SOILS2SEA DELIVERABLE NO. 3.2

BONUS

SCIENCE FOR A BETTER FUTURE OF THE BALTIC SEA REGION

Upscaling methodologies

SOILS2SEA

Reducing nutrient loadings from agricultural soils to the Baltic Sea via groundwater and streams

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SOILS2SEA

Reducing nutrient loadings from agricultural soils to the Baltic Sea via groundwater and streams

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1. Background and objectives

The Baltic Sea Action Plan and the EU Water Framework Directive both require substantial additional reductions of nutrient loads (N and P) to the marine environment. The BONUS Soils2Sea project conducts research on a widely applicable concept for spatially differentiated regulation, exploiting the fact that the removal and retention of nutrients by biogeochemical processes or sedimentation in groundwater and surface water systems shows large spatial variations. By targeting measures towards areas where the local removal is low, spatially differentiated regulation can be much more cost-effective than the traditional uniform regulation.

To design and evaluate the effectiveness of spatially differentiated regulation requires improved knowledge on the nutrient transport and removal processes at local scale. Soils2Sea therefore conducts field studies with comprehensive data collection and modelling at four sites in Denmark, Sweden, Poland and Russia. Furthermore, Soils2Sea will conduct scenario analyses at the Baltic Sea Basin scale to assess how different regulatory measures as well as changes in land cover, agricultural practices and climate may affect the nutrient losses from the entire Baltic Sea basin to the Baltic Sea.

Evaluating the impacts of local scale spatially differentiated measures at a scale such as the 1.8 million km^2 Baltic Sea Basin poses a particular challenge. Multi-basin hydrological and nutrient models at this scale (e.g. Donnelly et al., 2013) are not able to simulate local scale spatially differentiated measures, because i) the models operate at a much coarser spatial resolution than the measures; ii) they often do not include local scale data but rather aggregated data which can vary in quality and resolution between countries; and iii) they often have simplified process descriptions adequate for the input data complexity and model scale, but sometimes inadequate for simulating specific local scale measures such as field scale crop rotations, Such measures can be simulated by comprehensive and data demanding local scale models (Hansen et al., 2014a; however, for computational and data access reasons these models are not operational at the Baltic Sea Basin scale. Therefore, other methods must be applied for upscaling the results from suitable local scale models to models operating at the Baltic Sea scale. Bronstert et al. (2007) provide one of the very few examples reported in literature of this type of upscaling based on dynamic combinations of small and large scale models.

The objective of the present deliverable report is to describe the upscaling methodologies that have been developed for use in Soils2Sea.

2. Soils2Sea upscaling approach

2.1 General

Soils2Sea uses E-HYPE, a pan-European application (Donnelly et al., 2016) of the Hydrological Predictions for the Environment (HYPE) code (Lindström et al., 2010) as the modelling tool for the Baltic Sea Basin. For the small scale studies in the case areas Soils2Sea uses different numerical and analytical modelling tools that can provide the necessary detailed descriptions required for analyzing spatially differentiated regulations, but that at the same time are not applicable to the Baltic Sea Basin. Hence, the objective of the upscaling in Soils2Sea is to transfer knowledge from local scale models into HYPE for use in simulating the impacts of spatially differentiated measures at the Baltic Sea Basin scale.

The basic hypothesis is that our small scale models, with their more advanced process descriptions and ability to utilize more of the existing system data, have sufficient predictability and that our case studies have sufficient representativeness to allow model outputs to be used for constraining the large scale model.

The methodology applied comprises the following steps:

- *STEP 1 – Compare concepts*. Check the consistency of the concepts used in the local model and in E-HYPE. Identify possible needs for refined process representation or calibration of E- HYPE.
- *STEP 2 – Identify additional data requirements.* Assess whether additional data are required for E-HYPE at Baltic Sea Basin scale, for instance for new process descriptions, recalibration or evaluation of simulation results.
- *STEP 3 – Recalibration of E-HYPE*. Recalibrate HYPE if required.
- *STEP 4 – Upscaled E-HYPE parameters.* This is the core of the upscaling procedure. The local scale models are used to create relationsships for how E-HYPE parameters should be modified to enable E-HYPE to simulate the effects of local scale processes in the scenario analyses.
- *STEP 5 – Use E-HYPE for Baltic Sea Basin simulations.*

2.2 Groundwater

The upscaling approach to assess reduction of nitrate in groundwater is described in Appendix A. The procedures and outcomes of the five steps can be summarized as follows:

STEP 1- Compare concepts.

Local scale models were established for two Danish catchments, Norsminde (101 km^2) and Odense (486 km^2) using two independent models for simulating nitrate leaching from the root zone (NLES) and for simulating flow and transport processes in surface water and groundwater (MIKE SHE). The NLES simulated N-leaching is aggregated to a grid corresponding to the grid scale of the hydrological models i.e. a 100 m grid in Norsminde and a 200 m grid in Odense. HYPE was setup using one subcatchment in Norsminde and two subcatchments in Odense. HYPE operates with three soil layers of which the upper two should represent the tillage and rootzone layers and the lower one can be characterized as corresponding to the groundwater zone.

In the local scale models all flow and solute transport is accounted for by MIKE SHE, while NLES is confined to providing a nitrate source to MIKE SHE at the bottom of the root zone. The concepts for simulating nutrient processes in the rootzone and nitrate reduction in groundwater are very different between the local scale models and E-HYPE. In the local models NLES accounts for denitrification in the root zone, while MIKE SHE calculates nitrate reduction in groundwater by introducing a redox interface somewhere in the saturated zone, above which nitrate is conservative and below which nitrate is reduced instantly. The E-HYPE model assumes that nitrate denitrification can take place in all three soil layers as a function of a decay parameter *K*, the pool of inorganic N, the concentration of inorganic N, soil moisture content and temperature. Furthermore, when the upper layers become saturated, inorganic N can also leach directly from the upper soil layers to streams.

MIKE SHE is calibrated against both discharge and groundwater head data, while the E-HYPE is only calibrated against discharge data. Hence, there is no guarantee that the two models have a comparable simulation of the split between surface near flows and groundwater flows. Similarly, the split of reduction of nitrate between surface water and groundwater cannot be assumed identical in the two modelling approaches. To ensure some consistency the following three analyses were made:

- Use a baseflow filter on observed discharge data as well as on simulated flows from E-HYPE and MIKE SHE and check that the baseflow fractions are comparable within a certain tolerance.
- Ensure that the leaching of nitrate from the root zone in the two models are comparable within a certain tolerance.
- Ensure that the groundwater fraction of the total nitrate reduction that takes place between the bottom of the root zone and the outlet of the catchment are comparable in the two models within a certain tolerance.

STEP 2 – Identify additional data requirements.

To make the analyses for the consistency checks there is a need for some data sets not presently used by E-HYPE:

- A map of baseflow fraction based on analysis of observed discharge data.
- A Baltic Sea Basin map with estimates of the nitrate leaching from the root zone for today's situation. The map shown in Andersen et al. (2016) will be used.
- A Baltic Sea Basin map with estimates of the fraction of nitrate reduction that occurs in groundwater. Our assessment is that previous maps on this are so much off that they are useless. As shown in Appendix B Soils2Sea has moved state-of-the-art forward on this issue by preparing a map that we believe is much better, albeit far from perfect. We will use this map.
- Transport of water and solutes such as nitrate in porous media from the root zone to the river may take several years. It is therefore important to take this lag time of response into account when simulating the effects of remediation measures. For this purpose we have calculated lag times for Poland (Appendix D).

STEP 3 – Recalibration of E-HYPE.

E-HYPE is recalibrated in order to meet the consistency checks described in step 1 on baseflow fraction, N-leaching and groundwater reduction using the additional data described in step 2. If HYPE results are within the accepted tolerance, no recalibration is performed. This is presently in process and will be reported in Deliverable 5.1.

STEP 4 – Upscaled E-HYPE parameters.
MIKE SHE was used to predict how much spatially differentiated regulation can increase nitrate reduction in groundwater for the two Danish catchments Norsminde and Odense. This resulted in a relationship, where the obtained increase in nitrate reduction in groundwater can be derived from the percentage of arable land within the catchment. HYPE was subsequently used on 10 catchments to assess how the parameters in the denitrification rate description for soil layer 3 should be changed to match the increase in nitrate reduction in that layer caused by spatially differentiated regulation. This resulted in a relationship, where the change in the denitrication parameter is a function of the average N-leaching to layer 3 and the soil moisture in layer 3.

In conclusion, this learning process involving both MIKE SHE and E-HYPE, now makes it possible to modify E-HYPE parameters to predict effects of spatially differentiated regulation with respect to nitrate reduction in groundwater.

STEP 5 – Use E-HYPE for Baltic Sea Basin simulations.
This will be initiated soon and reported in Deliverable 5.4.

2.3 Surface water

The upscaling approach to assess N-reduction and P-retention in surface water is described in Appendix C. The procedures and outcomes of the five steps can be summarized as follows:

STEP 1- Compare concepts.
An analytical model was derived to assess the N-reduction and the P-retention in the hyphorheic zone using detailed geometrical characterization of the stream system. This model was tested against the tracer tests conducted for the Tullstorp stream. The analytical tool includes some of the same key concepts as E-HYPE, such as stream length, slope (hydraulic head loss) and mean residence time.

