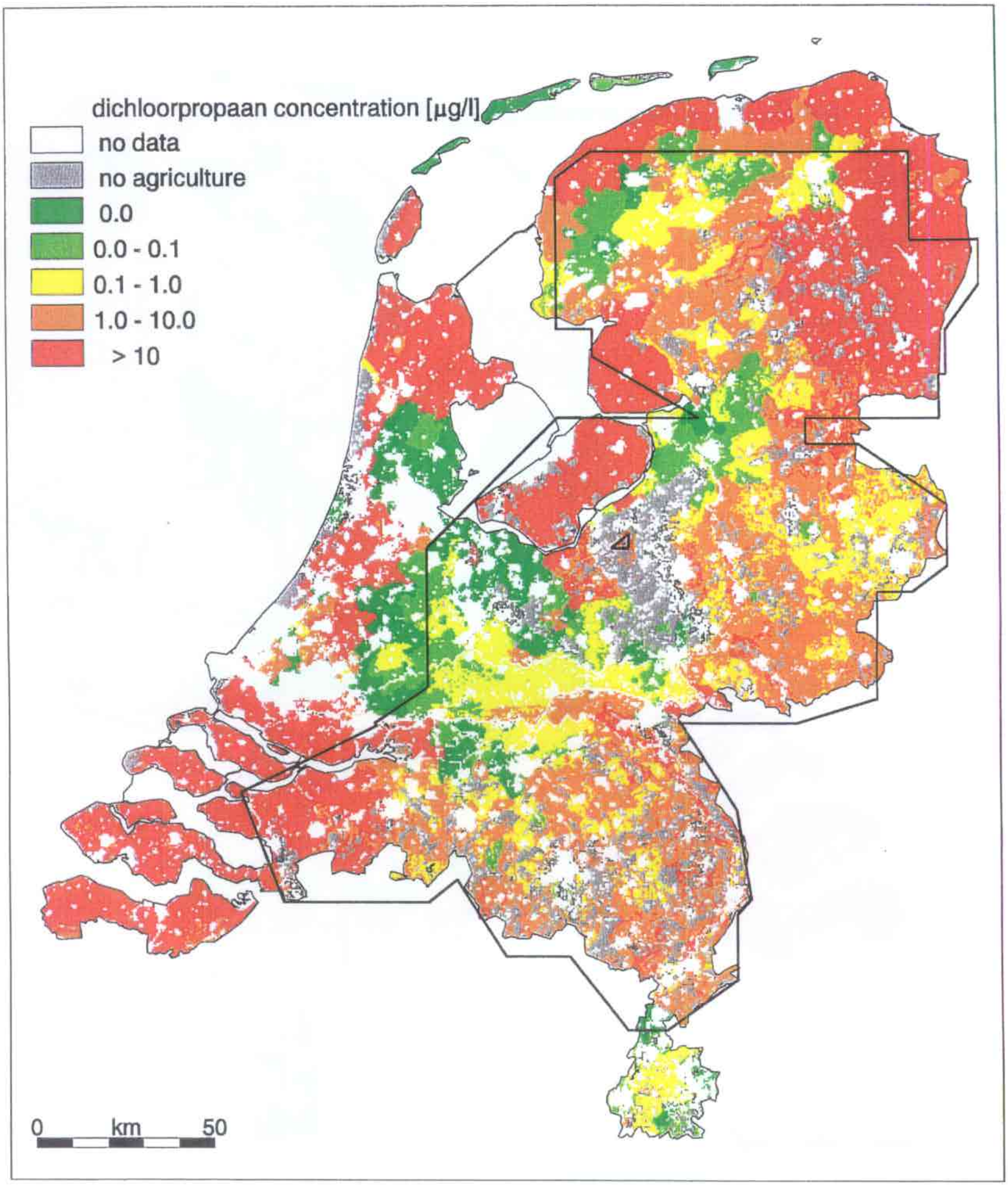


Figure 5.14 *Leaching of 1,2-dichloropropane to shallow groundwater, historical, 1970.*

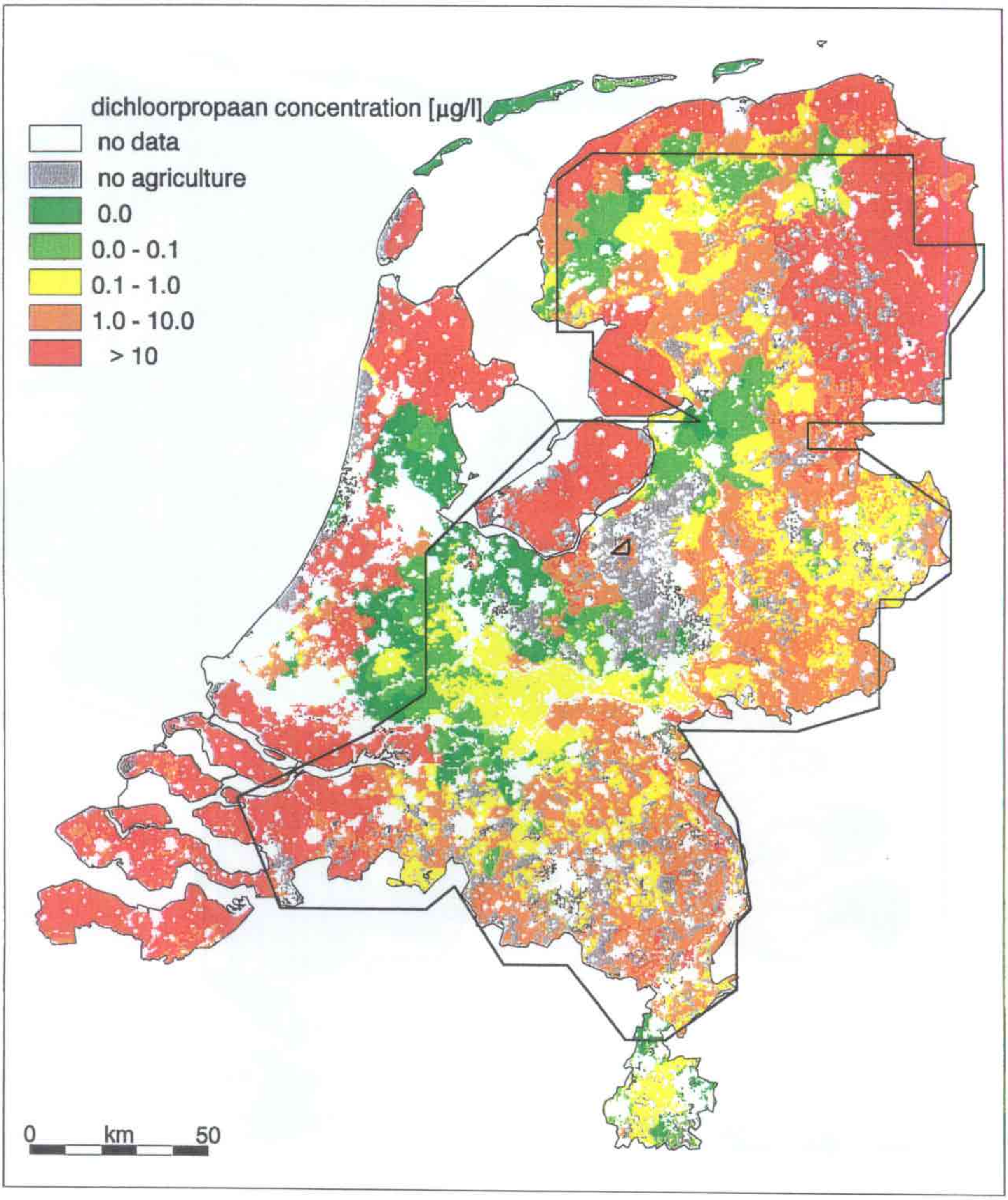


Figure 5.15 *Leaching of 1,2-dichloropropane to shallow groundwater, historical, 1980.*

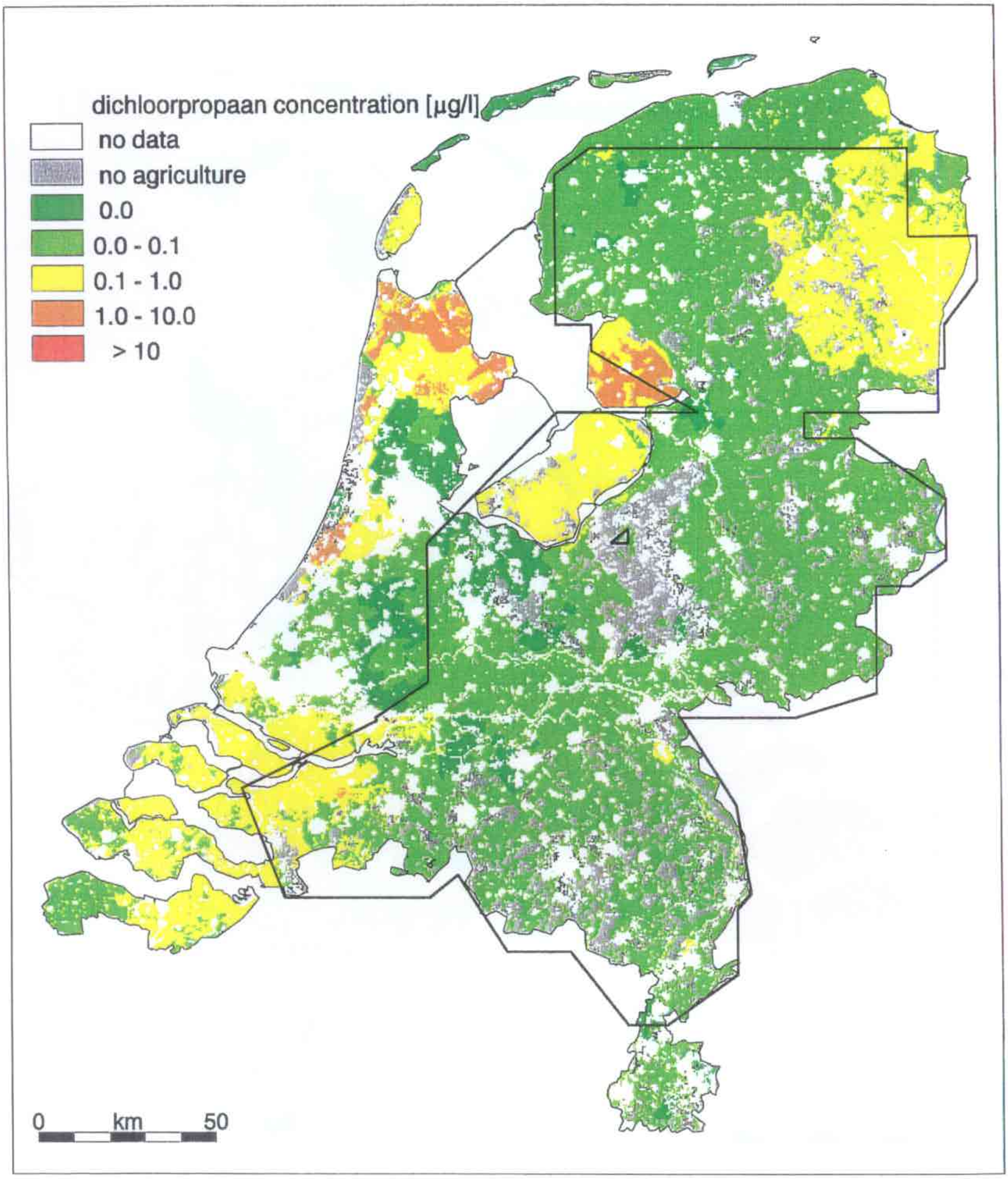


Figure 5.16 *Leaching of 1,2-dichloropropane to shallow groundwater, historical, 1990.*

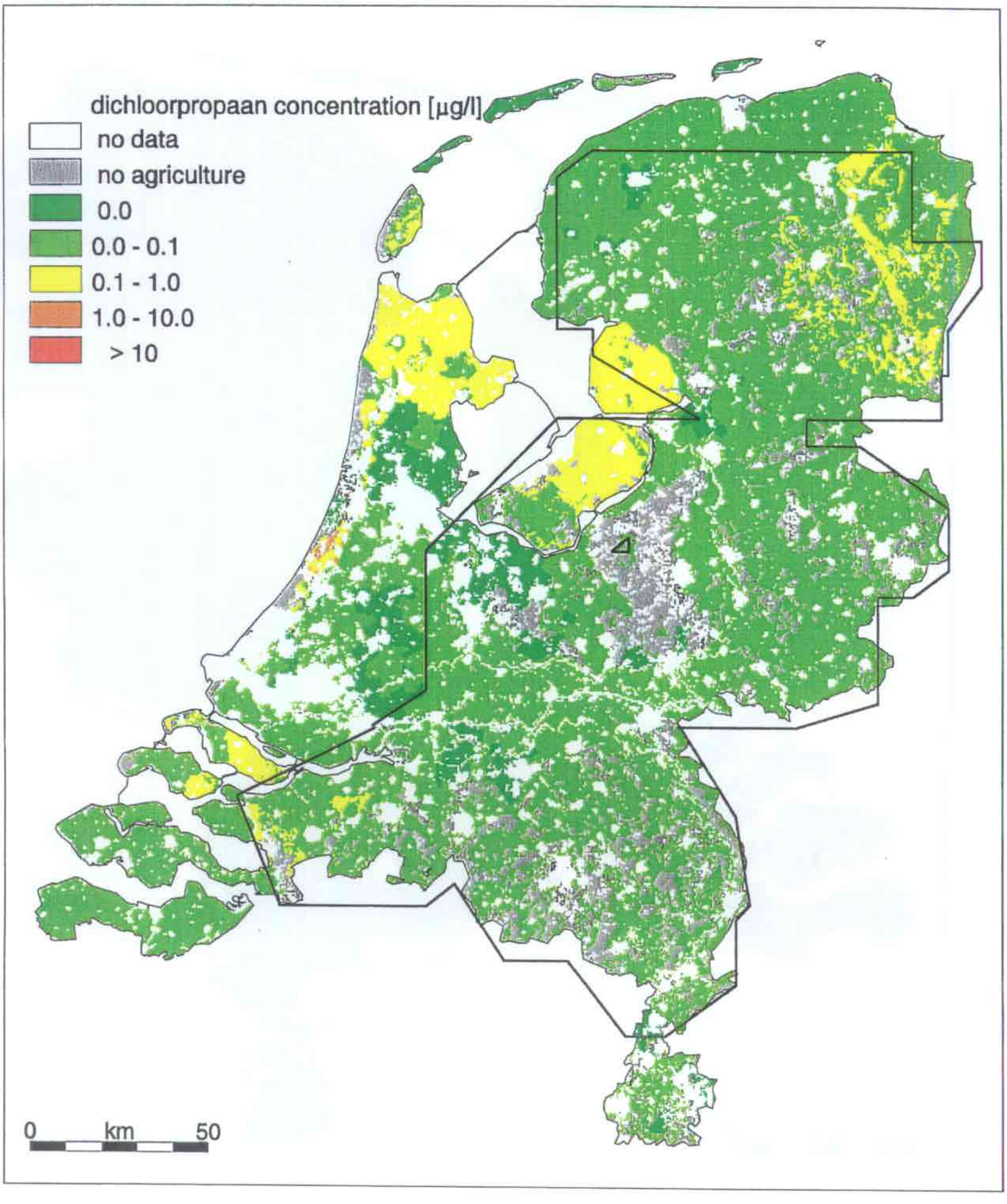


Figure 5.17 *Leaching of 1,2-dichloropropane to shallow groundwater, historical, 1995.*