It was not considered necessary to make adjustments to E-HYPE to ensure consistency of concepts.

STEP 2 – Identify additional data requirements.
Not relevant in this case.

STEP 3 – Recalibration of E-HYPE.
This is presently in process and will be reported in Deliverable 5.1.

STEP 4 – Upscaled E-HYPE parameters.

The detailed, analytical model is used to calculate the effect of specific remediation actions, such as replaced substrate in streams, construction of riffle-and-pool sequences or riparian zones. By mathematically requiring that the retention and decay of nutrients in such detailed models are transferred (conserved) in the parameterization of HYPE (Riml and Wörman, 2011), we can assure that important effects of remediation actions are reflected in E-HYPE.

As a conclusion an equation was developed for estimating the change in a E-HYPE parameter required for simulating the impacts of specific remediation measures (see Appendix C for details).

STEP 5 – Use E-HYPE for Baltic Sea Basin simulations.
This will be initiated soon and reported in Deliverable 5.4.

3. Discussion

All models are scale dependent, and a model that is parameterised and calibrated to make predictions at a particular scale does often not have predictive capabilities at smaller scales (Beven, 1995; Refsgaard et al., 1999). To adequately analyse impacts of local scale spatially differentiated measures at the Baltic Sea Basin scale it is therefore required to combine small scale and large scale models.

There are a variety of approaches to calibration in large-scale modelling, but common to all large-scale models is that it is impossible to calibrate in detail at every single observation point. The E-HYPE model regionalises parameters making them general, land use or soil-type specific depending on the process represented by the parameterisation. Performance in single observations points is compromised to achieve the best possible performance across the model domain (Donnelly et al. 2016). This means that for a single given catchment, large-scale models most likely use less calibration data and may be less fitted to the data that is available. On the other hand, the variation in performance can be used to estimate the uncertainty in simulating ungauged basins within the domain. Nevertheless, performance for a given catchment is generally considerably poorer than when compared to smaller scale models. Regarding nutrient calibration, there is also the potential for equifinality in nutrient reduction processes (Beven, 2006) because many combinations of parameter values with different splits between reduction in surface water and in groundwater can provide the same overall N-reduction. At the E-HYPE scale, there is not always enough observation data to separate out whether reduction occurs in surface or groundwater. To help solve this, we use simulation results from small scale models as proxy observational data in recalibrations of E-HYPE. This is possible, because E-HYPE is able to reach the same final calibration targets (water and nutrient fluxes at river gauging stations) via many different combinations of intermediate results (such as local scale flows, nutrient transport and reduction/retention). The recalibration in reality implies constraining E-HYPE to reproduce results at small scales that are comparable with those from detailed small scale models. In this way we expect that the equifinality level in E-HYPE will be reduced such that it to a greater extent will simulate the “right answers for the right reasons”. This improves the confidence in model predictions, when E-HYPE is used in scenario analyses to assess impacts of future changes in climate, land use and agricultural practice.

In both the groundwater and surface water cases the impact of spatially differentiated measures exploiting small scale heterogeneities in natural or modified systems is calculated explicitly by the small scale models and subsequently used to modify a parameter value in E-HYPE, so that E-HYPE can reproduce the same effect. In the groundwater case the small scale model is a numerical model (MIKE SHE/NLES), while for the surface water case it is an analytical model. However, the upscaling principle is the same: *Use a small scale model to derive a relationship by which E-HYPE parameters can be modified to simulate the desired impact*.

Although use of different models in the same study are not uncommon, few other studies (e.g. Bronstert et al., 2007) have utilised this in an upscaling approach, where the local scale model is applied to train or develop a relationship for the large scale model. With the increased use of large scale models and the need to describe impacts of local scale interventions at the large scale, we believe that this approach hold a large potential for further development and wide application.

A critical assumption in this regard is that the calibration of E-HYPE made against data from small scale models will also be valid in other parts of the Baltic Sea Basin. The issue here is not whether the large scale model simulation can match the small scale model in all aspects, but rather to which extent the large scale model can reproduce the same sensitivities to changes in system properties (spatially differentiated measures).

Another critical issue is whether the upscaling relationship, derived under present climate and land use will also be valid under future conditions with changes in both land use and climate. This can to some extent be tested by using both the local scale models and E-HYPE for the scenario analyses on land use and climate changes in the case study areas.

The analyses of lag time in Poland (Appendix D) illustrates a limitation of the overall modelling approach as lag time in the soil system in the order of years to decades, as assessed many places in Poland, cannot currently be taken into account explicitly by E-HYPE due to insufficient data availability. While E-HYPE can simulate aquifer interactions, accurate representation of these processes in the model requires observations of both surface water discharge and nutrient as well as data on groundwater, and too less such data from Poland have been available for the Soils2Sea project. Hence, the scenario studies analysing the changes in nutrient loads due to changes in land use and climate will show the results when a new quasi steady state situation has been achieved, and the final effects can be assessed as the differences between the future steady state situation and today’s situation. Appendix D then provides information for Poland on the expected lag time for this effect to occur. We have not analysed the lag times for the entire Baltic Sea Basin. It is our assessment, however, that the lag times in other parts of the region are generally smaller than in Poland. For instance, lag times in Denmark are typically in the order of a couple of years (Hansen et al., 2014b; Højberg et al., 2015). The reasons for this difference are that the unsaturated zone generally are not so deep in Denmark and that most of water flows in more shallow aquifer systems rather than through deep regional aquifers like in Poland.

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Appendix A

Upscaling of hydrological modelling of nitrate transport and fate

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Abstract

New and more cost-effectively ways of regulating N-losses from agriculture is needed to reach the reduction targets set for i.e. the Baltic Sea. This has in recent years lead to the idea of implementing a spatially targeted regulation of agriculture, which takes into account the spatial variability of natural N-reduction in groundwater by focusing on decreasing N-loss from areas with low reduction, instead of the present uniform regulation. The expected impacts of introducing such a spatially targeted regulation can be simulated with local-scale models with a spatial detailed description of the N-reduction in groundwater, but cannot directly be simulated with the large-scale more conceptual models used for large basins i.e. the Baltic Sea basin. In this paper we present a methodology to upscale knowledge on the impact of a spatially targeted regulation from two local-scale physically based MIKE SHE catchment models to the large-scale and more conceptual E-HYPE model with the aim of using this model to simulate how spatially targeted regulation may affect nutrient loads to the Baltic Sea. We conclude that E-HYPE using this upscaling methodology is likely to be able to simulate the correct trends and order of magnitudes at the Baltic Sea basin scale and as such provides a sound base for large scale policy analysis. However, we do not expect it to be sufficiently accurate to be useful for detailed designing of local-scale measures.

Keywords: Nitrate reduction, Groundwater, Modelling, Spatially targeted regulation, Upscaling

Highlights:

- Spatially targeted regulation of agriculture can cost-effectively decrease N-loads
- We use local-scale models to simulate impact of a spatially targeted regulation
- We develop a methodology to upscale knowledge from local- to large-scale models
- The upscaling enables E-HYPE to simulate the impact for the Baltic Sea basin

1. Introduction

Large nutrient loads have resulted in severe environmental problems in the Baltic Sea during the past decades. In order to address the eutrophication problem the Baltic Sea Action Plan (BSAP) was adopted in 2007 setting very specific goals on who much the nutrients Nitrogen and Phosphorous must be decreased to achieve a “good status” in the Baltic Sea by 2021 (Backer et al., 2010). Many actions to decrease the nutrient loading have already been implemented in the Baltic Sea countries, but the abatement targets are still not met and more actions are therefore needed. In this paper the focus is on nitrogen.

Agriculture is the principal source of N pollution in most of the Baltic basin countries, except for the forested northern countries, Sweden and Finland, where the natural background contribution is largest (EEA, 2005). The loss of nitrogen from agriculture occurs mainly as leaching of excess nitrate from the root zone, which is then transported to surface waters via groundwater. Along the flow path from the source to the catchment outlet nitrate can be naturally transformed and thereby removed by reduction processes. Nitrate reduction is a microbial process occurring under anoxic conditions and in the presence of an electron donor and occurs both in soils, groundwater and surface waters. This removal of nitrogen is in some studies referred to as “N-retention”, but in this study we will use the term “N-reduction”. In this study we mainly focus on the N- reduction occurring below the root zone i.e. in groundwater and surface waters. We define here the N-reduction percentage in groundwater (GW%) as the amount of N reduced in groundwater divided by the N-leaching from the root zone. The N-reduction percentage in surface water is defined as the amount of N reduced in surface water divided by the amount of N transported to the stream from groundwater and other N-sources (e.g. point sources).

The N-reduction in surface water and groundwater in the Baltic Sea basin was estimated by Wulff et al. (2014) to vary between the catchments in the basin from less than 20% in some catchments to more than 80% in others. Whether the N-reduction mainly occurs in groundwater or surface waters is also expected to vary across the Baltic Sea basin, however little literature exists on this, with the exception of a national scale study from Denmark, where groundwater reduction is the dominating sink (Højberg et al., 2015) and a regional scale study from southern Sweden, where groundwater reduction is thought to remove 10-25 % of the gross N-loads to seas (Arheimer and Brandt, 1998).

In the groundwater zone (below the root zone) nitrate is transported conservatively in the oxic part, but is reduced when transported below the redox-interface, which marks the transition from oxic to anoxic conditions (Hendry et al., 1984; Postma et al., 1991; Hansen et al., 2008). The amount of nitrate reduced in the groundwater is therefore dependent on the depth of the redox interface and the groundwater flow paths. Subsurface heterogeneity leads to spatial variations in depth of the redox interface and in groundwater flow paths. The amount of N-reduction in groundwater can therefore vary greatly, not only between catchments, but also within the catchment itself at very local scales (Hansen et al., 2014).

Agricultural regulations to decrease the N-loss from the root zone at present specify the same abatement requirements for all areas. The abatement requirements are thus spatially uniform and do not take local variations in groundwater reduction or surface water reduction into account. Identification of areas with high and low natural reduction along the flow path from below the root zone to the catchment outlet would potentially make it possible to decrease N-loads to surface waters more cost-effectively (Jacobsen and Hansen, 2016). This has in recent years lead to the idea of implementing a spatially targeted regulation of nitrate losses from agriculture. This has especially gained considerable attention in Denmark, where it was included in the recent recommendations of the Danish Nature and Agriculture Commission (NLK, 2013) and now has been included as a measure in the second version of the Danish Water Plans adopted on 1. July 2016. The idea behind a spatially targeted regulation approach is to take into account the spatial variability of natural N-reduction in groundwater i.e. by focusing on decreasing N-loss from areas with low reduction, instead of a uniform regulation where all areas have to decrease by the same amount. Applying a spatially targeted regulation would potentially mean that the required decrease in nutrient use at the source would be smaller than when applying a uniform regulation. A spatially targeted regulation can either be implemented at the source by decreasing N-leaching from the root zone on areas with low reduction, e.g. by changing the crop or the management practice on the field, or it can be implemented where water from areas with low reduction is discharging into surface waters, e.g. in the form of constructed wetlands that can reduce some of the discharging nitrogen (Langergraber et al., 2011).

The effectiveness of regulatory measures for decreasing nitrate loads can be assessed using hydrological and nutrient flux models (Vagstad et al., 2009). To exploit the locally (field and farm-scale) variability in reduction, however, requires simulations at scales resolving the field and farm-scale variability. This is extremely data demanding and requires detailed process descriptions of the hydrological models (Hansen et al., 2014; Karlsson et al., 2016).Modelling of nitrate processes at large scale, such as the Baltic Sea basin, has been carried out by Arheimer et al. (2012) and Wulff et al. (2014) to assess the impacts of international regulations that are applied uniformly across larger regions or even the entire basin. In the case of Wulff et al. (2014) the model resolution required measures to be considered at an aggregated river basin scale and even if the approach used by Arheimer et al. (2012) is considered high-resolution for the Baltic Sea basin (ca 350 km^2 per computational subbasin) and does take into account some subbasin scale variability in groundwater and surface water reductions, to create such a model that operates at field (hectare) scale would be both computationally, but more importantly model input demanding. It is therefore difficult to use these large-scale models to simulate the impacts of local-scale spatially targeted measures because they cannot spatially resolve the measures and often have inadequate process descriptions, in particular for describing the spatial variability in groundwater processes. Therefore, there is a need to combine the knowledge and results achieved by local- and large-scale modelling in an upscaling procedure.

Upscaling can be done in a variety of ways (Bloschl and Sivapalan, 1995; Refsgaard et al., 1999; Vereecken et al., 2007). The most common upscaling approach in distributed hydrological modelling is the *effective parameter approach* assuming that the process equations and system data originating from smaller scale are applicable at a larger scale and that effective parameters exist that can reproduce the mean behaviour of the system observed at larger scale. This assumption is often justifiable (Refsgaard, 1997; Henriksen et al., 2003) while it in other cases has to be rejected (Beven, 1995). Another approach used in distributed models is the *distribution function approach*, where the statistical distributions, but not the geo-referenced locations, of system data and parameters are represented in the model (Andersen et al., 2001; Herbst and Diekkruger, 2002). The use of hydrological response units in semi-distributed models like HYPE (Arheimer et al., 2012; Stromqvist et al., 2012) can be seen as a kind of statistical distribution function approach, although the continuous properties here are replaced by categorical data such as soil type and land use.

A somewhat different upscaling approach is the *dynamic upscaling approach*, where model results from a model with a finer resolution of the computational units is utilized in a large-scale model with coarser resolution. There are only few examples of this upscaling approach (Bronstert et al., 2007; Hansen et al., 2008). In the study by Bronstert et al. (2007) a HBV model was set up for the Rhine basin. For 3 meso-scale catchments within the Rhine basin small-scale models were set up using the distributed physically-based model WASIM-ETH and simulated stream discharge from these models were used in the calibration of the large-scale HBV model. The effect of land use changes was simulated with the small-scale models and these results were then used as input to the HBV model in order to simulate land use change for the entire Rhine basin.

The objectives of this study are i) to introduce a methodology to upscale knowledge from local-scale models to a large-scale model simulating water and nutrient flows for the entire Baltic Sea basin; and ii) to test it on predicting impacts of a spatially targeted regulation to decrease the N-load to the sea. We use the dynamic upscaling approach in a manner similar to Bronstert et al. (2007), extending this approach from only considering discharge to also including N-transport and reduction. The basic hypothesis in the presented upscaling methodology is that our local-scale models, with their more detailed description of the system in terms of input data and more advanced process descriptions, have sufficient predictability and that our two case studies have sufficient representativeness to allow model outputs to be used for upscaling to the larger scale model.

2. Upscaling methodology

The aim of the upscaling presented in this paper is to develop a modelling tool that is able to simulate the impact of local-scale spatially targeted N regulation for the Baltic Sea basin. The spatially targeted N-regulation aims at exploiting the considerable spatial differences in the natural N-reduction taking place in groundwater and surface water. Our study focuses on N-reduction in groundwater. The tools we selected to describe large-scale and local-scale processes are E-HYPE and MIKE SHE, respectively. We use the Baltic Sea basin part of the pan-European E-HYPE v3.1 (Hundecha et al., 2016) water and nutrient flux model based on the HYPE model code (Lindstrom et al., 2010). The Baltic Sea basin covers 1.8 million km^2 represented by 7,145 subbasins in E-HYPE with a median (but variable) resolution of 215 km^2 MIKE SHE model is a high-resolution catchment model that can describe the process details required to assess impacts of local-scale spatially targeted N regulation.

2.1. Study areas

The study areas used to develop the upscaling methodology are the two Danish catchments: Norsminde catchment, located on the east coast of Jutland, and Odense catchment, located on the island of Funen (figure 1). Norsminde catchment covers an area of 101 km^2 and discharges into the Norsminde Fjord. The most downstream gauging station in the stream covers an area of 85 km^2 (figure 1a). Odense catchment covers an area of 486 km^2 (catchment to Kratholm monitoring station; figure 1d) and is an upstream subcatchment of the larger Odense Fjord basin (1025 km^2). Both Norsminde Fjord and Odense Fjord are sensitive water bodies with respect to nitrogen, and a significant decrease of the N-load to the fjords are required to obtain good ecological status (Danish Nature Agency, 2016).

The land use in both catchments is dominated by intensive agriculture (figure 1b and 1e). A part of the agricultural area in the catchments is used for permanent grass, which will not be utilized in a spatially targeted regulation of agriculture. The agricultural area in rotation (from now on called arable land), suitable for applying a spatially targeted regulation, makes up 62% of the catchment area in Norsminde and 61% in Odense.

The climate in Norsminde and Odense catchments is temperate and humid. The catchments are located in glacial landscapes from the Weichsel glaciation, where the upper geology is dominated by clayey till with smaller units of glacial melt water sand and post-glacial freshwater peat (figure 1c and f). Due to the predominance of clay till soils the two catchments are to a large extent tile drained.

In both Norsminde and Odense catchment observations of daily discharge are available from the stream monitoring stations. Observations of Total-N concentrations are available at the downstream monitoring station in Norsminde for the period 2000-2007 with 14-48 samples per year. In Odense observations of Total-N concentrations are available at the downstream station with 114-183 samples per year in the period 2000-2009 and 16-17 samples per year from 2010 and until present.