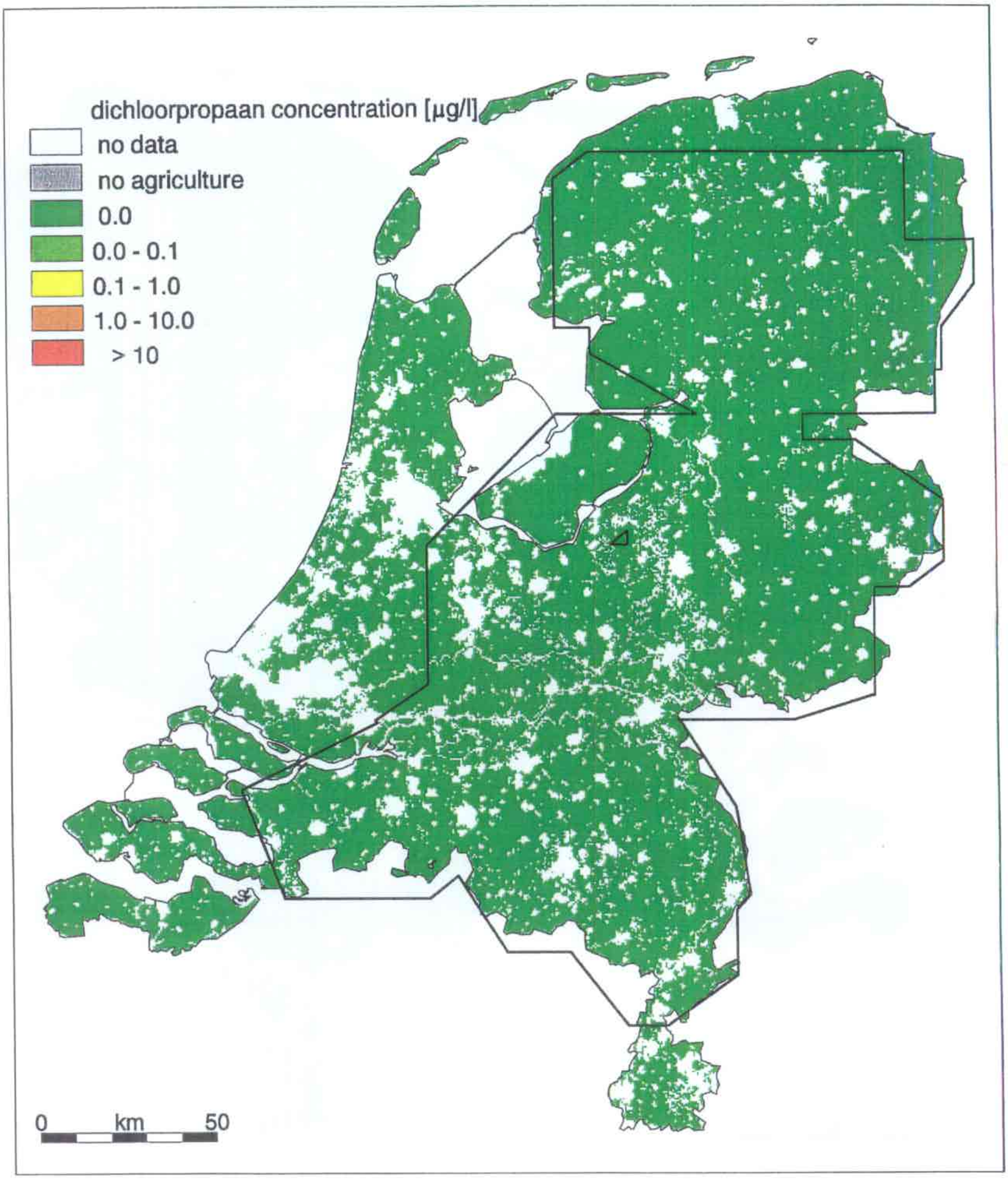


Figure 5.18 *Leaching of 1,2-dichloropropane to shallow groundwater, scenario EC, 2000 and 2020.*

6. MODELLING PATHLINES AND TRAVEL TIMES

6.1 Introduction

This chapter describes the modelling of pathlines and travel times in saturated groundwater, using the module LGMFLOW, as necessary for the calculation of concentration in abstracted groundwater (Appendix E and Chapter 7).

In this part of the study, the module LGMFLOW was used to generate pathlines and travel times. One of the input items for LGMFLOW are the maps of groundwater heads in aquifers, the fluxes across the aquitards, and the fluxes between the top aquifer and the top system and the top aquifer and rivers. This input data was generated previously using the module LGMSAT (Chapter 2).

The pathlines and travel times were generated for most *phreatic* and *semi-confined* locations for drinking-water supply (section 2.3). The abstraction locations in the vicinity of large rivers (*river system*) have not been considered.

Appendix B lists the data for the 165 abstraction locations for which pathlines and travel times were generated. The data is given separately for each of 15 refined model areas. The data in Appendix B is a subset of data in Appendix A. The following abstraction sites, originally available in Appendix A do not occur in Appendix B:

- abstractions which are not located in the 15 refined models;
- abstraction locations labelled as *river system*;
- abstraction locations for which the calculated control breakthrough curves (Appendix E) were not suitable. This occurred (a) for deep semi-confined abstractions, for which the travel time was extremely long, (b) for abstractions which received relatively too large portion of their rate from the model boundary, instead of having their source within the model area, and (c) for semi-confined abstraction locations in the vicinity of large rivers.

The abstraction locations listed in Appendix B concern the locations selected for calculation of pathlines, travel times and concentrations, as post-processing of the groundwater potential results previously produced by LGMSAT. Evidently, as all factors which influence the geohydrology must be taken into account, LGMSAT uses as input not only <u>all</u> drinking-water abstractions, as listed in Appendix A, but also the abstractions for industrial purposes (sections 2.5 and 2.6).

In Appendix B, each abstraction site is characterized, among other items by the number of aquifers where abstraction takes place. As the geohydrological system in LGM consists of four aquifers, the number of well screens can vary between 1 and 4. Appendix B contains also the sequence number of aquifers where abstraction takes place. This is because in most cases the actual well screen cuts across a number of aquifers. The aquifer numbers vary between 1 (top aquifer) and 4 (the deepest aquifer). As an example, the abstraction LGM-no.=720, nieuw-loosdrecht, in model *u2*, takes place in two aquifers, 2 and 3.

Table 6.1 summarizes the data from Appendix B. The total mumber of abstractions for calculation of pathlines, travel times and concentration breakthrough curves is 165.

Table 6.1 *Summary of drinking-water abstractions used for concentration calculation (permit 1988).*

	number abstractions	total rate (m^3 $year^{-1}$)
phreatic abstractions	76	282.10×10^6
semi-confined abstractions	89	457.66×10^6

The calculation of pathlines by LGMFLOW is based on the following approximations:

(a) the *x*- and *y*-components of the groundwater velocity in an aquifer are independent of the depth (*z*-coordinate);
(b) the groundwater velocity in *z*-direction in an aquifer is calculated by linear interpolation of the vertical velocity at the top and bottom of the aquifer;
(c) the vertical velocity in an aquitard is assumed constant, no horizontal flow there.

The LGMFLOW module was used in two modes: (a) *forward tracking* mode, needed for calculation of concentration breakthrough curves (section 6.2), and (b) *backward tracking* mode, needed for generation of pathlines (section 6.3).

6.2 Pathlines from Top of the System

LGMFLOW was used to calculate pathlines and travel times in the forward tracking mode, i.e. when particles move in the same direction as groundwater flow. The pathlines start at the top of the system, from the top-grid starting-points. The *z*-level of these points is either ground level or, if groundwater head in the top aquifer is below the top of that aquifer, the phreatic level (in the top aquifer). In *x*/*y*-space, the top-grid starting-points were located in the middle of square-shaped cells, so-called ARC/GRID cells. The size of the ARC/GRID cells, denoted as CELLSIZE, is 250 × 250 m. The number of cells in *x*- and *y*-directions is denoted as NCOLS and NROWS, respectively. The geometry of ARC/GRID model area is illustrated in Figure 6.1. It should be noted that there is no functional relationship nor dependency between the location and density of (a) the ARC/GRID cells and (b) the finite-element base grid. In our case, as a matter of coincidence, the size of both the ARC/GRID cells and the base grid was identical, namely 250 × 250 m.

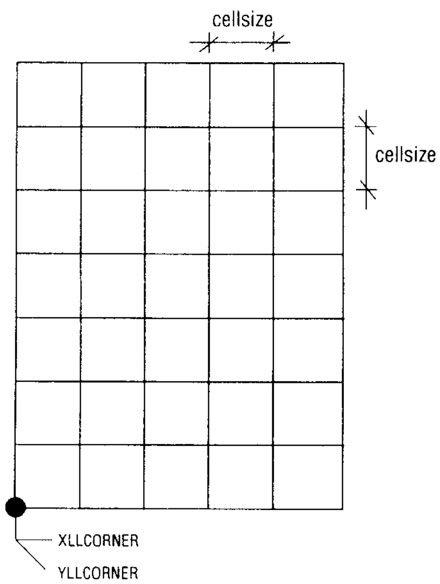


Figure 6.1 *Geometry of the model area for forward tracking from top-grid starting-points.*

In each of the NCOLS × NROWS cells, the number of top-grid starting-points, denoted by variable ICF, is defined. The ICF starting-points are uniformly distributed over the area of the ARC/GRID cell. The possible options are shown in Figure 6.2 (ICF= 1, 4, 9 or 16). In this study, one top-grid starting-point per cell (250 × 250 m) was used, i.e. ICF=1.

The top-grid starting-points are located over the entire area of each refined model. At the end of particle tracking, the final "destination" of each pathline is known. The forward pathline tracking is terminated if:

- the maximum user-specified tracking time was exceeded;
- the pathline ended in an abstracting well;
- the pathline ended in a draining river;
- the pathline ended on the periphery of an aquifer (outward boundary flow);
- the pathline ended on top of the model system (upward flow, drainage flux).

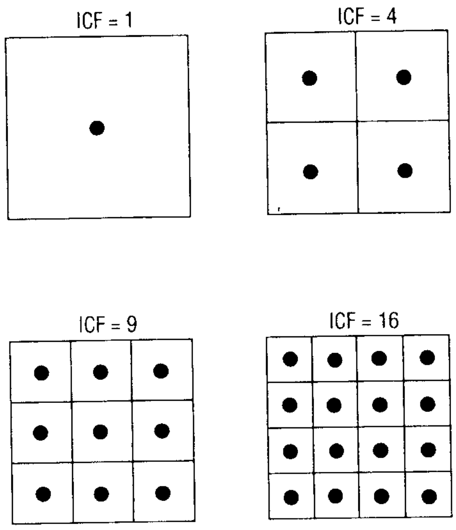


Figure 6.2 *Options for location of top-grid starting-points.*

The maximum tracking time used for forward tracking varied per refined model area, i.e. it was either 5000 or 9000 years. The value of 9000 years was used in model areas with deeply located, semi-confined abstractions, such as in the model area *eindhoven* (model code *e2*).

The capture zone of an abstraction well is an area formed by those top-grid starting-points that have been captured by the given abstraction well. The resulting capture zones for 165 abstraction locations (drinking-water supply) are presented in Appendix C. The full information about the 165 abstractions is contained in Appendix B.

Based on the results given in Appendix C the following characterizations can be made:

- groundwater abstractions from *phreatic systems*, i.e. where the hydraulic resistance of aquitards (including the top system) is relatively low, are characterized by:
  (a) a more-or-less contiguous (non-fragmented) capture zone,
  (b) a capture zone that starts at the abstraction location, and has a rather smooth (convex) shape (often unilaterally oriented or ellipse-shaped), and
  (c) a relatively high fraction of the area where travel time is less than 25 years (red dots).

- groundwater abstractions from *semi-confined systems*, i.e. where the hydraulic resistance of aquitards (including the top system) is relatively high, are characterized by:

(a) a discontinous (fragmented) capture zone, in most cases, e.g. as patches of sub-capture zones;
(b) a capture zone that sometimes is strongly dislocated with respect to the abstraction location itself, and has an uneven (concave, rugged) shape, and
(c) a relatively high fraction of the area where travel time is greater than 50 years (green dots).

Examples of groundwater abstractions from typically *phreatic systems* are:
- refined model *utrecht* (model code *u2*): LGM-no. = 747, soestduinen
- refined model *utrecht* (model code *u2*): LGM-no. = 737, beerschoten
- refined model *utrecht* (model code *u2*): LGM-no. = 749, zeist

Examples of groundwater abstractions from typically *semi-confined systems* are:
- refined model *utrecht* (model code *u2*): LGM-no. = 720, nieuw-loosdrecht
- refined model *utrecht* (model code *u2*): LGM-no. = 739, bunnik
- refined model *utrecht* (model code *u2*): LGM-no. = 741, cothen

Examples of groundwater abstractions from *semi-confined systems* where groundwater (partly) originates from *rivers* are:
- refined model *utrecht* (model code *u2*): LGM-no. = 722, linschoten
- refined model *utrecht* (model code *u2*): LGM-no. = 787, nieuwegein("totaal")
- refined model *utrecht* (model code *u2*): LGM-no. = 843, lexmond(delaak)

As noted before, for groundwater abstractions from *semi-confined systems*, the capture zone is sometimes strongly dislocated with respect to the abstraction location itself. Typical examples of this feature are:
- refined model *veluwe* (model code *v2*): LGM-no. = 194, twello
- refined model *veluwe* (model code *v2*): LGM-no. = 144, fikkersdries
- refined model *eindhoven* (model code *e2*): LGM-no. = 523, oirschot
- refined model *eindhoven* (model code *e2*): LGM-no. = 494, eindhoven-groote.heide

In a number of cases it can be seen that the model area was too small to include the whole capture zone, such as for:
- refined model *polder* (model code *p2*): LGM-no. = 10136, harderwijk_I
- refined model *polder* (model code *p2*): LGM-no. = 58, fledite
- refined model *eindhoven* (model code *e2*): LGM-no. = 494, eindhoven-groote.heide

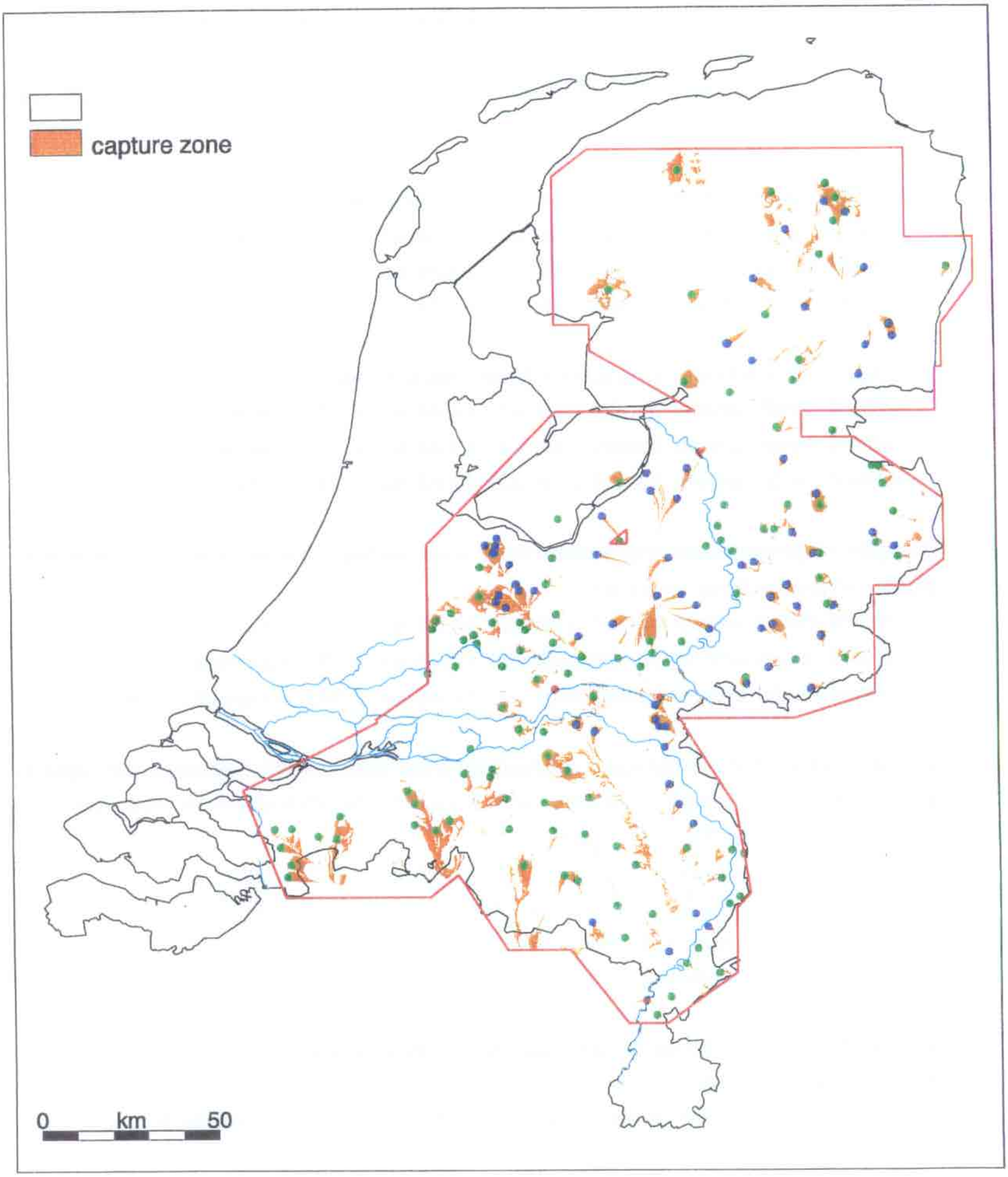


Figure 6.3 *Capture zones calculated within 15 refined model areas, with the location of all abstractions for drinking-water supply (blue:phreatic, green:semi-confined, red:river system).*

The capture zones depicted in Appendix C could be used, for example, for delineating the well-head protection areas. In this study, the capture zones were used as background information for understanding the shape of the control breakthrough curves, as discussed in Appendix E.

The capture zones are summarized in Figure 6.3. The orange-coloured region represents pathline locations from which a well is reached. Figure 6.3 contains the capture zones for the 165 abstractions from Appendix C. However, Figure 6.3 also contains the capture zones for the *river-system* abstractions.

The total area of the 165 capture zones is 1703 km^2. This area was calculated as the sum of the 250 × 250 m cells from which a pathline ended in an abstraction well.

6.3 Pathlines from Well Screens

Subsequently, LGMFLOW was used to calculate pathlines and travel times in the so-called backward tracking mode, i.e. when particles move in the direction opposite to the groundwater flow. Unlike the forward tracking pathlines, the backward tracking pathlines were not used for the subsequent calculation of concentration breakthrough curves. The backward tracking pathlines were used for a better understanding the flow patterns, both around a specific abstraction location and in a bigger area (regional flow).

The backward tracking pathlines start at points along lines which are uniformly distributed around a well location. The current concept for locating starting-points around a well in a finite element is depicted in Figure 6.4. The example concerns a quadrilateral element. It is important to note that wells, in general, do not coincide with nodes but can be located anywhere in an element. In the example, four starting-points are used, i.e. the uniform angle is 90 degrees. The starting-points are located at a certain distance beyond the boundary of the element in which the well is located.

Galerkin finite-element discretization is used to generate groundwater velocity fields in x- and y-directions. The velocities are continous across the element boundaries. As wells are not located in grid nodes, the groundwater velocities in the direct vicinity of wells can show an irregular pattern. This is inherent to the discrete nature of the finite-element method. In principle, the irregularities are smaller if a well is located close to a node. With the current procedure, the irregularities in the velocity field can cause pathline tracking to take place in a direction other than is expected (mostly in the opposite direction). This occurs only for certain wells and for certain pathline starting-points along a well. As the current grid generator, LGMGRID, does not generate nodes at well locations, the pathline tracking problem could be alleviated by (a) locating the starting-points in a circle (constant radius) around a well in combination with (b), using a denser finite-element grid around a well.

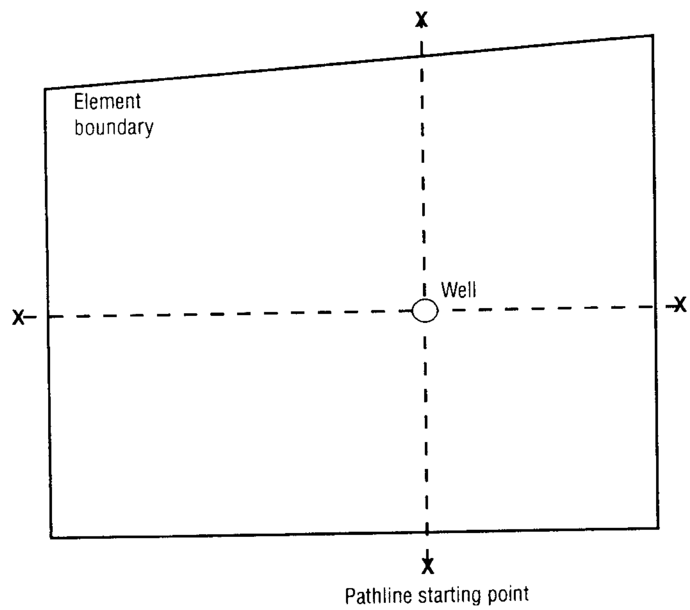


Figure 6.4 *LGM-2 concept for locating starting-points around a well in an element.*

The z-level of the starting-points is at the well-screen bottom. The number of starting-points around a well screen is set at 90. Recall that well screens are assumed to be located across the full depth of an aquifer. If an abstraction takes place in more than one aquifer, the "ring" of pathlines starts at the bottom of each aquifer, i.e. at the bottom of each well screen. The numbers of aquifers where abstraction takes place are listed in Appendix B.

Since the backward tracking pathlines were not used for calculation of concentrations, as an example, only 10 pathlines patterns are presented in Appendix D. For a better understanding of the flow patterns, Appendix D also includes the relevant capture zones for the convenience of the reader. The capture zones are identical to those presented in Appendix C.

As is explained in section 6.1, the pathline calculation is based on (a) groundwater velocity in x/y-plane, constant for each depth within the thickness of an aquifer, (b) linear interpolation of z-velocity within the thickness of an aquifer, and (c) constant z-velocity across aquitards. From this it follows that when starting at different level in an aquifer, the resulting pathline pattern is, in principle, different in each case. For example, for an abstraction from a single phreatic aquifer, the shape of the area enclosed by a pathline pattern will remain more-or-less the same, and the size of the area will increase with the increasing depth of the starting level. However, when starting from a multi-well-screen abstraction in a multi-aquifer system, where there are significant hydraulic resistances of aquitards, the pathline patterns for each well screen will, in principle, be different in both shape and location. These effects are illustrated by the 10 examples in Appendix D.

6.4 Evaluation

The particle-tracking method currently used leads to good results for pathlines and travel times, both in forward and backward tracking mode. However, in some cases, due to the discrete nature of the finite-element discretization, problems can occur in the immediate vicinity of wells. The probability that problems will occur is smaller if a well is located closer to a node. A solution to this problem would be (a) to locate the starting-points in a circle (constant radius) around a well, in combination with (b) using a denser finite-element grid around a well.

The density of the base grid used for the 15 refined models was 250 × 250 m, based on the conclusion of the study by Kovar *et al.* (1996). This states that when using LGM for calculating pathlines, travel times and breakthrough curves for phreatic and semi-confined abstractions, it is required to use the base grid element size of 250 × 250 m in the entire capture zone.

The accuracy of pathlines and travel times in the vicinity of wells would further increase if the finite-element grid around abstraction locations were locally refined. The refinement could be, for example, 100 × 100 m within the radius of 200 m around each well. The grid refinement would have a positive effect, not only on backward pathline tracking but also on forward pathline tracking, specifically for the algorithm for capturing pathlines in a well consisting of multiple-aquifer well screens. Currently, due to the relatively coarse grid around a well (250 × 250 m), some forward pathlines can end up in a wrong well screen, i.e. either in a well screen above or below the well screen where the pathline would have ended if a higher grid resolution were used. The above-mentioned errors in pathline destination cause slight deviations in the shape of the control breakthrough curves (Appendix E) for the specific well screens in a multiple-aquifer system and thus also for the control breakthrough curve for the total abstraction.

7. MODELLING CONCENTRATIONS AT ABSTRACTION WELLS

7.1 Introduction

This chapter discusses the calculation of concentration breakthrough curves of nitrate and pesticides at abstraction locations. The calculation of concentration variation of nitrate and pesticides in abstracted groundwater is, in a way, the core of this study.

Before calculating the concentration variation, so-called control breakthrough concentration curves were generated. A control breakthrough curve is the concentration variation in time in abstracted groundwater, resulting from the solute load at the system top, specified as an instantaneous increase of concentration from 0 to 100 concentration units at the zero time. The solute input concentration 100 is spatially constant over the entire model area and the initial concentration within the groundwater system is assumed to be zero. The method and the results for control breakthrough curves are documented in Appendix E.

The control breakthrough concentrations and the concentration of nitrate and pesticides were calculated by the module LGMCAM. The functionality of LGMCAM is discussed in Appendix E (section E.1).

A solute load on the system top (i.e. on top of saturated groundwater) that can be variable in both space and time forms a part of the LGMCAM concept. The input concentration of nitrate and the three pesticides was specified at the following eight time instants:
1) six for *historical solute load*: 1950, 1960, 1970, 1980, 1990, 1995;
2) two for *scenario (future) solute load*: 2000, 2020.

The (economic) scenarios, *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*) are described in Chapter 3. For nitrate, use is made of the so-called scenario *EC(plus)*. The leaching fluxes of nitrate and pesticides are described in Chapter 4 and 5, respectively. The concentrations of nitrate and pesticide leaching into saturated groundwater were calculated as constant values within 500 × 500 m grid cells.

The concentration variation in abstracted groundwater is calculated for a 1000-year period, starting in 1950. The calculation is done for each solute, at each of 165 abstraction locations (76 phreatic, 89 semi-confined). Evidently, for practical reasons, it is not possible to present all concentration breakthrough curves in this report. Therefore the results will be presented in a number of maps, for a selected number of time instants. However, in addition to those maps, this report also includes a number of concentration breakthrough curves for the purpose of illustration. This is done for 14 selected abstraction sites:

1) refined model *utrecht* (model code *u2*): LGM-no. = 745, groenekan;
2) refined model *utrecht* (model code *u2*): LGM-no. = 749, zeist;
3) refined model *utrecht* (model code *u2*): LGM-no. = 739, bunnik;
4) refined model *veluwe* (model code *v2*): LGM-no. = 144, fikkersdries;
5) refined model *achterhoek* (model code *a2*): LGM-no. = 26, gorssel-tJoppe;
6) refined model *achterhoek* (model code *a2*): LGM-no. = 139, noordijkerveld;
7) refined model *achterhoek* (model code *a2*): LGM-no. = 136, lochem;
8) refined model *achterhoek* (model code *a2*): LGM-no. = 36, vorden.(dennenwater);
9) refined model *achterhoek* (model code *a2*): LGM-no. = 10124, olden-eibergen;
10) refined model *yssel* (model code *y2*): LGM-no. = 175, espelose-broek-links;
11) refined model *drenthe* (model code *d2*): LGM-no. = 19, valtherbos;
12) refined model *drenthe* (model code *d2*): LGM-no. = 16, noordbargeres;
13) refined model *limburg* (model code *l2*): LGM-no. = 370, ospel;
14) refined model *eindhoven* (model code *e2*): LGM-no. = 551, son.

The selection of the above listed 14 abstraction sites was done using a number of criteria: (a) phreatic and semi-confined, (b) fast and slow response, (c) the number and location of well screens, and (d) the possibility of occurrence of relatively high concentration at the site.

7.2 Results for Nitrate

The variation of nitrate concentration in time was calculated for 165 abstraction locations (76 phreatic, 89 semi-confined). Use is made of a single scenario, namely *EC(plus)*. Appendix F contains, as an example of the results, the concentration breakthrough curves for 14 selected abstraction sites. Figures 7.1 and 7.2 show the spatial distribution of nitrate concentration calculated for 2020 and 2050, respectively. The nitrate concentrations 25 and 50 mg NO_3 l^{-1} are used in the Netherlands as guide value concentration and maximum admissible concentration, respectively.

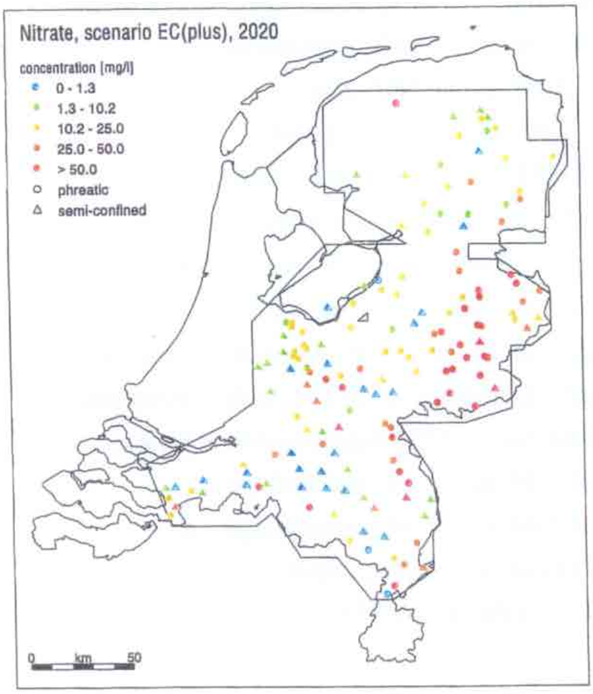


Figure 7.1 *Nitrate concentration in abstracted groundwater, scenario EC(plus), 2020.*

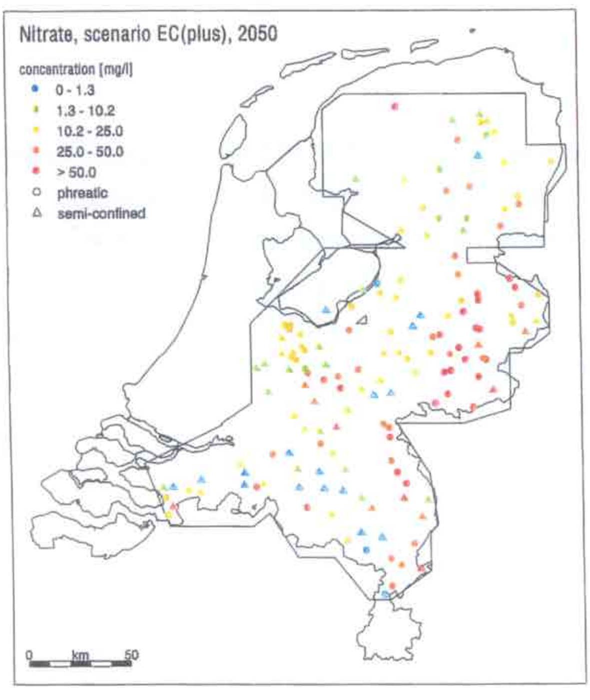


Figure 7.2 *Nitrate concentration in abstracted groundwater, scenario EC(plus), 2050.*

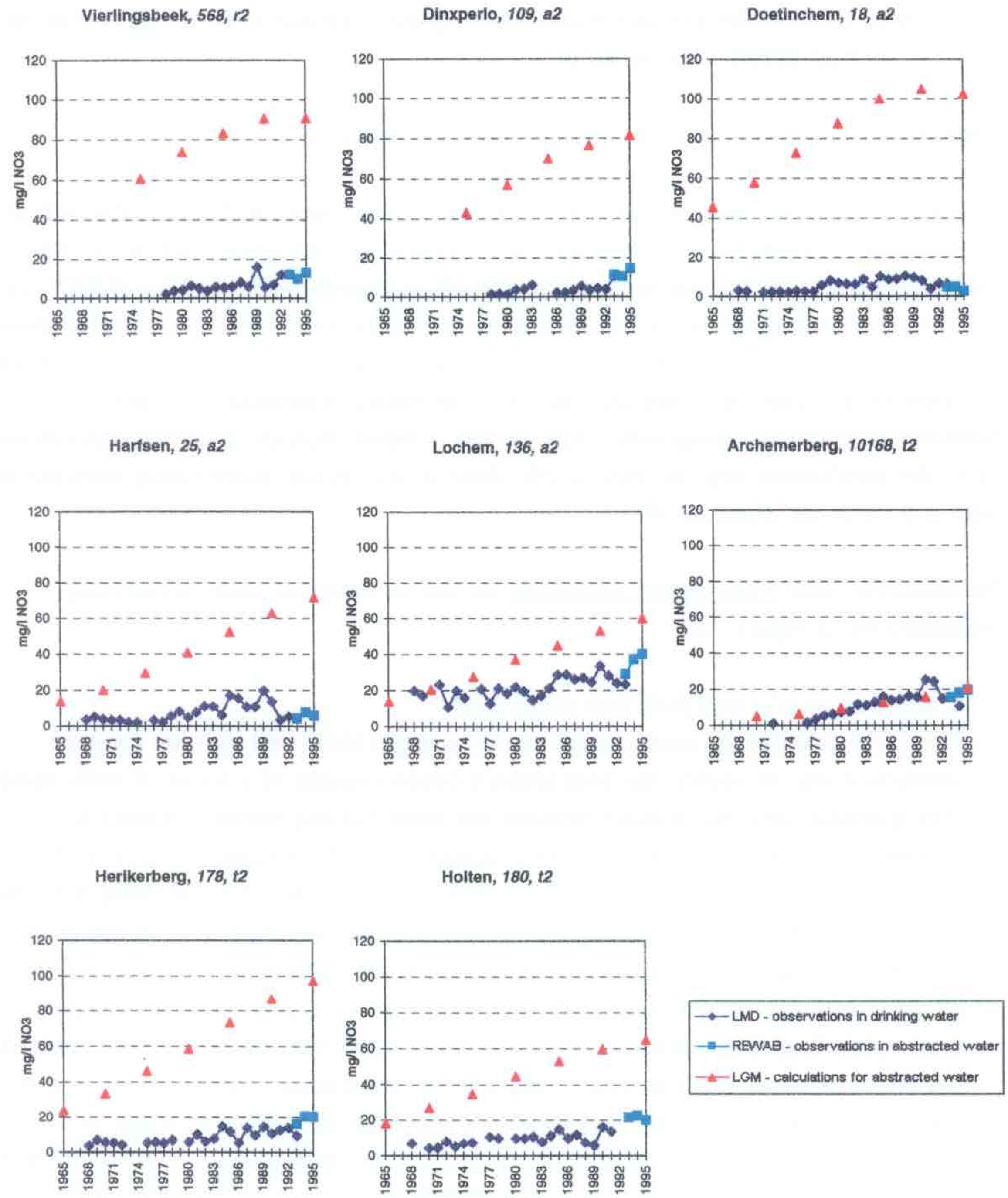


Figure 7.3 *Comparison of calculated and observed nitrate concentrations for selected abstraction sites (name, LGM-no., refined model area code).*

According to the calculation results shown in Figures 7.1 and 7.2, a large number of groundwater abstractions, and consequently the related groundwater-based drinking-water production in Oost-Gelderland and Noord-Limburg will continue to be threatened in the future by too high nitrate concentrations.