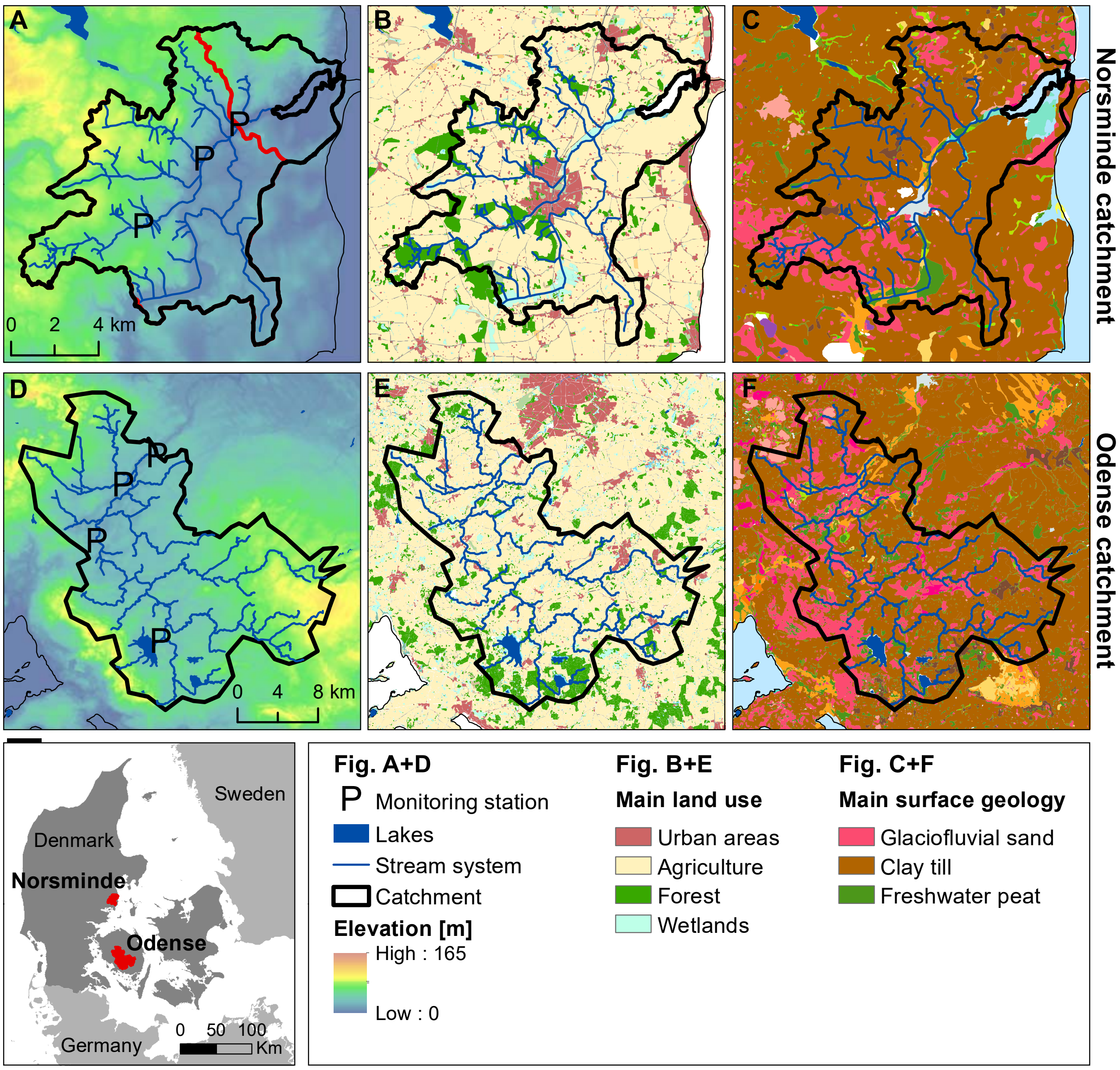

Figure 1 Norsminde and Odense catchment. A+D) Topographic elevation (meters above sea level), stream system and location of monitoring stations. B+E) Main land use classes. C+F) Main surface geology classes.

2.2. Models

2.2.1. Local-scale models (MIKE SHE/NLES)

The local-scale models comprises the MIKE SHE catchment model simulating flow and transport and the agricultural field scale model NLES simulating N-leaching from the root zone as an N source for MIKE SHE

N-leaching model - NLES

The N input to the MIKE SHE models for Norsminde and Odense catchment is N-leaching from the root zone. Leaching of N from agricultural areas is calculated using the NLES model (Kristensen et al., 2003; Kristensen et al., 2008), whereas standard values for N-leaching are used for all non-agricultural areas. NLES is an empirical model that does not include flow descriptions in the root zone. NLES calculates a yearly N-leaching based on input data on crop rotation, input of fertilizer and manure, N-fixation, percolation, soil type and content of organic matter and clay in the soil.

NLES is calibrated against N-leaching observations from Denmark and therefore reflects these observations and the agricultural practice behind. The yearly N-leaching from NLES is afterwards disaggregated to daily values using results from the physically-based root zone model Daisy (Abrahamsen and Hansen, 2000) set up for typical crop rotations and soil types in the area. Daisy is also used to calculate the monthly percolation used in the NLES calculations. The N-leaching results for all areas are afterwards aggregated to grid corresponding to the grid size of the hydrological model i.e. a 100 m grid in Norsminde and 200 m grid in Odense.

Input data to NLES on crop rotation and application of fertilizer and manure is for the period 1990 to 2000 based on land use data available on parish level from the Danish Statistical Databank. After 2000 crop and fertilizer/manure data are available on field block or field level from national agricultural databases (data from The General Agricultural Register (GLR in Danish) and data on yearly fertilizer use and area of catch crops from The Danish AgriFish Agency). Soil data used in NLES is from typical Danish soils.

Model code – MIKE SHE

MIKE SHE is a distributed physically-based hydrological model. MIKE SHE includes process descriptions for evapotranspiration, snowmelt, 2D overland flow, 1D unsaturated flow, 3D saturated flow, macropore flow, tile drainage and 1D river flow. The computational units are georeferenced grid cells and all model parameters can vary for each grid cell. Transport can be simulated as particle tracking or advection-dispersion (Havnø et al., 1995; Refsgaard and Storm, 1995).

Model set up – MIKE SHE

Norsminde

The MIKE SHE model for Norsminde is based on of the model set ups from He et al. (2015) and Hansen et al. (2014), but a few changes are made to the model. The model has a grid size of 100 m x 100 m and vertically the saturated zone of the model is divided into 24 computational layers of varying thickness. The flow in the unsaturated zone is described by the two-layer model in MIKE SHE and flow in the saturated zone as three-dimensional (3D) saturated flow. The climate input used in the model is obtained from the Danish Meteorological Institute's (DMI) 10 km grid for daily precipitation and 20 km grid for reference evapotranspiration and air temperature (He et al., 2015).

The Norsminde model is set up as a transient model for the period 1995 – 2007. The model is inversely calibrated using the parameter estimator PEST (Doherty, 2005) against daily discharge data from the 3 monitoring stations (figure 1a) and 690 hydraulic head measurements in 108 wells for the period 2000-2003 (He et al., 2015).

The transport of N in MIKE SHE is simulated using particle tracking. The N-leaching from NLES is applied as a daily input to MIKE SHE for the period 2000-2007, where a particle is released for each 2 kg of N added to a grid cell. The particles are released on the water table and tracked until they reach a sink (stream, drain, well or fjord/ocean). When a particle crosses the redox interface it is assumed to be completely reduced (Hansen et al., 2014). Point source data are added to the simulated N-load from groundwater to the stream outside the MIKE SHE framework. The same is done with the N-reduction in the stream system. The amount of N-reduction in the stream is calculated simplistically on a subcatchment level (4 subcatchments within Norsminde) and is defined as a function of the stream length.

Odense

The MIKE SHE model for Odense is based on the model setup by Karlsson et al. (2015) and Sonnenborg et al. (In preparation). The model has a grid size of 200 m x 200 m and the saturated zone is divided into 7 computational layers. The flow in the unsaturated zone is described with the full Richards' equation and flow in the saturated zone as 3D saturated flow. The climate input in the Odense model is also obtained from the DMI climate grid (Karlsson et al., 2015).

The Odense model is a transient model running from 1990-2010. The model is calibrated with the global optimization algorithm Population Simplex Evolution method (PSE) using the AutoCal tool in MIKE SHE. The calibration is done for the period 2004-2007 using daily discharge data from the four monitoring stations (figure 1d) and hydraulic head data from 455 wells (Karlsson et al., 2015).

The N-transport in the Odense model is simulated in a similar way as in the Norsminde model using particle tracking and a redox-interface. The N-leaching input to MIKE SHE is changed from the original model setup to the NLES estimated daily N-leaching. In the Odense model it is assumed that all reduction within the catchment is occurring in groundwater and that reduction in the stream system is negligible (Sonnenborg et al., In preparation).

2.2.2. Large-scale model (HYPE)

Model code - HYPE

The HYPE model is a semi-distributed model mixing conceptual and physical descriptions for different hydrological and nutrient processes (Lindström et al. 2010). Calculations are made on a daily time-step in coupled subbasins. The subbasins are divided into hydrological response units (HRUs), which are a calculation unit with a unique land use and soil type. The HRUs can be divided in up to three soil layers that can have different thicknesses. Model parameters are either the same for the whole model domain or are related to soil type or land cover depending on the process they represent. Some of the processes included in HYPE are evapotranspiration, snowmelt, surface runoff, surface erosion, macro pore flow, tile drainage, groundwater outflow from each soil layer and routing of water through the river network. Rather than coupling with a leaching model, HYPE simulates the continuous transformation of nutrients from the surface, via the soil and in surface waters. Processes simulated include plant uptake, turnover, mineralisation, denitrification and erosion.

Model set up – HYPE

Two applications of the HYPE model were used in the study to test the upscaling procedure. Both the Norsminde and Odense catchments (to the monitoring station) were extracted from the existing E-HYPE v3.0 model (Hundecha et al., 2016) which has since been extended to simulate water quality using the same methods used in the previous water quality version (Donnelly et al., 2013). The model inputs come from continental and global scale databases. For example, precipitation and temperature are from the 0.5 degree WFDEI database (Weedon et al., 2014). Norsminde catchment consists of 1 subbasin in the E-HYPE model and Odense catchment consists of 2 subbasins. Testing the upscaling in Norsminde and Odense on this model version, hereafter referred to as E-HYPE, will demonstrate how the upscaling could work when extended to the entire Baltic Sea catchment.