Comparison of calculated and observed nitrate concentrations

In Figure 7.3, the calculated nitrate concentrations are compared with those observed at eight abstraction locations. The observed values consist of two series, namely (a) the observations from the national monitoring network of drinking-water quality (LMD), i.e. after the abstracted groundwater has undergone the purification, and (b) the observations from the water-quality registration system of drinking-water supply companies (REWAB). The latter system came into operation in 1993, the values expressing the quality of abstracted groundwater. Essentially, there is only a minor decrease in nitrate concentration due to the purification step. In other words, there is only minor discontinuity between the LMD and REWAB series in 1993.

The following three hydrological limitations should be observed when interpreting concentrations in Figure 7.3:

1) *Simplification of a well field into single well:*
   The LGM calculations were carried out for a single ideal well location for each abstraction site. In reality, the abstraction location consists of a series of wells spread over a certain area, the distance between the wells varying between dozens to hundreds of metres. The idealized well location in LGM is supposed to be at the "centre of gravity" of the actual well locations, the well rate in LGM being the total of all (small) well rates. The schematization into a single well location introduces an error in the resulting calculated concentrations. If the calculation were carried out with all wells acting at an abstraction site, the impact of small parcels with a relatively large solute load (e.g. nitrate from maize) would have been modelled more accurately. On the other hand, small parcels would also be encountered with relatively low solute load, thus "counterbalancing" the impact of small parcels with peak loads. It is assumed that the error introduced by using one-well representation is relatively small. Moreover, one can recall that the concentrations of nitrate and pesticides in the leaching flux is available as constant values within grid cells of 500 × 500 m. Taking into account all wells within a well field would be only meaningful if at the same time the concentration of the leaching flux would be provided at a higher resolution scale, e.g. for grid cells of 250 × 250 m.

2) *Operational changes in well locations:*
The calculated concentrations should represent the average total well rate at the "centre of gravity" for a fixed spatial configuration of wells. Similarly, the observed concentrations (LMD and REWAB) are expected to reflect the average total well rate for a fixed spatial configuration of wells. The observed values should be the "average" concentration of the mixed contribution from all wells within a well field, as modelled by LGM. However, as the nitrate content in drinking water at some abstractions became higher than the maximum admissible concentration (50 mg NO_3 l^{-1}), the water supply company has installed additional (new) wells in a relatively "clean" area. These extra wells - not accounted for in LGM - were intended to deliver groundwater of good quality, to be mixed with the poor-quality groundwater, thus producing an acceptable nitrate quality of groundwater of the entire abstraction site. This induced mixing occurred, for example, at Lochem, LGM-no. = 136, refined model area *achterhoek* (*a2*), where at the end of 1980s one extra well was placed about 1 km south-east of the existing well field. Without this extra well, the concentration at Lochem would have started increasing rather rapidly from the end of 1980s. It should be noted here that as groundwater at Lochem is aerobic, denitrification plays a minor role. The general consequence of operational changes in well locations (such as at Lochem) for this study is that at such locations the observed concentrations are lower than the concentrations that would have been observed without additional "clean" wells. These operational changes are not accounted for in LGM.

3) *Assumption of constant well rate:*
In this study, the concentration variation of nitrate and pesticides in abstracted groundwater is calculated starting from 1950. As steady-state groundwater flow is simulated, the applied abstraction rates are assumed to be constant in time from 1950. Use is made of maximum-permitted abstraction rates, as in effect for the year 1988. However, in reality the actual abstraction rates (a) were in most cases lower than the 1988 maximum-permitted abstraction rates, and (b) the actual abstraction well rate was not constant from 1950 but usually gradually increasing in time from the start of operation (mostly after 1950):

(3a) *actual abstraction rates smaller than 1988 maximum-permitted abstraction rates:*
In general, a higher well rate would lead to (i) earlier occurrence of a certain concentration level, and (ii) a deviation in the final concentrations. The first happens because with lower well rate it takes longer to withdraw the "original" (resident, old) groundwater, which is of better quality than current groundwater recharge. The latter is caused by the fact that a higher well rate implies a greater

area of the capture zone and thus the effect of solute leaching in that extra portion of the capture zone. The deviation can be both positive and negative, depending on the quality of groundwater recharge (solute leaching flux) in the extra portion of the capture zone.

(3b) *actual abstraction well rates not constant in time from 1950:*
The actual abstraction "history" is different for each abstraction location. Some abstractions have gradually increased the well rate from 1950. At other locations the abstraction rate has been maintained constant or increased stepwise from the onset of abstraction (e.g. 1960) up to now. The discrepancy between the constant 1988 maximum-permitted abstraction rates used and the actual temporal variation of abstraction rate per specific site has an effect on the accuracy of the calculated concentration variation of nitrate and pesticides in abstracted groundwater. In general, compared to reality, a higher well rate would lead to an earlier occurence of a certain concentration level. An explanation for this is given at the previous item 3a. However, the error caused by the assumption of steady-state well rates from 1950 decreases in time. It should be noted that the abstraction rates beyond the year 1995 have to be assumed as constant values anyway, being an extrapolation of the current situation.

From Figure 7.3 it follows, in general, that the calculated concentrations are higher than observed concentrations. In addition to the previously listed three hydrological limitations, the discrepancy between observed and calculated concentrations can also be attributed to two solute quality-related causes, namely (1) because the actual concentrations in nitrate leaching flux are higher (on agricultural land 1.5 to 10 times) than the calculated ones, and (b) because denitrification was disregarded. The two factors have an adverse effect on resulting concentrations. While the first one implies that with realistic leachate concentrations (higher than modelled), the resulting calculated breakthrough concentrations would be higher, the latter one (if denitrification were included) would mean that the calculated breakthrough concentrations would be lower. As the observed concentrations are higher than the calculated ones, it was concluded that denitrification, as an error source, is dominant over the underestimation of nitrate concentrations.

The following gives background information on the two solute quality related causes:

1) *Underestimation of concentration in nitrate leaching flux in agricultural areas:*
Recalled is that agricultural areas represent a major source of nitrate leaching, both in terms of percentage of the total area and the amount of nitrate leaching. The impact of

forested and urban areas is relatively small. Boumans & Van Drecht (1998) have compared the calculated output of the module NLOAD with nitrate concentrations observed in shallow groundwater in sandy soil areas in the Netherlands. NLOAD is applied for the assessment of leaching on agricultural land, i.e. not in forest and urban areas. The observation data were drawn from the Dutch national network for monitoring the effectiveness of animal waste control policy (LMM). The validation was done for grid blocks of 500 × 500 m. The results of the validation are discussed in section 4.2.1. A major conclusion is that NLOAD underestimates the actual leaching fluxes into saturated groundwater, especially at shallow groundwater tables. The actual concentration in leaching fluxes can be 1.5 to 10 times higher than the NLOAD-calculated concentration. The difference between actual and calculated concentrations depends, for instance, on the depth of the groundwater table, the error decreasing with increasing depth of the groundwater table. There are two possible reasons for the discrepancy: (a) denitrification between land surface and shallow groundwater tables, which plays, in reality, a less important role than it was assumed to have in NLOAD calculations, and (b) lack of knowledge about spatial distribution of nitrate load. The latter, serving as input for NLOAD, is available as constant value in each municipality.

2) *Denitrification in saturated groundwater:*
Another reason for the discrepancy between observed and calculated nitrate concentration can be denitrification in saturated groundwater. The effect of denitrification in "deep" groundwater on concentration of abstracted groundwater was documented in, for example, KIWA (1989) and KIWA (1991). Denitrification of nitrate in deep saturated groundwater occurs either by redox reaction between nitrate and pyrite (e.g. KIWA, 1989) or by mineralization of organic material (c.f. KIWA, 1991). Denitrification is discussed further in this section.

The only abstraction site out of the eight examples in Figure 7.3 where the calculated and observed concentrations match reasonably well is Archemerberg, LGM-no. = 10168 in refined model area *twente* (*t2*). A possible explanation could be that in this area the groundwater is aerobic, which means that denitrification cannot take place. In general, aerobic groundwater occurs below relatively thick unsaturated zones.

The second-best matching abstraction site is Lochem, LGM-no. = 136, in refined model area *achterhoek* (*a2*). As previously mentioned, denitrification does not play a major role here because of aerobic groundwater. The cause of observed nitrate concentrations being lower than calculated ones since about the end of 1980s is that an additional abstraction well was installed in an area with low a nitrate concentration in the groundwater.

Denitrification most probably, with the exception of Archemerberg and Lochem, plays a major role at the other six abstraction sites, which would explain the depicted deviation. A special case is the abstraction site Vierlingsbeek, LGM-no. = 568 in refined model area *ruhrdalslenk* (*r*2). Here the denitrification takes place due to oxidation of pyrite, i.e. redox reaction between nitrate and pyrite, resulting in nitrogen gas, sulphate and a decrease in pH. The changes in sulphate concentrations and pH have actually been observed (KIWA, 1989).

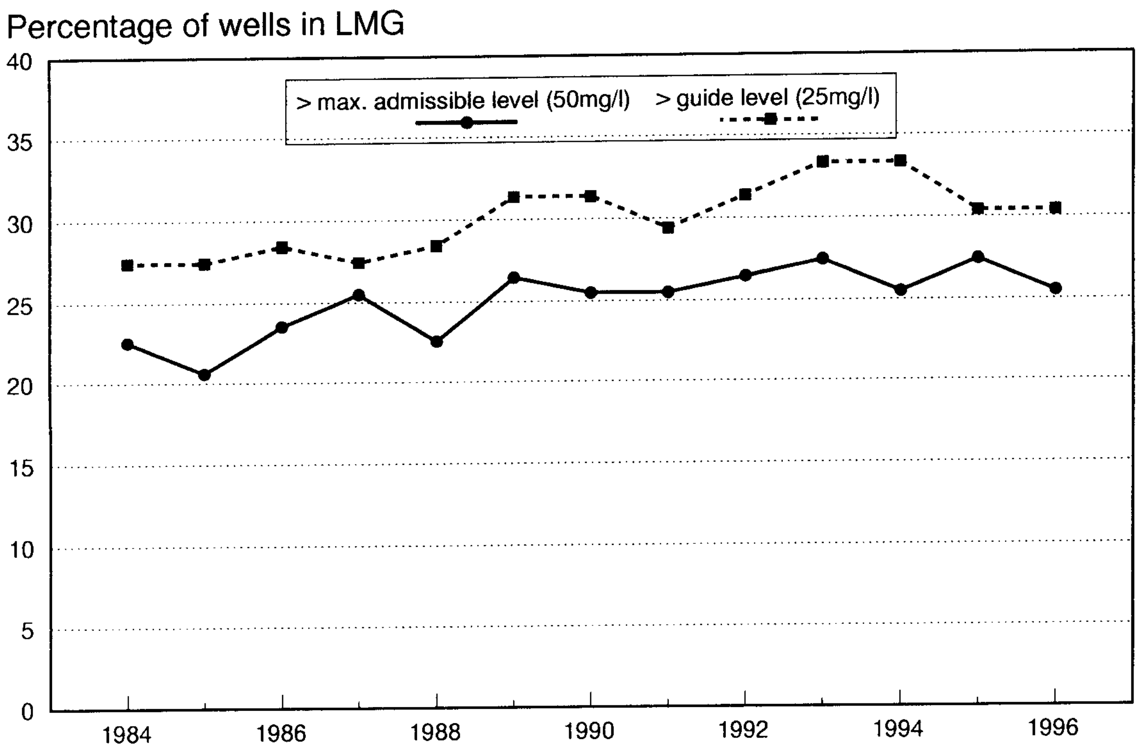


Figure 7.4 *Nitrate variation in deep groundwater (average 10 m below land surface) in sandy soil areas.*

Impact of denitrification in saturated groundwater

The impact of denitrification in saturated groundwater is illustrated by Figures 7.4 and 7.5. Both figures are reproduced from Fraters & Boumans (1997). Figure 7.4 depicts the variation in time of nitrate concentration in saturated groundwater in sandy soil areas of the Netherlands. Groundwater concentrations were obtained from samples between 5 and 15 m below land surface, the average being 10 m below land surface. The travel time to this depth is in the range of one or more decades. The figure is based on data for the period 1984-1996 from the Dutch national groundwater quality monitoring network (LMG). The two lines show the percentage of wells in which a concentration exceeds a given value. The observed data are presented in terms of the nitrate concentrations 25 and 50 mg NO_3 l^{-1}. The latter two are used in the Netherlands as guide value concentration (25 mg NO_3 l^{-1}) and maximum admissible concentration (50 mg NO_3 l^{-1}), respectively. A statistically significant increase (from 23% to 26%) occurs for the number of wells in which the maximum admissible concentration is exceeded.