2.2.3. Comparison of model concepts

In the local-scale models all flow and solute transport is accounted for by MIKE SHE, while NLES is confined to providing a nitrate source to MIKE SHE at the bottom of the root zone. The concepts for simulating nutrient processes in the root zone and nitrate reduction in groundwater are very different between the local-scale models and E-HYPE. In the local models NLES accounts for denitrification in the root zone, while MIKE SHE calculates nitrate reduction in groundwater by introducing a redox interface somewhere in the saturated zone, above which nitrate is conservative and below which nitrate is reduced instantly. The E-HYPE model assumes that nitrate denitrification can take place in all three soil layers as a function of a decay parameter *K*, the pool of inorganic N, the concentration of inorganic N, soil moisture content and temperature. Furthermore, when the upper layers become saturated, inorganic N can also leach directly from the upper soil layers to streams.

The HYPE model and MIKE SHE/NLES model are therefore very different in the way the two models describe N-fluxes in the system and it is not straightforward to compare results from the two models. Figure 2 shows conceptual sketches of the fluxes of N-loads from source to catchment outlet in the two models.

MIKE SHE has only two N-fluxes to the stream system; via groundwater flow and via tile drain flow. Saturated flow in MIKE SHE is simulated in 3D and N can be transported to the stream system via both shallow and deep groundwater flow paths over the entire depth of the saturated zone. The tile drains are located at 1 m depth, at the bottom of the root zone, and will be active when the water table is above the drain level. N-transport is not simulated via overland flow in when applying particle tracking in MIKE SHE, because particle tracking is restricted to the saturated zone. However, since overland flow is of little importance in the two catchments, this will have negligible effect on the simulated N-load to the stream.

HYPE includes three N-fluxes out of the upper two layers covering the root zone; 1) N-leaching to the deeper layer three that constitutes the groundwater zone, 2) a horizontal flux directly to the stream system and 3) tile drain flow to the stream system. The last two N-fluxes are only active when the water table rises into the root zone making the bottom of the root zone saturated. From the groundwater zone there is one N-flux via groundwater flow to the stream system. HYPE also includes a possible N flux to the stream with surface runoff either due to saturation excess or infiltration excess. This flux is negligible in our two case studies and is therefore not included in the following equations.

Because the two models do not include the same N-fluxes, comparing N-loads and groundwater reduction between the two models is not trivial. The N-load from the root zone is defined as follows for the two models, with MIKE SHE terms (subscript *M*) on the left hand-side and HYPE terms (subscript *H*) on the right-hand side and where notation is defined in figure 2:

$$L_{M\_leach} = L_{H\_leach} + L_{H\_UL} + L_{H\_TD} \qquad \textbf{(eq.1)}$$

The N-load to the stream system is defined by:

$$L_{M\_GW} + L_{M\_TD} = L_{H\_GW} + L_{H\_UL} + L_{H\_TD} \qquad \textbf{(eq.2)}$$

Finally, the fraction of the total N-leaching from the root zone that is reduced in the groundwater is defined by:

$$\frac{L_{M\_leach} - (L_{M\_GW} + L_{M\_TD})}{L_{M\_leach}} = \frac{(L_{H\_leach} + L_{H\_UL} + L_{H\_TD}) - (L_{H\_GW} + L_{H\_UL} + L_{H\_TD})}{L_{H\_leach} + L_{H\_UL} + L_{H\_TD}} \qquad \textbf{(eq.3)}$$

$$= \frac{L_{H\_leach} - L_{H\_GW}}{L_{H\_leach} + L_{H\_UL} + L_{H\_TD}}$$

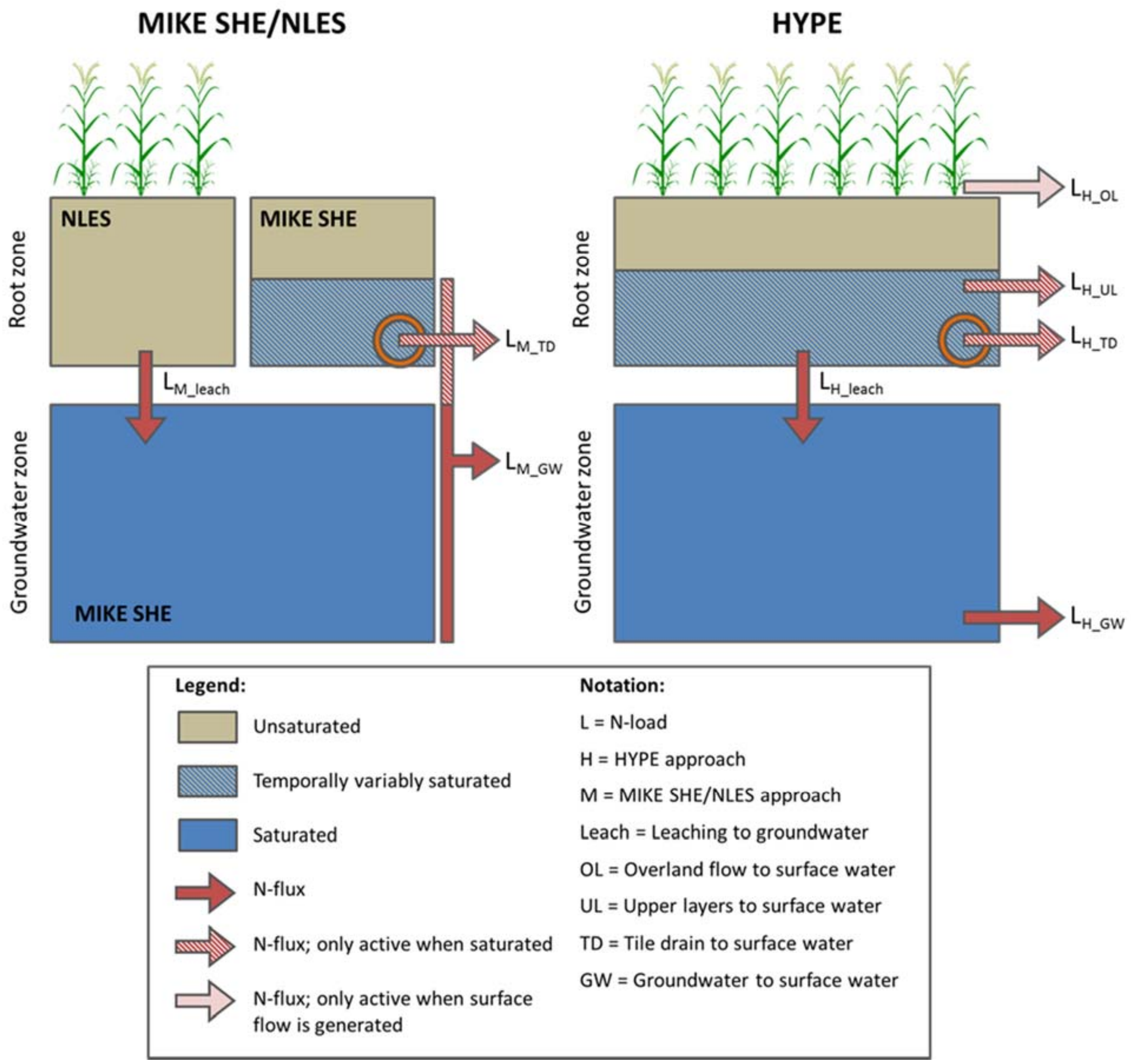

Figure 2 Conceptual drawing of the N-loads in MIKE SHE/NLES and HYPE approach

2.3. Upscaling approach

In order to enable the HYPE model to simulate the impacts of spatially targeted regulation on the N-load to the Baltic Sea we therefore need to modify the current E-HYPE model setup. This modification needs to be based on information from local scale, making it essential to apply an upscaling procedure. The effective parameter approach and the distribution function approach are not directly applicable for our purpose, because we want to simulate the impact of spatial differences within one computational unit of the large-scale model. To do this with HYPE requires that parameter values in HYPE are changed to reflect the impact of the spatially targeted regulation, and we have no a priori information on how to do this. Therefore, we adopt the dynamic upscaling approach, where the basic idea is that the HYPE through our two case studies, Norsminde and Odense, is learning by simulation results from the local-scale MIKE SHE models, or in other words that we learn from MIKE SHE simulations how to parameterise HYPE.

The upscaling methodology consists of three steps: (1) Baseflow fraction, (2) N-reduction in groundwater and (3) Impact of spatially targeting. Assuming that E-HYPE needs to have a reasonably accurate description of the split between baseflow and surface near flow and between N-reduction in groundwater and in surface water to enable it to accurately simulate the impact of spatially targeting, Step 1 and Step 2 can be seen as consistency checks and, if necessary, recalibration. In order to make this possible for the entire Basin Sea basin the consistency check/recalibration must be done against data sources that are available for the entire basin. Step 3 is then to use HYPE to simulate the impact of a spatially targeted regulation using some upscaling relationships. The upscaling methodology is illustrated in figure 3 and will be described in more details in the following sections.