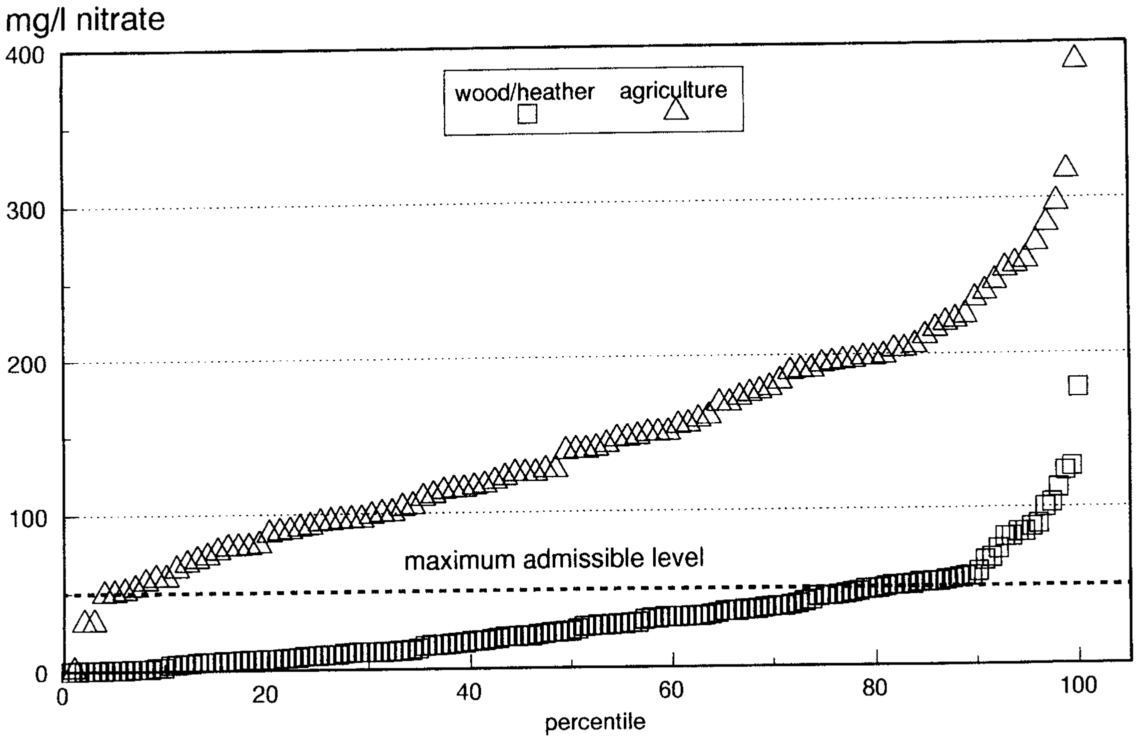


Figure 7.5 *Nitrate concentration in shallow groundwater of nature areas (woodland/heather) and agricultural land.*

Figure 7.5 shows the nitrate concentration in shallow groundwater of nature areas (woodland/heather) and agricultural land. The samples were taken at 100 agricultural farms and at 150 woodland/heather locations. The samples were taken from the first metre of shallow phreatic groundwater. For example, the average depth of phreatic groundwater table at the 100 farm locations in the period 1992-1995 was 1.25 m. The travel time to this depth is in the range of one or more years. The sampling was done in the framework of the Dutch national network for monitoring the effectiveness of the animal-waste control policy (LMM). Figure 7.5 indicates that at 20% of woodland/heather locations and at 95% of the agricultural land locations, the nitrate concentration in shallow groundwater exceeds the maximum admissible concentration (50 mg NO_3 l^{-1}).

A comparison of Figures 7.4 and 7.5 leads to the conclusion that the nitrate concentration in deep groundwater is significantly lower than that in shallow groundwater, thus providing evidence for the discrepancy shown in Figure 7.3. This indicates that denitrification plays an important role in saturated groundwater. Therefore in a follow-up study denitrification will have to be taken into account. As the denitrification process is complex, it may then be needed to also simulate the concentration of other (less reactive) solutes, such as sulphate. Examples of approaches for modelling denitrification on a pumping site scale are given in Boukes (1997, 1998).

7.3 Results for Pesticides

As for nitrate, the variation in concentration for atrazine, bentazone and 1,2-dichloropropane was calculated for 165 abstraction locations (76 phreatic, 89 semi-confined). The simulations were carried out for three scenarios, namely *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*). For practical reasons, only the results for scenario *EC* are presented in this section. The concentration breakthrough curves for 14 selected abstraction sites are attached as examples of results:
- Appendix G, atrazine, scenario *EC*;
- Appendix H, bentazone, scenario *EC*;
- Appendix I, 1,2-dichloropropane, scenario *EC*.

Figures 7.6 and 7.7 show the spatial distribution of atrazine concentration calculated for 2020 and 2050, respectively. According the the Netherlands Water Supply Companies Directive, the value 0.1 μg l^{-1} is the maximum admissible concentration for a single pesticide (for details refer to section 5.1).

Figures 7.8 and 7.9 show the spatial distribution of bentazone concentration calculated for 2020 and 2050, respectively.

Figures 7.10 and 7.11 show the spatial distribution of 1,2-dichloropropane concentration calculated for 2020 and 2050, respectively.

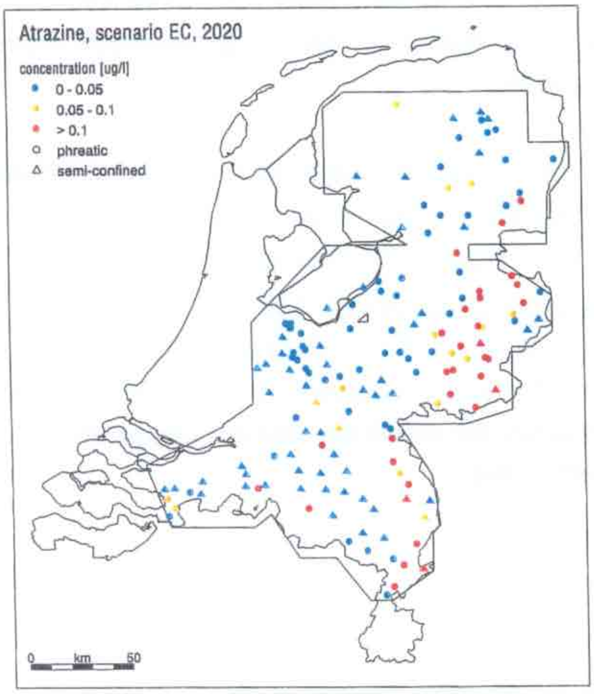


Figure 7.6 *Atrazine concentration in abstracted groundwater, scenario EC, 2020.*

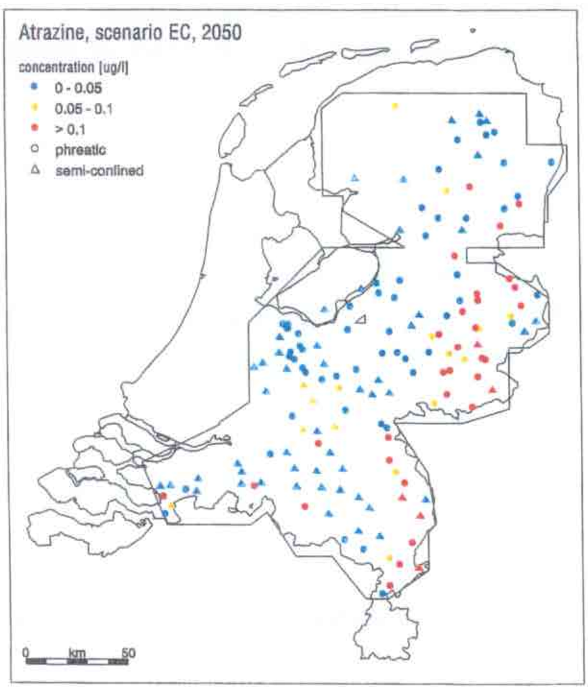


Figure 7.7 *Atrazine concentration in abstracted groundwater, scenario EC, 2050.*

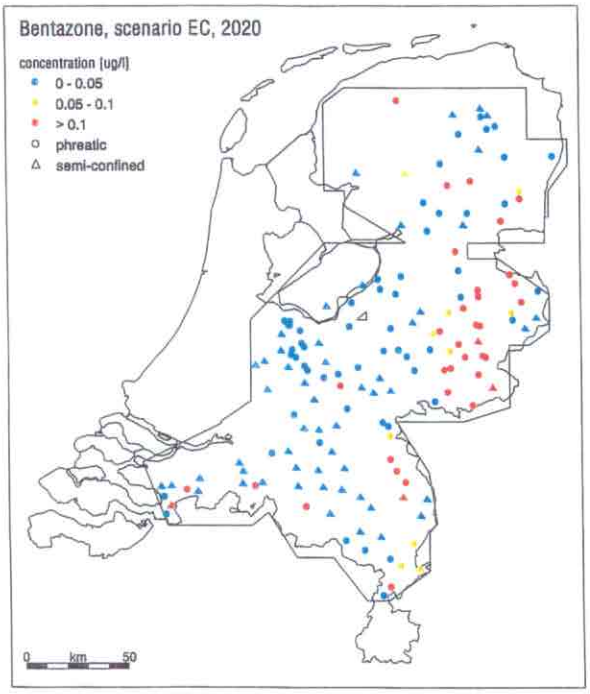


Figure 7.8 *Bentazone concentration in abstracted groundwater, scenario EC, 2020.*

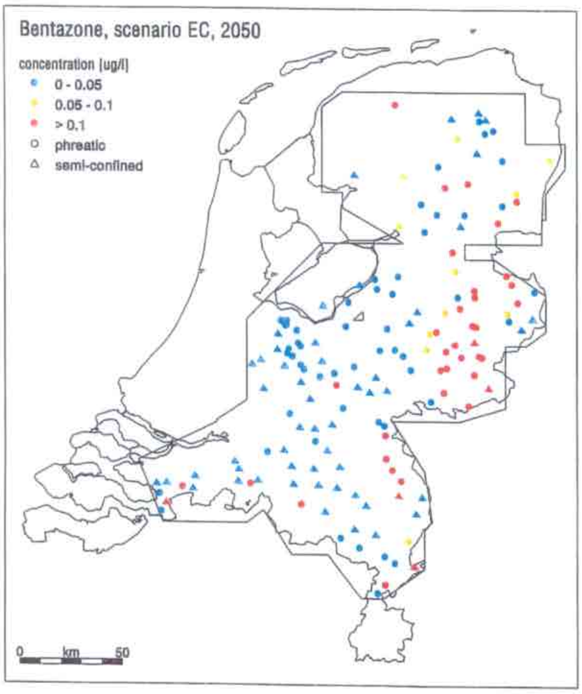


Figure 7.9 *Bentazone concentration in abstracted groundwater, scenario EC, 2050.*

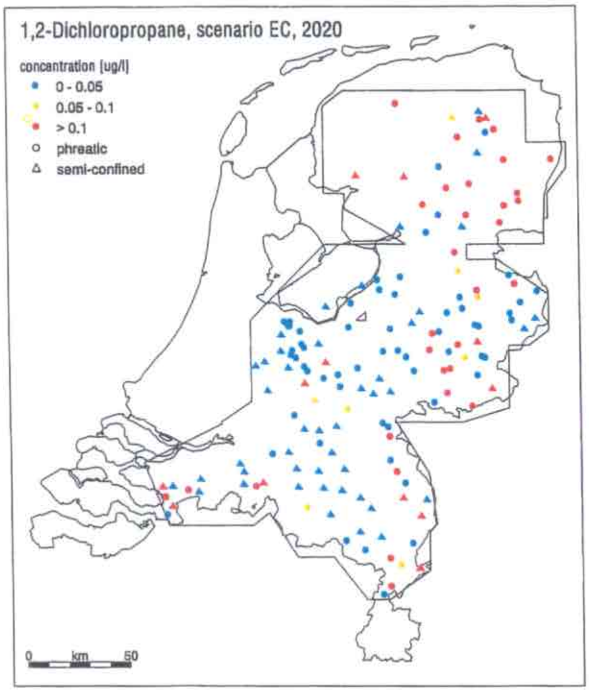


Figure 7.10 *1,2-Dichloropropane concentration in abstracted groundwater, scenario EC, 2020.*

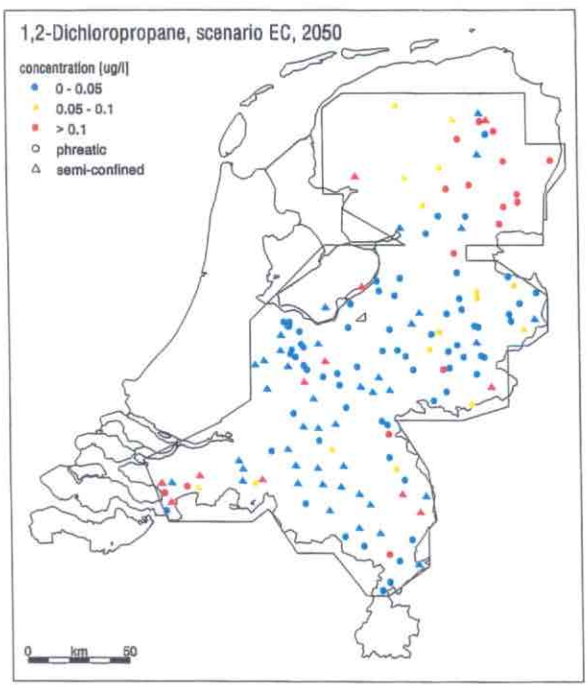


Figure 7.11 *1,2-Dichloropropane concentration in abstracted groundwater, scenario EC, 2050.*

According to the calculation results shown in Figures 7.6 through 7.11, a large number of groundwater abstractions, and consequently the related groundwater-based drinking-water production in Drenthe, Overijssel, Oost-Gelderland and western part of Noord-Brabant will be threatened in the future by too high pesticide concentrations.

Comparison of calculated and observed pesticide concentrations

Figure 7.12 depicts the calculated pesticide concentrations in abstracted groundwater for two abstraction locations (Dinxperlo and Noordbargeres), together with the pesticide concentrations observed at the two locations.

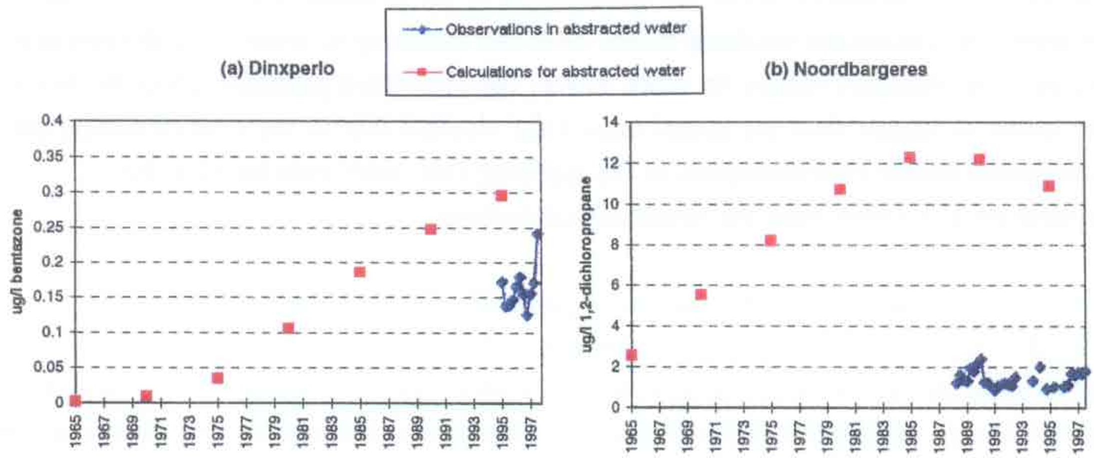


Figure 7.12 *Comparison of calculated (scenario EC) and observed pesticide concentrations for two selected abstraction sites: (a) bentazone, dinxperlo, LGM-no. = 109, refined model area a2; (b) 1,2-dichloropropane, noordbargeres, LGM-no. = 16, refined model area d2.*

The pesticide observation data for the abstraction sites Dinxperlo and Noordbargeres were received from and used with permission of the water-supply company Waterleidingmaatschappij Oostelijk Gelderland WOG) and Waterleidingmaatschappij Drenthe (WMD), respectively.

Just as for nitrate concentrations (section 7.2), here too three hydrological limitations should be observed when interpreting concentrations in Figure 7.12:

1) *Simplification of well field into single well;*
2) *Operational changes in well locations;*
3) *Assumption of constant well rate.*

From Figure 7.12 it follows that the calculated pesticide concentrations for the two abstraction locations are higher than observed concentrations. In addition to the three hydrological limitations previously discussed (section 7.2), the discrepancy between observed and calculated pesticide concentrations is attributable to three other causes:

1) *Uncertainty in pesticide leaching flux:*
   The validation of GEOPESTRAS is discussed in section 5.2. The conclusion is that the uncertainty in the regionalized (500 × 500 m) pesticide leaching values assessed by GEOPESTRAS can be considerable.
   Most probably, the actual leaching values (concentrations) can, as "averages per 500 × 500 m block, vary between at least half and twice the calculated leaching value. No information is available about the distribution of the possible uncertainty in space nor whether the calculated leaching fluxes have the tendency to over- or underestimate the actual concentration values. In other words, the calculated pesticide concentrations can be lower or higher than the actual ones. One recalled that in the case of nitrate the calculated nitrate concentrations in the leaching flux were concluded to be systematically lower than the actual concentrations.

2) *Decay (degradation, transformation) of pesticides in saturated groundwater:*
   Another reason for the discrepancy between observed and calculated pesticide concentration can be decay (degradation, transformation) of pesticides in saturated groundwater. Unlike for denitrification, no literature references on the observed effect of pesticide decay on the scale of a capture zone are known to the authors, i.e. as observed in abstracted groundwater. Pesticide decay is discussed further in this section.

3) *Sorption of pesticides in saturated groundwater:*
   Finally, the discrepancy between observed and calculated pesticide concentration can be also caused by sorption of pesticides in saturated groundwater. The sorption potential depends on the availability of organic matter in subsurface. As very little is known about the spatial distribution of organic matter, sorption was disregarded in this study. A "side-effect" of sorption is that, due to longer residence time of pesticides in the subsurface environment, the decay of pesticides can have greater effect compared to the case without sorption. It is assumed that sorption has relatively smaller impact than degradation of pesticides.

Figure 7.12a depicts the variation of observed and calculated bentazone concentrations at the abstraction site Dinxperlo in the refined model area *achterhoek*. According to the information of the water-supply company, the well-screen filters are regularly reflushed (de-ironing) by temporary infiltration of groundwater. This can also result in a decrease in bentazone concentrations.

The first study towards predicting the future pesticide concentrations in abstracted groundwater in the Netherlands was carried out by Beugelink (1987). This study concerned the future concentrations of 1,2-dichloorpropane in groundwater abstracted at the previously mentioned location Noordbargeres. Beugelink (1987) reports that during the period August 1985 to December 1986 the observed 1,2-dichloropropane concentrations - as an average for the entire well field - increased from 0.7 to 1.7 $\mu g\ l^{-1}$.

The results of the current LGM study for the Noordbargeres location within the refined model area *drenthe* are shown in Figure 7.12b. Because (a) the method used by Beugelink (1987) for calculation of the pesticide leaching flux into the shallow groundwater was different from that used in the current study (Chapter 5), (b) another spatial extent of leaching flux was assumed, and (c) a different groundwater flow model was used, the results achieved by Beugelink (1987, Figure 5) and those in this study are, strictly speaking, not comparable.

Decay (degradation, transformation) and sorption of pesticides in saturated groundwater

Contrary to the unsaturated zone, relatively very little is known about the fate of pesticides in saturated groundwater, i.e. about their transport, decay (degradation, transformation) and sorption. Van den Berg *et al.* (1990) mention that well-documented information was available on only 10 pesticides and their metabolites. For a number of constituents, no decay was detected. For other constituents, it was shown that decay can occur only under certain conditions. Van den Berg *et al.* (1990) state that, due to different environmental conditions it is only partially possible to extrapolate the knowledge about the pesticide behaviour in the unsaturated zone to that in the saturated groundwater, possible specific conditions being:
- availability of oxygen and, if oxygen is not available, the redoxpotential;
- amount, composition and activity of the microbial biomass;
- availability of primary sources of carbon and energy;
- availability of nutrients.

plays a less important role than advection, and (b) the effect of diffusion is negligible. In the previous sections in this chapter it was made plausible that denitrification and, probably to lesser extent, also decay (degradation) of pesticides can play a major role in saturated groundwater. Neither denitrification nor decay of pesticides in saturated groundwater were taken into account.

Conclusions

A comparison was made of calculated and observed concentration values, between 1965 and 1997 for (1) nitrate and pesticides at a number of selected abstraction sites, and for (2) nitrate for aggregated volume abstracted at phreatic sites (Chapter 8). Because the calculated concentrations are higher than the observed ones, denitrification and degradation of pesticides in saturated groundwater are concluded to have a greater impact on the results than the uncertainty in the concentrations of nitrate and pesticides in the leaching flux. Therefore, the modelled concentrations can be concluded to be the worst-case outcome.

8. CONCENTRATION RESULTS AGGREGATED FOR MODEL AREA

8.1 Introduction

This chapter presents the concentration results from Chapter 7 by means of bar charts. The concentration bar charts show concentration variation in time (1950-2050).

The solute concentrations were calculated for 165 abstraction locations (76 phreatic, 89 semi-confined). Separately, bar charts are presented for the totalized abstraction rate of phreatic and semi-confined abstractions. As discussed in section 6.1, the total well rate of the 76 phreatic abstractions is 282.10 $\times$ 10^6 m^3 $year^{-1}$, and the total well rate of the 89 semi-confined abstractions is 457.66 $\times$ 10^6 m^3 $year^{-1}$.

8.2 Nitrate

The simulations for nitrate were carried out for a single scenario, namely scenario *EC(plus)*. Figure 8.1 shows the calculated nitrate variation for phreatic and semi-confined abstractions as a fraction of the totalized well rate.

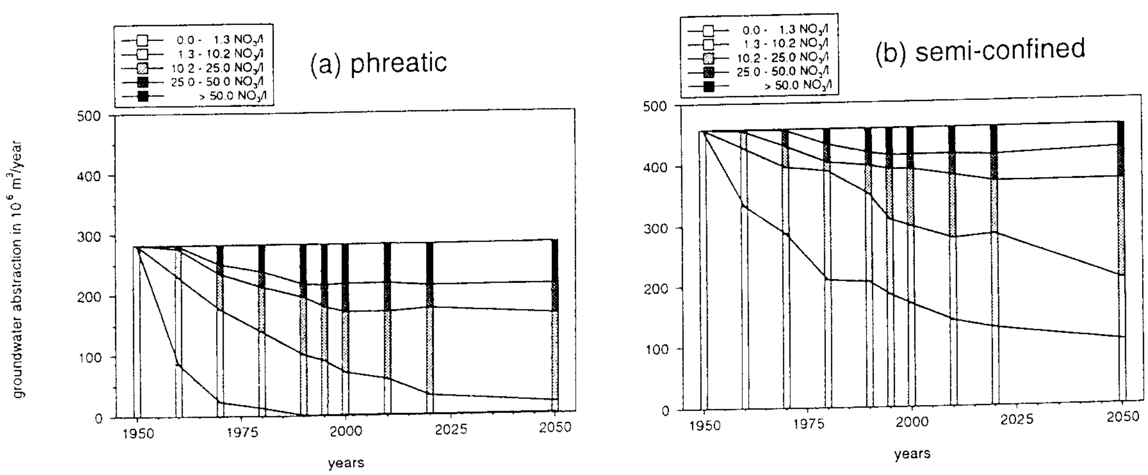
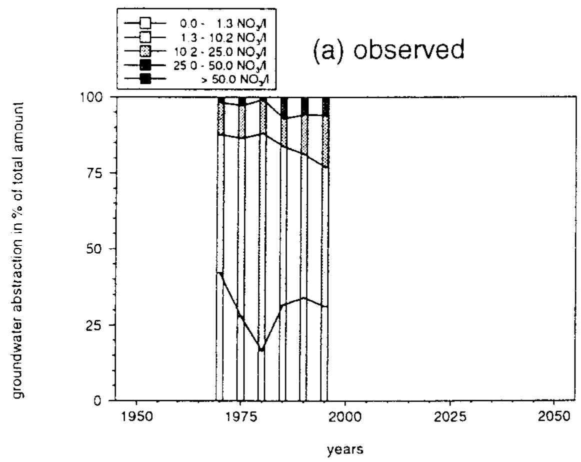


Figure 8.1 *Calculated nitrate variation for (a) phreatic and (b) semi-confined abstractions as a fraction of totalized well rate. Scenario EC(plus).*

Figure 8.2 depicts the observed and calculated nitrate concentration for phreatic abstractions shown as a fraction of 100% well rate. The observed data applies only to 1970, 1975, 1980, 1985, 1990 and 1995. It should be noted that the total number of wells - and thus also the total well rate - used for preparing the observed data varied per year. In this respect, the observed data is not fully comparable with the calculated data, prepared using 76 phreatic abstractions with the constant total well rate of 282.10×10^6 m^3 $year^{-1}$. When comparing Figures 8.2a and 8.2b, it can be seen that the calculated nitrate variation shows significantly higher concentrations than the observed variation. This is consistent with the findings reported in section 7.2 (c.f. Figure 7.3). The most plausible explanation for the discrepancy is that denitrification in deep saturated groundwater was not taken into account.



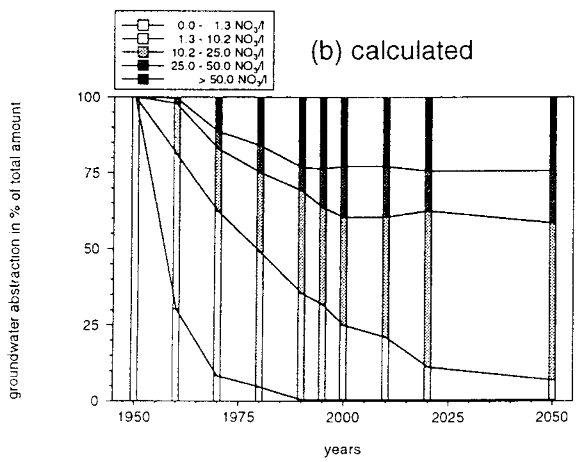


Figure 8.2 *(a) Observed and (b) calculated nitrate variation for phreatic abstractions as a fraction of 100% well rate.*

8.3 Pesticides

Simulations of concentration variation for atrazine, bentazone and 1,2-dichloropropane were carried out for three scenarios, namely *Divided Europe* (*DE*), *European Coordination* (*EC*) and *Global Competition* (*GC*).

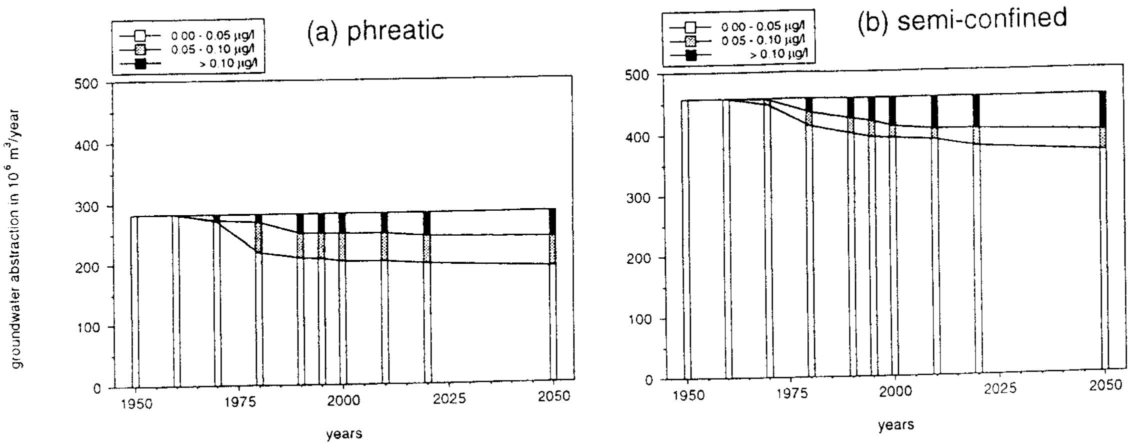


Figure 8.3 *Calculated atrazine variation for (a) phreatic and (b) semi-confined abstractions as a fraction of totalized well rate. Scenario EC.*

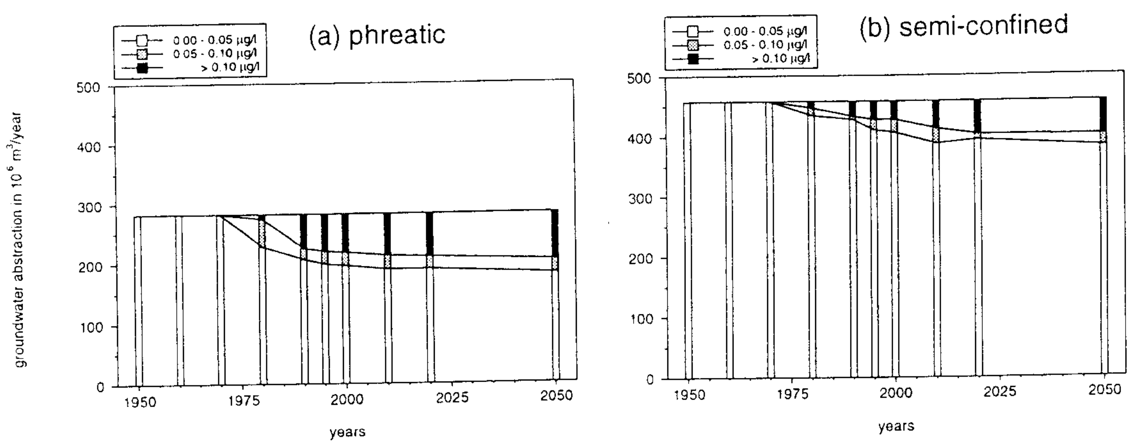


Figure 8.4 *Calculated bentazone variation for (a) phreatic and (b) semi-confined abstractions as a fraction of totalized well rate. Scenario EC.*

The calculated variation of atrazine and bentazone in time for the *EC* scenario is shown in Figures 8.3 and 8.4, respectively. The results for scenarios *DE* and *GC* are not shown as they are very similar to those for the *EC* scenario. The similarity can be explained when considering Table 5.2.

The calculated variation in 1,2-dichloropropane in time is shown in Figure 8.5. The results for 1,2-dichloropropane are identical for all three scenarios (see Table 5.3).

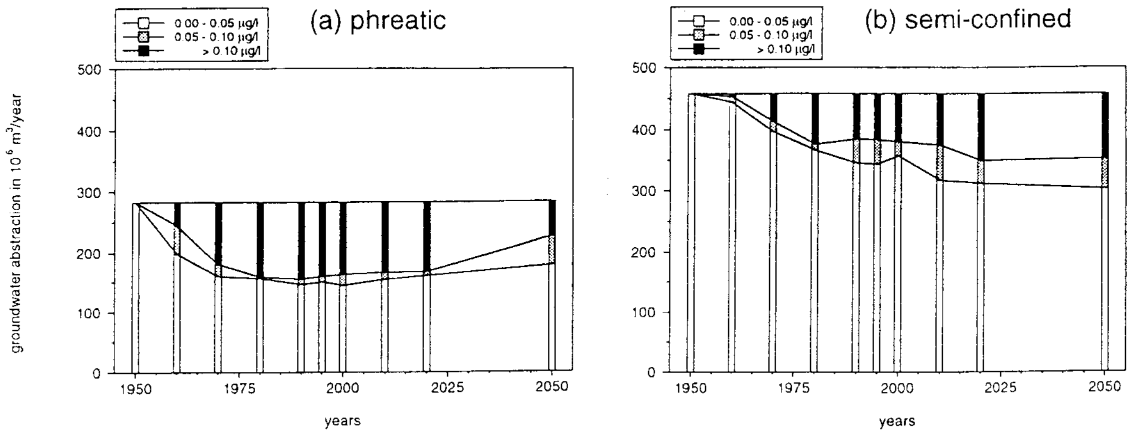


Figure 8.5 *Calculated 1,2-dichloropropane variation for (a) phreatic and (b) semi-confined abstractions as a fraction of totalized well rate. Scenarios DE, EC and GC.*

Analogous to the results for nitrate, it is expected that the calculated pesticide concentrations are higher than the actual concentrations. The reader is referred to section 7.3 for discussion of possible causes in this discrepancy, such as ignoring the decay of pesticides in saturated groundwater.

8.4 Evaluation

The reader should refer to section 7.4 for the discussion of the three factors influencing the reliability of concentrations modelled at individual abstraction sites, namely (1) the reliability of the modelled groundwater flow, (2) the reliability of the modelled leaching fluxes, and (3) the reliability of the modelled solute transport and transformation processes in saturated groundwater. As the aggregated results originate from the calculations done for individual abstraction sites, the three listed factors are also valid for the aggregated results. However, as the current section focuses on the discussion of the aggregated results, the evaluation will be done solely on the basis of Figures 8.1 through 8.5.

As mentioned before, in general, it is expected that the calculated concentrations at many abstraction sites will be higher than concentrations actually occurring in abstracted groundwater. This is especially due to disregarding denitrification of nitrate, and decay (degradation) of pesticides in saturated groundwater, Consequently, the modelled concentrations can also be considered as the worst-case outcome. The worst-case nature of the calculated aggregated values is also documented in Figure 8.2, which shows a comparison of observed and calculated concentrations for aggregated phreatic abstractions.