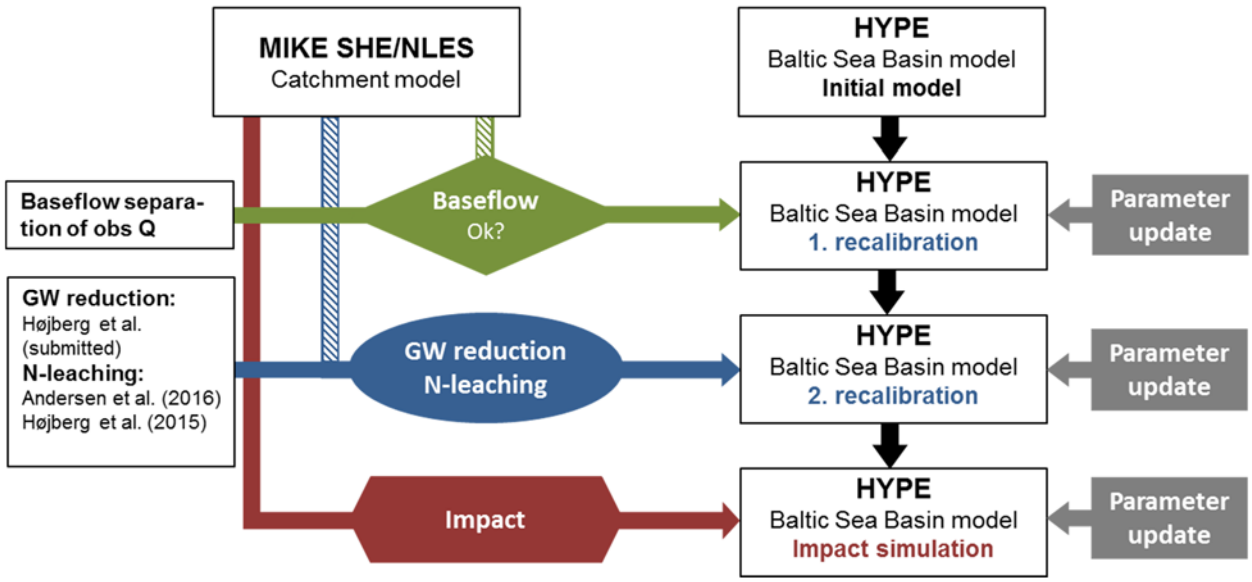

Figure 3 The upscaling methodology consists of three steps: (1) Consistency check of baseflow fraction (green box), (2) upscaling of nitrate reduction in groundwater (blue box) and (3) upscaling of impact of spatially targeting (red box).

2.3.1. Step 1 - Baseflow fraction

The first step in the methodology is to do a consistency check of the split between shallow and deep flow in the model. This is done by checking and if needed calibrating the baseflow fraction of the simulated discharge in HYPE against the baseflow fraction of the observed hydrograph. The baseflow fraction is estimated by applying the baseflow filter BFLOW (Arnold et al., 1995) to observed and simulated hydrographs. BFLOW uses three different passes where the hydrograph is separated in several steps. We will use pass 3, giving the lowest baseflow fraction. The baseflow fraction (BF%) is calculated as the long-term total baseflow divided by the total discharge. The baseflow fraction is used as a soft criterion in the calibration, so that the calibration is deemed acceptable if the mean value of baseflow from HYPE is within +/- 20% of the observed baseflow fraction.

2.3.2. Step 2 - N-reduction in groundwater

After the flow has been checked/recalibrated, the next step is to calibrate the groundwater reduction in HYPE. We have found it necessary to also calibrate the N-leaching in HYPE alongside with the groundwater N-reduction. The parameters that are calibrated in this step are; a time constant for denitrification (d^{-1}), a parameter for decay of humus to fast N (d^{-1}), a parameter for mineralisation of fast N to inorganic N (d^{-1}) and a parameter controlling the crop uptake.

Constraining the N-leaching in E-HYPE requires a best possible estimate of N-leaching. Simulated values are used, as leaching observations are not systematically available. At the Baltic Sea basin scale this is a 10 km grid scale map produced by Andersen et al. (2016) (figure 7 pp. 14). For Denmark we use N-leaching based on the NLES model on a 500m grid scale produced for use in the Danish national N-model (Højberg et al., 2015).

For constraining the groundwater N-reduction in HYPE we are utilising a new map with estimates on the groundwater reduction for the Baltic Sea basin produced by Højberg et al. (Submitted). This map is based on the available national studies supplemented with expert judgement. For the Danish subbasins the groundwater reduction map is based on the results from the Danish National N-model (Højberg et al., 2015).

2.3.3. Step 3 - Impact of spatially targeted regulation

The final step in the upscaling methodology is to derive relationships based on the local-scale models to estimate how HYPE parameters can be adjusted to allow HYPE to simulate the impacts of spatially targeted regulation of agriculture. First the local-scale models for Norsminde and Odense are applied to simulate and assess the impact of applying a spatially targeted regulation at 100-200 m scale. We then need to find a way to transfer the results gained from the local-scale impact modelling for Norsminde and Odense to the rest of the Baltic Sea basin to estimate the expected impact in each subbasin. We assume that the denitrification parameter in E-HYPE can be modified to obtain the expected impact, i.e. changes in denitrification rate are used as a proxy for obtaining the changes resulting from applying a targeted regulation of agriculture. This was developed into generic relationships that use information readily available in E-HYPE and hence are transferable to all subbasins in the Baltic Sea basin. The upscaling relations will be presented in the results section (section 3.2).

2.4. Impact scenario of spatially targeting in Norsminde and Odense

In order to assess the potential impact of applying a spatially targeted regulation, an impact scenario was defined and run for Norsminde and Odense catchments using the local-scale models. It is the results from these impact scenarios that will be upscaled to the HYPE model. In the impact scenarios the N-leaching in the two catchments was redistributed so that the largest N-leaching rate is relocated to the area with the largest N-reduction and the smallest leaching rate to the area with lowest N-reduction. This means we simulate the effects of moving around the present land use and agricultural practice in the catchments without having to change it.

In the models, the redistribution of the present N-leaching is done at the MIKE-SHE grid scale (100 m in Norsminde and 200 m in Odense) by ranking the N-leaching and the total N-reduction (i.e. the sum of groundwater and surface water reduction) over all model grids in the catchments. We then relocate the leaching inputs, so that the highest ranked leaching grid is located at the grid with the highest ranked reduction and, followed by the second ranked leaching and reduction and so on (figure 4). This redistribution of N-leaching is only done between arable land grids.

To calculate the impact of this redistribution, the spatially targeted scenario is run through map-based N-load models for Norsminde and Odense developed in the study by Hansen et al. (Submitted). These models are based on the MIKE SHE/NLES models described in section 2.2.1, but are not dynamic in time. The models calculate a total mass balance for a given time period by multiplying maps of N-leaching, groundwater N-reduction and surface water reduction. The calculations are done for each grid cell in the catchments and then afterwards summed for the whole catchment.

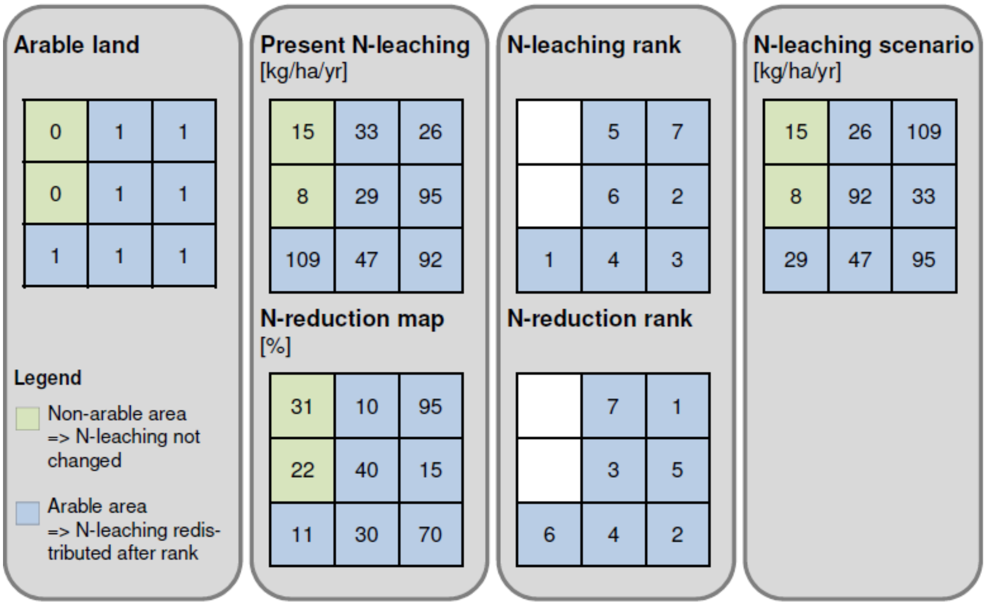

Figure 4 Spatially targeting of N-leaching based on N-reduction map. The N-leaching is redistributed so that the highest N-leaching is placed on the area with highest N-reduction and vice versa. This is done by ranking the N-leaching map and the N-reduction map. The N-leaching rate with rank 1 is then moved to the grid cell with N-reduction rank 1 i.e. in the above example the 109 kg/ha/yr in the lower left corner of the present N-leaching map is moved to upper right corner in the N-leaching scenario.