After this, conclusions will be drawn with regard to the aggregated concentrations. The results for phreatic and semi-confined systems are presented and discussed separately. Obviously, the characterization "phreatic" and "semi-confined" refers to the geohydrological response. The variation in the concentration of nitrate and pesticides for a specific abstraction depends not only on the geohydrological response characteristics but also on the spatial distribution of the concentrations in the leaching flux.

Nitrate (Figure 8.1)

The results concern the scenario *EC(plus)*. The nitrate concentration in the phreatic systems increases more rapidly (deterioration of groundwater quality) than the concentration in semi-confined systems, due to the faster geohydrological response of the former. Conversely, due to the slow response of semi-confined systems, a relatively large portion of groundwater in these systems retains its good quality for a much longer period of time than in phreatic systems.

The results of calculations, namely the comparison of Figures 8.1a and 8.1b lead to the conclusion that especially phreatic abstractions have been affected up to now and will be affected in the future by nutrient application since 1950. The calculated, relatively stronger impact of nutrient application on phreatic abstractions is being supported by changes observed in the chemical composition of groundwater in time (e.g. Mülschlegel, 1992, Figures 6.2.8. and 6.2.9; and VROM/DGM, 1997, Figure 1).

From Figure 8.1a it can be seen that in phreatic systems the calculated nitrate concentration levels of 50 and 25 mg l^{-1} (the two uppermost lines) stabilize between 2000 and 2050. From 2000 onwards, about 75% and 60% of the total volume of abstracted groundwater has a nitrate concentration lower than 50 and 25 mg l^{-1}, respectively.

In semi-confined systems (Figure 8.1b), the calculated nitrate concentration level of 25 mg l^{-1} stabilizes after 2020. The percentage of groundwater with concentrations lower than 50 mg l^{-1} increases slowly after 2020. In 2050, about 90% and 80% of the total volume of abstracted groundwater have a nitrate concentration lower than 50 and 25 mg l^{-1}, respectively. Obviously, the nitrate concentrations in semi-confined groundwater will start decreasing (improvement of groundwater quality) much later than in phreatic systems, i.e. much later than 2050.

Pesticides (Figures 8.3 through 8.5)

The results concern the scenarios *DE, EC* and *GC*. Regarding atrazine (Figure 8.3), the difference between the three scenario results is very slight. The same holds for bentazone (Figure 8.4). The results for the three scenarios for 1,2-dichloropropane, which are identical are presented in Figure 8.5. The improvement in quality at phreatic abstractions in Figure 8.5 takes place significantly more rapidly than for semi-confined abstractions. This is caused by a sharp decrease in the application of 1,2-dichloropropane after 1990 (section 5.4).

The conclusions with respect to pesticides are similar to those for nitrate. As the phreatic systems respond faster than semi-confined systems: (a) the phreatic groundwater deteriorates faster, ending up with a higher percentage of groundwater - compared to semi-confined systems - with a concentration below a certain level, while (b) the semi-confined systems deteriorate more slowly, ending up with a lower percentage of groundwater with a concentration below a certain level. On the other hand, the phreatic systems are "cleaned" faster than the semi-confined systems, as a consequence of the termination of pesticide leaching into saturated groundwater.

9. EFFECT OF TERMINATION OF NITRATE LEACHING

In this chapter a specific application of LGM is discussed. LGM was used to calculate the future concentration of nitrate if the nitrate leaching to saturated groundwater in the entire capture zones is completely terminated in 2000. The results will be compared to those of scenario *EC(plus)*, in which the nitrate leaching to groundwater will exist after 2000.

The calculation was carried out for the refined model area *achterhoek* (model code *a2*) in the eastern part of the country (Oost-Gelderland). This area was selected because most abstractions there are phreatic with a relatively fast response, i.e. with a steep rise of the breakthrough control curve. Another reason for selecting this area is that the nitrate load is very high here.

The calculated nitrate variation for phreatic abstractions, as a fraction of totalized phreatic well rate for scenario *EC(plus)* is shown in Figure 9.1. It can be seen that water quality has deteoriorated rapidly and will remain constant after 1995.

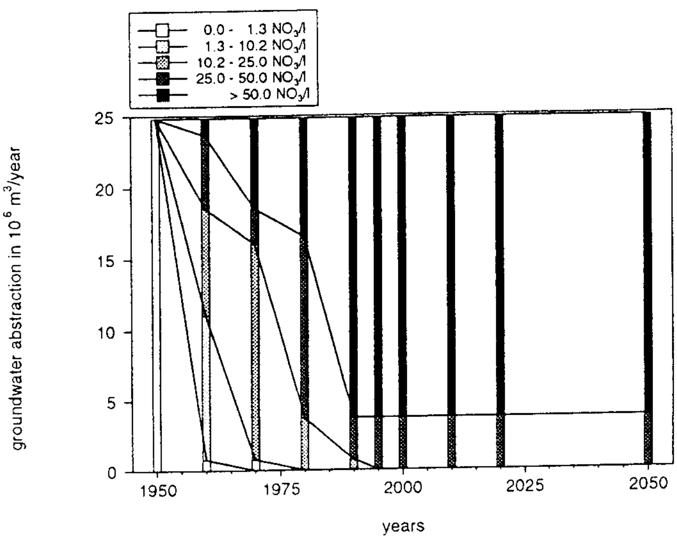


Figure 9.1 *Calculated nitrate variation for phreatic abstractions in refined model area achterhoek (a2) as a fraction of totalized well rate. Nitrate leaching continued after 2000 according to scenario EC(plus).*

The calculated nitrate variation for nitrate leaching terminated in 2000 is shown in Figure 9.2. In this case the nitrate concentration in abstracted groundwater will decrease rapidly after 2000.

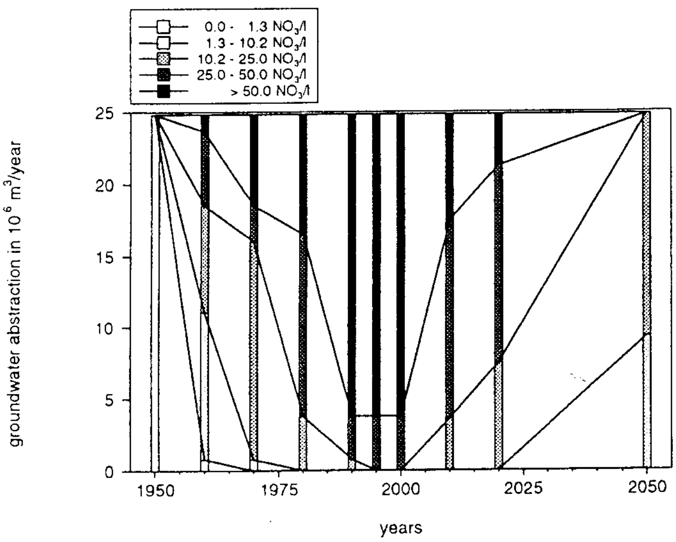


Figure 9.2 *Calculated nitrate variation for phreatic abstractions in the refined model area achterhoek (a2) as a fraction of totalized well rate. Nitrate leaching stopped in 2000.*

10. LAND-USE MAPS

This chapter gives an overview of land-use maps used throughout this study. Due to the different stages of development of various models, it was not possible to apply a single land-use map. The land-use distribution in space was assumed as constant during the entire simulation period from 1950 onwards. The following land-use maps were used:

10.1 Land Use for LGMSAT (Groundwater Recharge)

The LGM module LGMSAT simulates groundwater heads and fluxes in a multi-aquifer system (Chapter 2). One of the input parameters is the groundwater recharge rate (section 2.4). The recharge rate is calculated as the difference between precipitation and actual evapotranspiration. The amount of actual evapotranspiration depends greatly on land use. The land use information for LGMSAT was composed of two datasets (Pastoors, 1992, section 3.6):

- satellite images (Landsat-TM and SPOT) for the sandy soil areas of the Netherlands for images made in 1986. Sandy soils are located mostly in the central, eastern and southern part of the country. The images are used to prepare the dominant land use in 500 × 500 m grid blocks;
- land-use statistics data for the remaining part of the country (mostly grassland), for 1985. The land-use statistics data contain information within 500 × 500 m blocks.

The following eight land-use categories are distinguished in the resulting land-use map (Pastoors, 1992): grass, (fodder) maize, arable land, surface water, decidious forest, coniferous forest, unclassified forest and urban areas.

10.2 Land Use for Nitrate Leaching

The assessment of nitrate leaching is discussed in Chapter 4. Use is made of the land-use map LGN1 (Thunissen *et al.*, 1992). The land-use map is used for both for calculation of groundwater recharge rate and for the load of nitrate on soil. The groundwater recharge rate is the long-term annual average rate between 1961 and 1990 (Van Drecht & Scheper, 1998).

The LGN1 map was created by classification and additional visual interpretation of the Landsat Thematic Mapper images. Most of the images were taken in the summer of 1986. For some areas use was made of satellite images taken in the summer of 1984, 1987 and 1988. The LGN1 land-use data are available for 25 × 25 m grid cells.

The original LGN1 data are available on a 25 × 25 m scale. For the purposes of this study, the original data were upscaled for 500 × 500 m grid cells, containing the percentage of the land-use types occurring within each 500 × 500 m grid cell.

10.3 Land Use for Pesticide Leaching

The assessment of pesticide leaching is discussed in Chapter 5. Referring to Tiktak *et al.* (1996, section 3.1.1), use is made of the land-use map LGN1 (Thunissen *et al.*, 1992). The land-use map is applied for calculation of groundwater recharge rate only, as long-term annual average rate between 1961 and 1990 (Van Drecht & Scheper, 1998).

Citing Tiktak *et al.* (1996, section 3.1.1), the procedure applied at RIVM for running GEOPESTRAS was as follows: *"Using the information of 400 25×25 m^2 grid cells, the relative area of various agricultural crops was calculated for each of the 500×500 m^2 grid cells used in this study. All arable crops were taken together, and finally two classes were distinguished: meadows (61% of total agricultural area) and arable land (39% of total agricultural area). The final land-use map is assigned the dominant of these two land-use types".*

It should be noted that the amount of pesticides used on the land was not based on the LGN1 land-use map. According to Tiktak *et al.* (1996, section 2.3), the pesticide doses were derived from the data in the Information System on Pesticide Use, ISBEST (Merkelbach *et al.*, 1998). ISBEST provides information on the total pesticide use per municipality, the area on which the pesticide has been applied, and the average dose on those parcels where the pesticide has actually been used. The ISBEST database is created on the basis of land-use information obligatorily provided by farmers, at the beginning of the month May in each calendar year.

10.4 Evaluation

By using different land-use maps for the calculation of (1) groundwater recharge rate and (2) leaching fluxes, some inconsistency in the modelling procedure is introduced. Locations where groundwater recharge rate is calculated for grass, while the nitrate leaching at the same spot is calculated for maize can be mentioned as examples. Another inconsistency stems from the fact that the maps used are partly prepared for different time periods. In conclusion, as the general pattern in land use has not changed significantly between the map dates used, it is expected that though the results can be locally (within capture zones) influenced, the aggregated results will not be significantly affected.

11. GROUNDWATER PROTECTION MEASURES

For a better understanding of the further proposed measures, one must bear in mind the difference in the behaviour of phreatic and semi-confined systems. The concentration in the phreatic systems increases more rapidly (deterioration of groundwater quality) than the concentration in semi-confined systems due to the faster geohydrological response of the former. Conversely, due to the slow response of semi-confined systems, a relatively large portion of groundwater in these systems retains its good quality for a much longer period of time than in phreatic systems. Consequently, once solute leaching into saturated groundwater terminates, the concentrations in semi-confined groundwater will also start decreasing (improvement of groundwater quality) much later than those in phreatic systems and the rate of decrease will be smaller.

Tables 11.1 through 11.3 provide information needed for designing prospective measures for improvement of the quality of abstracted groundwater. The tables are based on the LGN2 land-use map of the Netherlands (Noordman *et al.*, 1997). Table 11.1 shows the surface area of land use for phreatic and semi-confined abstractions. The total surface area of the capture zones is 1703 km^2. The surface area of 76 phreatic and 89 semi-confined abstractions is 605 and 1098 km^2, respectively.

Table 11.1 *LGN2-based land use within the total capture zone area (1703 km^2) for phreatic and semi-confined abstractions.*

Land use	Area (km^2)	Phreatic (km^2)	Semi-confined (km^2)
Agriculture	973	317	656
Nature (forest/heather, etc.)	450	217	233
Urban area	152	60	92
Other (lakes, rivers, etc.)	128	11	117

Tables 11.2 and 11.3 show the total capture zone area for four selected ranges of travel times from the top of the saturated groundwater system to well screens. It is assumed that the protection measures would be implemented in 2000.

Table 11.2 *Surface area contribution for various travel time ranges within total capture zone area (km^2) of phreatic abstractions (605 km^2). The percentages in the parentheses refer to 605 km^2.*

Travel time (years)	Period (ca.)	Agriculture (km^2) (%)	Nature (km^2)	Urban area (km^2)	Other (km^2)
< 25	< 2025	86 (14.2%)	62	15	1
25-50	2025-2050	48 (7.9%)	42	12	1
50-100	2050-2100	52 (8.6%)	49	14	2
> 100	> 2100	131 (21.7%)	64	19	7
		317 (52.4%)	*217*	*60*	*11*

Table 11.3 *Surface area contribution for various travel time ranges within total capture zone area (km^2) of semi-confined abstractions (1098 km^2). The percentages in parentheses refer to 1098 km^2.*

Travel time (years)	Period (ca.)	Agriculture (km^2) (%)	Nature (km^2)	Urban area (km^2)	Other (km^2)
< 25	< 2025	73 (6.6%)	37	11	1
25-50	2025-2050	63 (5.7%)	28	9	2
50-100	2050-2100	96 (8.7%)	34	10	13
> 100	> 2100	424 (38.7%)	134	62	101
		656 (59.7%)	*233*	*92*	*117*

The information in Tables 11.1 through 11.3 can be of use in designing the groundwater protection measures in capture zones. One should keep in mind that the calculation results can be considered to be a worst-case outcome. Though the approach will also hold for pesticides, it is illustrated for nitrate only. Agriculture can be recalled as the main source of the nitrate load, which is significantly more important than the load from other land-use types:

a) In phreatic systems (Figure 8.1a), the concentration level of 50 mg l^{-1} stabilizes between 2000 and 2050. From 2000 onwards, about 75% of the total volume of abstracted groundwater has a nitrate concentration lower than 50 mg l^{-1}. In other words, the concentration of 50 mg NO_3 l^{-1} is exceeded in 25% of the volume of abstracted groundwater. From Table 11.2 it follows that nitrate concentration will further increase after 2050 (about 50 years hence). In 2050, 134 km^2 (86+48) of the 317 km^2 agricultural land contributes to nitrate concentrations in the wells. The area of 134 km^2 is 22.1% of the capture zone area. By 2100, an additional 52 km^2 (8.6%) will contribute, thus moderately increasing the nitrate concentrations;

b) In semi-confined systems (Figure 8.1b), the percentage of groundwater with concentrations lower than 50 mg l^{-1} increases slowly from 2020 to 2050, i.e. groundwater deteriorates slowly. In 2050, about 90% of the total volume of abstracted groundwater has a nitrate concentration lower than 50 mg l^{-1}. Further increases of nitrate concentration will take place after 2050. Namely, in 2050 only 136 km^2 (73+63) of the 656 km^2 agricultural land contributes to nitrate concentrations in the wells. The area of 136 km^2 forms 12.3% of the capture zone area. By 2100, an additional 96 km^2 will contribute, thus slightly increasing the nitrate concentrations. The nitrate concentrations will increase considerably during the period after 2100. The increase will be much higher than that for phreatic abstractions. The reason is that during the period after 2100, an additional 38.7% of the capture zone area will originate from agricultural land, while this will be only 21.7% at phreatic sites.

In many cases, the groundwater protection zones in the Netherlands are based on the calculated 25-year travel-time zones. From Tables 11.2 and 11.3 it can be concluded that in this case only 27.1% (164 km^2) and 11.1% (122 km^2), respectively, of the surface area of phreatic and semi-confined capture zones would be protected.