3. Results

3.1. Impact of a spatially targeted regulation in Norsminde and Odense

The results of the impact scenarios for Norsminde and Odense are seen on figure 5. In both catchments the spatial targeted regulation, where high leaching rates are redistributed to areas with high N-reduction and vice versa, is seen to result in a decrease of the total N-load out of the catchments. Since the present N-leaching is just spatially redistributed, i.e. the total input is unchanged for the catchments, the decrease in N-load is the caused by the percentage of nitrate being reduced in the groundwater increases. In Norsminde the groundwater reduction is increased by 8% and in Odense by 15% (figure 5).

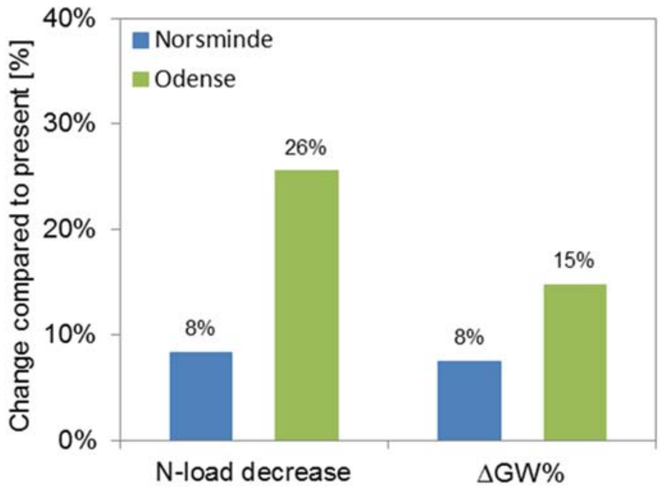

Figure 5 Impact of spatially targeted regulation in Norsminde and Odense catchment. On the graph is seen the resulting decrease in N-load at the catchment outlets and the increase in groundwater reduction percentage (ΔGW%).

3.2. Developing upscaling relationship for application at Baltic Sea Basin

The results gained from local-scale impact modelling for Norsminde and Odense must be upscaled to the rest of the Baltic Sea basin. We expect the impact of a spatially targeted regulation to be related to the local-scale spatial variation in groundwater reduction within a catchment, where a large variation will give a large impact and a small variation a small impact. However, no such information on local-scale variation is available at the Baltic Sea Basin scale. We show here that the impact can also be related to the fraction of arable land within a catchment i.e. the actual area where spatially targeted regulation can be applied. Since information about the fraction of arable land per subbasin is available in E-HYPE, this relationship can be utilized in the upscaling.

3.2.1. Assessing the impact based on arable land fraction

To estimate a relationship between the impact of spatially targeting and arable land, we have run the impact scenario described in section 2.4 for both Norsminde and Odense using different maps for areas in rotation and thus available for a spatially targeted regulation. These maps of arable land were based on the original map for the two catchments and a number of scenarios were then made by randomly adding or removing arable land to obtaining fractions of arable land between 0% (no areas in rotation, spatially targeting cannot be done on any areas) and 100% (all areas in the catchment can be used for spatially targeting).

This analysis resulted in a relationship between impact and arable land (figure 6) for Norsminde and Odense Catchments. We have fitted a 2nd order polynomial function to the data points in figure 6 (R^2 = 0.94):

$$\Delta GW\% = 0.26 * A^2 + 0.07 * A \qquad \textbf{(eq. 4)}$$

where *ΔGW%* is the increase in groundwater reduction percentage (GW%) and *A* is the fraction of arable land within catchment.

We expect this relationship between arable land and impact of targeted regulation to be scale dependent. Norsminde catchment (to the monitoring station) has a size of 85 km^2 and Odense catchment a size of 486 km^2. So we recommend not using this equation for catchments much above or below these catchment sizes. The median subbasin resolution in the E-HYPE model is 215 km^2, so it should be suitable using the equation with E-HYPE.

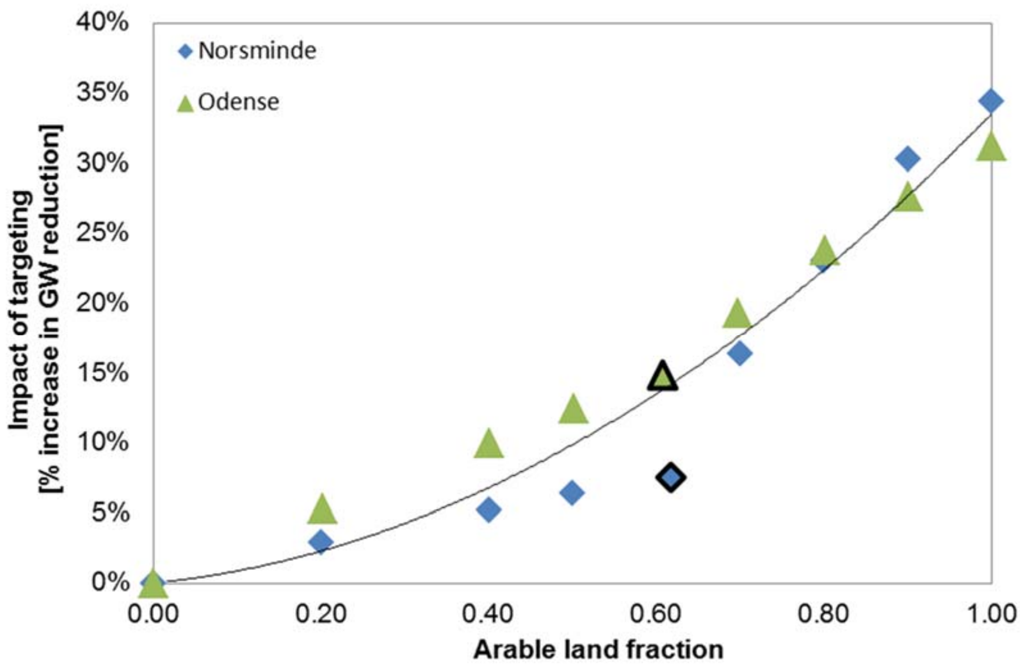

Figure 6 The impact of spatially targeting measures (increase in groundwater reduction) in Norsminde and Odense as a function of arable land (fraction of catchment area). The points with black border lines indicate the actual arable land % in Norsminde and Odense.

3.2.2. Changing the decay rate (denitrlu3) in HYPE

In order to have HYPE simulate the increase in groundwater reduction when a spatial targeted regulation is applied, the decay rate parameter for denitrification (denitrlu) must be increased. In the original HYPE model there is only one denitrification rate covering all three soil layers in HYPE. This means, that when the denitrification rate is changed it will affect both the N-reduction in the root zone (hence leaching) and the groundwater zone. In order to simulate the impact of spatially targeted regulation this needs to be two separate processes and the HYPE model code was therefore modified so that the denitrification rate for the groundwater zone in layer 3 can be changed independently of the root zone (denitrlu3).

Using equation 4 results in a different GW% increase for every subbasin in the Baltic Sea basin, which to simulate in E-HYPE requires a different increase in denitrlu3 for each of the 7,145 subbasins. Given that it is impractical to calibrate the increase in denitrlu3 for all of them, we propose a method to estimate the increase in denitrlu3 needed in each individual E-HYPE subbasin. This was done by studying how changes in the denitrlu3 parameter in E-HYPE affect the simulated change in GW%. To this we chose 10 subbasins spread across the Baltic Sea basin: Norsminde and Odense in Denmark, Kocinka and a small catchment near Gdansk in Poland, Parnu in Estonia, Pregolya in Poland/Kaliningrad Oblast (Russia) and Tullstorp and a small catchment south of Linköping in Sweden. For Linköping, Parnu and Pregolya, consisting of several subbasins in HYPE, the most downstream subbasin was chosen. For Odense we used all the 3 subbasins covering the Odense Fjord basin (therefore the names Odense 1, 2 and 3 in the following).

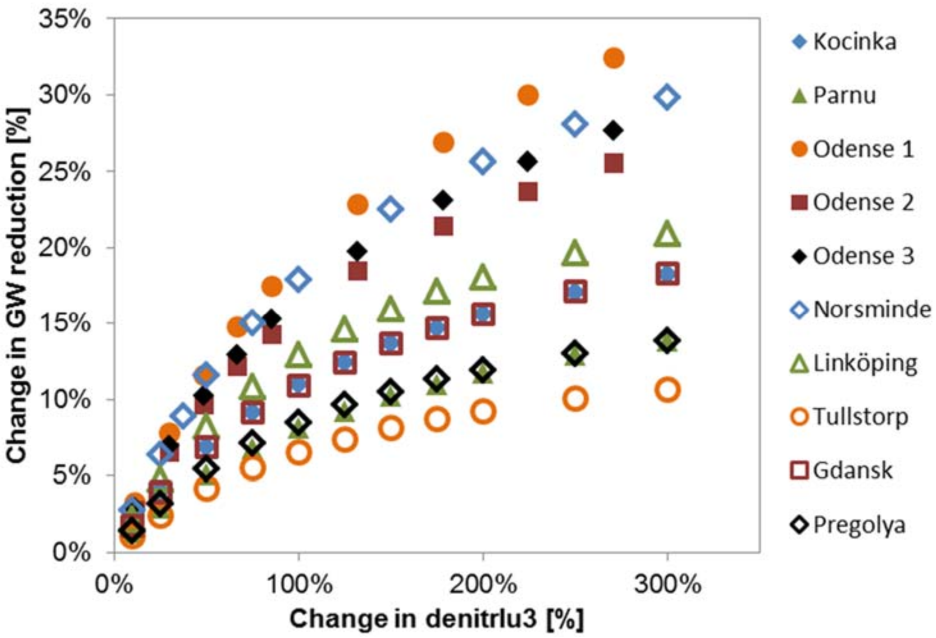

Figure 7 Change in decay rate denitrlu3 (percentage increase relative to initial value) and the simulated change in GW% (percentage increase relative to initial value) for 10 subbasins in HYPE