Evidently, policy measures aimed at the decrease in the nitrate leaching into saturated groundwater can be effective, mainly with regard to the agricultural land use. The nitrate leaching in forested areas is less important. It stems indirectly from the agricultural land use, being caused by atmospheric deposition. When considering the policy target time of 50 years from now, i.e. in the year 2050, a decrease of the nitrate leaching from agricultural land from about 2000 onwards would be more effective for phreatic abstractions than for semi-confined abstractions:

a) in phreatic systems, 22.1% (14.2%+7.9%) of the total capture zone is used for agriculture;
b) in semi-confined systems, only 12.3% (6.6%+5.7%) of the total capture zone is used for agriculture.

The considerations on the effectiveness of protection measures assume that nitrate load is equally distributed within phreatic and semi-confined abstractions throughout the entire model area. There are no indications that such an assumption should not hold.

In order to decrease sufficiently the nitrate concentrations at abstraction wells, a complete termination of leaching of nitrate into saturated groundwater within the capture zones would be the safest solution. As the complete termination is an extreme measure, a spatially optimized pattern of reduced nitrate leaching could be designed. From the above exercise (Tables 12.2 and 12.3) it follows that if no nitrate leaches within the travel time zone of 50 years, this would already result in a considerable reduction of nitrate concentrations in abstracted groundwater. Evidently, even better would be if nitrate leaching were terminated within the entire surface area of the capture zones.

Possible policy measures to achieve a decrease in nitrate concentrations in the leaching flux - preferably to zero - are:

1) a decrease in application rate of nutrients (manure and fertilizers) by determining the fertilizer requirements for a targeted yield;
2) a change in agricultural practices, such as attuning of the timing of fertilizer application to crop requirements;
3) a change in land use, such as converting agricultural land into forest.

If it is decided that nitrate leaching be terminated within the entire capture zone, or parts of it, a modelling analysis is required to assess the impact of those measures on the future development in nitrate concentrations. An example of the procedure to be followed was given for the *achterhoek* model area (Chapter 9). However, it should be recalled that denitrification was disregarded in this modelling analysis, while it can have a considerable decreasing impact on nitrate concentrations in abstracted groundwater.

If nitrate leaching is to be terminated within a capture zone, or a part of it, then it is recommended to delineate the areal extent of the protection zone by applying a stochastic (uncertainty) analysis. The reason for this is that a deterministic approach yields a best-possible outcome, being a single "value" of the protection zone geometry. On the other hand, as all model parameters used have an uncertainty associated with them, the size of the protection zone having a required level of confidence will be larger than the protection zone determined deterministically. Consequently, the larger protection zone will imply more certainty for avoiding leaching of nitrate into saturated groundwater within the actual capture zone. An example of the application of a stochastic (Monte Carlo) procedure for delineating capture zones is given in Kinzelbach *et al.* (1996). The methods already operational in the Netherlands for the reliability assessment of travel-time zones are described in KIWA (1997), IWACO (1998) and Uffink (1990).

As was said above, one of the inputs for establishing a groundwater protection zone are the calculated travel-time zones and, preferably, also their reliability. After a protection zone is delineated it might be necessary to carry out the modelling analysis for the concentration breakthrough curves for the anticipated groundwater protection zone. This study demonstrates a procedure for calculating deterministic values of the concentration breakthrough curves at pumping sites. However, it is recommended to develop a methodology enabling the calculation of not only the time-based deterministic concentrations, but also its reliability. An example of the latter forms the 10% and 90% bounds of the probability of occurrence. No ready-to-use methodology seems to be available for this purpose in the Netherlands. The proposed methodology could be also of use for designing the location and operation of groundwater-quality monitoring wells around the well-field to serve as an early warning system.

In conclusion, the most effective manner to ensure that the quality of abstracted groundwater improves sufficiently and rapidly in the future is to take measures leading to a complete termination of solute leaching (nitrate and pesticides) within the entire surface area of the capture zone of groundwater abstraction sites. Though using a Monte Carlo (uncertainty) analysis for delination of the capture zone boundaries would result in a larger surface area of capture zones than obtained by the deterministic approach used in this study, it would ensure protection of the entire capture zone.

12. CONCLUSIONS AND RECOMMENDATIONS

12.1 Conclusions

This study, carried out for 165 groundwater abstraction sites (76 phreatic, 89 semi-confined), focused mainly on the determination of the future concentrations of nitrate and three pesticides (atrazine, bentazone and 1,2-dichloropropane) in abstracted groundwater, aggregated for its total volume. Though the calculations were carried out for a longer period, the aggregated results are presented for the period up to 2050.

The application of LGM reported here has produced results which will be of use in:
(1) obtaining insight into the future development of concentrations in abstracted groundwater;
(2) designing possible policy measures for improving environmental quality, including the quality of abstracted groundwater as a source for drinking-water production, and
(3) evaluating the effectiveness of (future) policy measures for improving the quality of abstracted groundwater.

Applying the LGM - a numerical model comprising complex geohydrological system components and spatially variable (heterogeneous) data - yields more reliable results than obtained when using simplified modelling approaches based on analytical solutions.

The density of the finite-element grid used (base grid 250 × 250 m) was considered adequate. However, the accuracy of pathlines and travel times in the vicinity of wells would further increase if the finite-element grid around abstraction locations were to be locally refined.

The grid density of 250 × 250 m was too coarse for modelling the abstractions in the vicinity of large rivers (bank-infiltration abstractions). These abstraction sites were not considered in this study, primarily for this reason.

Diffuse pollution sources pose an important threat for the quality of groundwater. Main pollutants are nutrients - primarily nitrogen - and pesticides. The emission of nitrogen through agriculture has showed a decrease in the past. This occurred due to regulations restricting manure application, reduction of livestock and a decrease in the amount of fertilizers used. The regulations with regard to pesticide application have resulted in a considerable decrease in the applied pesticide.

The results of the calculations presented here, as well as groundwater quality observations at groundwater pumping sites, lead to the conclusion that the decrease in the amount of applied nitrogen and pesticides mentioned has not (yet) resulted in a significant improvement in the quality of abstracted groundwater. This is caused by the slow response of the groundwater system to changes in solute leaching and by the continued leaching of solutes into saturated groundwater.

Calculated and observed concentration values between 1965 and 1997 were compared for: (1) nitrate and pesticides at a number of selected abstraction sites, and (2) nitrate for an aggregated volume abstracted at phreatic sites. The calculated concentrations are (significantly) higher than the observed ones. The difference is caused by a number of factors. Important factors disregarded in this study are degradation (removal) of solutes in saturated groundwater and, of relatively smaller influence, the simplification of time-variable groundwater abstraction rates as constant values from 1950 on. These two factors lead, respectively, to higher concentration values and a greater increase in concentrations with time. However, the calculated concentrations are also affected by the uncertainty in the calculated concentrations in the nitrate and pesticide leaching flux. As the uncertainty in the leachate concentrations is considered less important than disregarding the solute decay, the modelled concentrations can be concluded to be a worst-case outcome. Though the effect of denitrification (degradation of nitrate) is considered more important than degradation of pesticides, the worst-case outcome holds both for nitrate and for pesticides.

Reliability of results

The reliability (uncertainty) of the calculated concentrations is affected by a number of factors. In this study, the uncertainty was not quantified, e.g. no sensitivity analysis was done. The uncertainty in the concentrations is expected to be especially affected by the following factors, arranged in order of importance:
(a) ignoring denitrification (removal of nitrate) in saturated groundwater,
(b) underestimation of the concentration of nitrate leaching fluxes,
(c) uncertainty in the concentration of leaching fluxes of pesticides,
(d) ignoring degradation of pesticides in saturated groundwater,
(e) schematization of time-variable abstraction rates as a constant well rate since 1950,
(f) uncertainty in the groundwater recharge rate (based on the relatively wet year 1988),
(g) uncertainty in the geohydrological parameters (transmissivities, etc.), and
(h) ignoring sorption of pesticides in saturated groundwater.

Of the above-listed factors (a) through (h), the "direction" (bias) of the error is known only for factors (a), (b), (d) and (h). The factor (e) leads to too high concentrations only in the first part of the calculation period (from 1950 to ca. 2000, depending on the date when a well-field was actually put into operation) but has probably no effect on the breakthrough concentrations at later times. Ignoring denitrification and degradation of pesticides, and sorption of pesticides results in calculated concentration breakthrough values in abstracted groundwater which are too high.

The validation of NLOAD results, using the 500 × 500 m grid-cell-averaged monitoring-network-based values, indicates that the calculated concentrations in the nitrate leaching flux in the agricultural areas are probably lower than the actually occurring concentrations. This would result in calculated nitrate concentration breakthrough values which are too low (b).

As the information on the uncertainty in the pesticide leaching concentrations (c) is scarce and very weak, and furthermore - contrary to nitrate - nothing is known about the bias, no conclusions can be drawn on the effect of this uncertainty source on the pesticide concentration breakthrough curves.

The effect of schematizing the time-variable abstraction rates as constant values (e), the effect of overestimating the groundwater recharge rate (f), the effect of the uncertainty in the geohydrological parameters (g), and the effect of pesticide sorption in saturated groundwater (h) are considered of less importance than items (a) through (d).

Nitrate

The results of calculations lead to the conclusion that especially phreatic abstractions have been affected up to now and will be affected in the future by nutrient application since 1950. The relatively stronger impact on phreatic abstractions is being supported by changes observed in the chemical composition of groundwater in time. The percentage of the abstracted groundwater volume with calculated concentrations higher than the drinking-water standard (50 mg l^{-1} NO_3) is not likely to increase during the period 1995-2020. About 25% of the total volume abstracted at phreatic sites in 2020 and 2050 will have concentrations higher than the drinking-water standard.

The percentage of the phreatic groundwater abstraction volume with calculated nitrate concentrations higher than 25 mg l^{-1} NO_3 will increase slightly during the 1995-2020 period, becoming about 40% in 2020.

When considering the total amount of groundwater abstracted at both phreatic and semi-confined pumping sites, the groundwater volume as a percentage exceeding the drinking-water standard during the 1995-2020 period, and further until 2050, is concluded to be 10% to 15%. The groundwater volume percentage with calculated nitrate concentrations higher than 25 mg l^{-1} NO_3 will also remain more-or-less constant during the period mentioned, being about 27%.

According to the results of calculations (Figures 7.1 and 7.2), an important part of groundwater-based drinking-water production in Oost-Gelderland and Noord-Limburg will continue to be threatened in the future by too high nitrate concentrations.

Currently, no policy measures exist that would on short term result in a balanced nutrient application, i.e. no leaching of nitrate into saturated groundwater. As a consequence, the nitrate concentrations in abstracted groundwater will decrease (very) slowly in the future.

Pesticides

According to the results of calculations (Figures 7.6 through 7.11), an important part of groundwater-based drinking-water production in Drenthe, Overijssel, Oost-Gelderland and western part of Noord-Brabant will be threatened in the future by an increase in pesticide concentrations.

It is concluded that about 10% of both phreatic and semi-confined pumping sites will face atrazine concentrations exceeding the drinking-water standard (0.1 µg l^{-1}). The differences between the three scenarios are very small at the end of the 1995-2020 period.

From the calculation results, the number of pumping sites where the drinking-water standard is exceeded for bentazone increase relatively fast in the beginning of the 1995-2020 period. The increase continues during this time period and beyond. The relatively high increase at the beginning of the 1995-2020 period is due to phreatic abstractions. There is hardly any difference between the results for the three scenarios.

Finally, 1,2-dichloropropane, a major contaminant in the soil-disinfectant 1,3-dichloropropene, was considered. The applications of 1,2-dichloropropane have been heavily reduced by 1990. The number of phreatic pumping sites where the drinking-water standard is exceeded decreases after the end of the 1995-2020 period. The number of semi-confined pumping sites exceeding the drinking-water standard increases during the

1995-2020 period. The number is likely to stabilize after 2020. The leaching fluxes and, consequently, the outcome for 1,2-dichloropropane were identical for the three scenarios.

Nitrate and pesticides

A number of groundwater abstraction sites will be threatened simultaneously by nitrate and one or more of the three pesticides considered. This will concern about 130×10^6 m^3 $year^{-1}$ in the year 2020, being about 18% of the total maximum-permitted groundwater abstraction rate of the 165 pumping sites studied. The relevant abstractions are located in Drenthe, Oost-Overijssel, Oost-Gelderland, Oost-Utrecht, western part of Noord-Brabant, and Limburg.

Measures for improving the quality of abstracted groundwater

Referring to Chapter 11, the most effective manner to ensure that the quality of abstracted groundwater improves sufficiently and rapidly in the future is to take measures leading to a complete termination of solute leaching (nitrate and pesticides) within the entire surface area of the capture zone of groundwater abstraction sites. As the complete termination is an extreme measure, a spatially optimized pattern of reduced solute (nitrate, pesticides) leaching could be designed, taking into account, among other factors, the effect of solute decay in saturated groundwater.

Possible policy measures to achieve the decrease in nitrate concentrations in the leaching flux - preferably to zero - are: (1) a decrease in the application rate of nutrients (manure and fertilizers), such as determining the fertilizer requirements for a targeted yield, (2) a change in agricultural practices, such as attuning of the timing of fertilizer application to the crop requirements, or (3) a change in land use, such as converting agricultural land into forest.

In order to ensure that leaching is terminated within the actual capture zone, the delineation of the protection zone boundaries should be carried out by applying a stochastic (Monte Carlo, uncertainty) analysis. Though this method would result in a larger surface area of capture zones than obtained by the deterministic approach used in this study, it would ensure protection of the entire capture zone.

12.2 Recommendations

Because of the follow-up of this study in the future, recommendations are given which result in an increase in the reliability of the calculated breakthrough concentrations:

i) Incorporation of solute decay, i.e. denitrification (removal of nitrate) and degradation of pesticides in saturated groundwater. Currently, solute decay can be included in LGMCAM as a spatially constant value of the half-lifetime constant. However, the spatial variability of the half-lifetime constant, which is different for aquifers and aquitards, would have to be taken into account. In addition, it might be necessary to introduce a simple reaction for the decay process, applying two or more parameters. Examples of approaches for modelling denitrification, probably also applicable on a pumping site scale, are given in Boukes (1997, 1998).

ii) Development of a calibration (inverse) method to assess: (1) the parameters governing denitrification and degradation of pesticides and (2) the uncertainty in the concentrations in the leaching fluxes. The latter could, for example, be a multiplication factor for the spatially variable concentration pattern calculated in this study. The inverse method would be based on observed concentration breakthrough curves and would not only provide the optimized values but also the linearized confidence intervals of these values. The inverse method would make use of the improved LGMCAM, as mentioned in (i).

iii) Extending the modelling procedure in LGMSAT-LGMFLOW-LGMCAM for time-invariant groundwater abstraction rates (instead of the current single value of abstraction rates) assumed as being constant from 1950 onwards. The variation in abstraction rates could be, for example, schematized stepwise. The groundwater flow could be considered as a consecutive series of steady states, i.e. it would not be needed to solve the transient problem.

iv) Using the groundwater recharge rate as input for LGMSAT for an average meteorological year. The current recharge rate is based on the meteo-data for the year 1988, which results in values about 20% higher than the average. Using a recharge rate higher than the average leads to a smaller surface area of capture zones, but has probably little effect on breakthrough concentrations calculated.

v) Using a denser finite-element grid in the vicinity of wells, for example, 100 × 100 m within the radius of 200 m around each well. This grid refinement would have a positive effect, not only on the accuracy of pathlines and travel times, but also on the concentration breakthrough curves for the abstraction sites.

vi) Improving the spatially variable parameterization of physical and physico-chemical processes governing transport and fate of solutes in saturated groundwater. Use can be made of data from existing national monitoring networks, namely the Dutch national groundwater quality monitoring network (LMG) and the Dutch national monitoring network of drinking-water quality (LMD).

In addition, it is proposed that a modelling procedure be developed to yield not only the deterministic values of concentration breakthrough curve at abstraction wells, but also the uncertainty bounds of the concentration variation in time. In principle, both the LGMCAD module (Uffink, 1990) and the LGMCAM module could be employed as a starting-point. The methodology would be useful for an uncertainty assessment of the effectiveness of the anticipated groundwater protection zones and for the design of groundwater-quality monitoring wells around a well-field to serve as an early warning system.

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