For these 10 subbasins we have analysed the change in GW% due to different changes in the decay rate denitrlu3 in layer 3 (figure 7). While the relationship between change in denitrlu3 and change in groundwater reduction was found to vary considerably between the different catchments, each of the catchments followed a clear relationship. Denitrification in HYPE is not only dependent on the decay rate denitrlu3, but also on soil moisture, soil water temperature, and nutrient levels within the soil (Lindstrom et al., 2010). To estimate how these factors affect the change in reduction with changes in denitrlu3, we first fitted individual logarithmic functions for each of the 10 catchments (all with $R^2 > 0.96$):

$$\Delta GW\% = a * \ln(\Delta Denitrlu3) + b \qquad \textbf{(eq. 5)}$$

where *ΔGW%* is the increase in GW% and *ΔDenitrlu3* is change in decay rate denitrlu3. The next step was to relate the *a* and *b* coefficients to some subbasin variables to describe how *a* and *b* varies between the 10 catchments. In this analysis 4 catchment variables, which are related to denitrification, were included:

Fraction of arable land, soil moisture in layer 3, soil water temperature in layer 3 and average N-leaching to layer 3.

A multiple linear regression analysis was done separately for coefficient *a* and *b*. Four different regression models were set up to test the effect of including 4 to 1 of the variables. The 4 variable models were constructed first, and then between each model the variable with the highest P-value (i.e. least statistical significance) was excluded. The 4-, 3- and 2-variable regression models were found to give almost the same curve describing the relationship between *ΔDenitrlu3* and *ΔGW* for the 10 subbasins. It was therefore chosen to use the 2-variable models:

$$a = 0.039 - 3.26E - 5 * SOIM + 0.0022 * N\_leach \qquad \textbf{(eq. 6)}$$

$$b = 0.099 - 7.52E - 5 * SOIM + 0.0043 * N\_leach \qquad \textbf{(eq. 7)}$$

where *SOIM* is soil moisture in layer 3 and *N_leach* is the average N-leaching to layer 3. When putting these equations for *a* and *b* back into the logarithmic function (eq. 5) we get the following expression to estimate the logarithmic model describing the relationship between *ΔDenitrlu3* and *ΔGW%* for a subbasin:

$$\Delta GW\% = (0.039 - 3.26E - 5 * SOIM + 0.0022 * N\_leach) * \ln(\Delta Denitrlu3) + (0.099 - 7.52E - 5 * SOIM + 0.0043 * N\_leach) \qquad \textbf{(eq. 8)}$$

The logarithmic models for each of the 10 subbasins are seen in figure 8A and B. The models for Norsminde, Kocinka and Gdansk are seen to perform well compared to the HYPE data points (RMSE for each model between 1.3-1.5%). For Kaliningrad, Linköping, Tullstrup and Odense2 the models perform reasonably (RMSE 2.4-2.9%), but for Parnu, Odense3 and Odense 1 the models do not perform very well (RMSE 3.1-4.5%).

When applying the upscaling it is the *ΔDenitrlu3* we want to estimate based on the *ΔGW%* estimated using equation 4. We therefore have to re-arrange equation 8 to the following upscaling relationship:

$$\Delta Denitrlu3 = \exp\left(\frac{\Delta GW\% - (0.099 - 7.52E - 5 * SOIM + 0.0043 * N\_leach)}{(0.039 - 3.26E - 5 * SOIM + 0.0022 * N\_leach)}\right) \qquad \textbf{(eq. 9)}$$

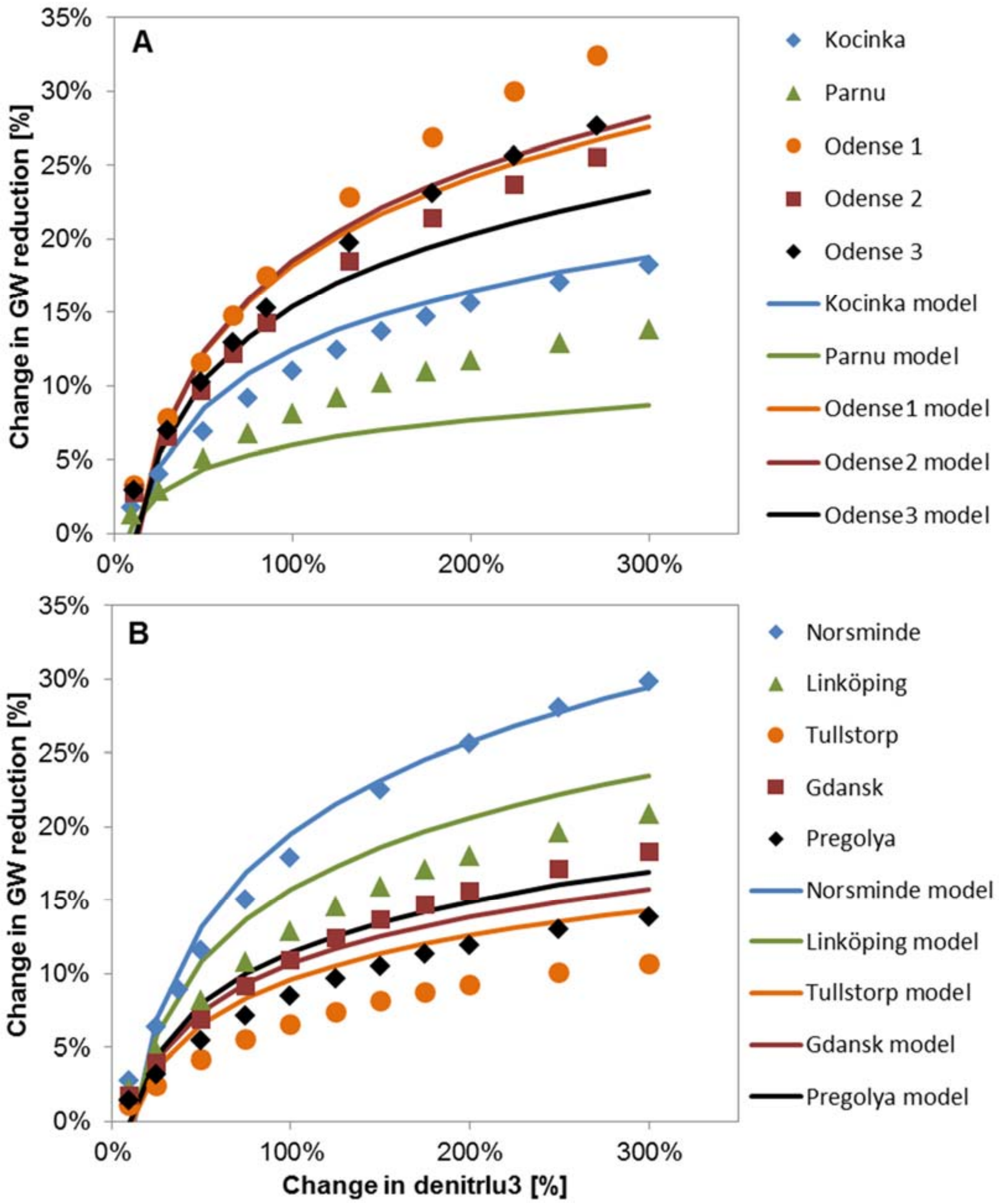

Figure 8 2-variable models for the 10 subbasins describing the relationship between change in decay rate parameter denitrlu3 and change in GW%

3.3. Test of upscaling approach

3.3.1. Step 1 - Baseflow fraction

The results from checking the baseflow fraction for E-HYPE is shown in table 1. For Norsminde, the local-scale MIKE SHE model is seen to have a baseflow fraction very close to the observed hydrograph, while the HYPE model has a higher baseflow fraction. For Odense the baseflow fractions for both MIKE SHE and HYPE are very close to the observed. In all cases the agreements are within the +/- 20% acceptance interval of the observed baseflow fraction and HYPE is therefore not recalibrated for flow. The simulated discharge at the most downstream station in Norsminde is seen in figure 9A for both the MIKE SHE model and HYPE model.

Table 1 Baseflow fraction (BF%) of total flow for the period 1/1/2000 – 31/12/2003 in Norsminde and Odense catchments. The baseflow filter BFLOW pass 3 has been used. The error of the BF% is compared to the observed BF%.

	Norsminde		Odense	
	BF%	Error	BF%	Error
Observed Q	53%		65%	
MIKE SHE	52%	-1%	62%	-3%
HYPE initial	70%	17%	71%	6%
HYPE recalibrated	70%	17%	71%	6%

3.3.2. Step 2 – N-reduction in groundwater

Table 2 shows the mass balance for Norsminde for the local-scale MIKE SHE model, the initial HYPE and recalibrated HYPE model as well as the target values. The target value for N-leaching is 38 N/ha/yr and for groundwater reduction 55%. The N-leaching and groundwater reduction in the MIKE SHE/NLES model is seen to be very similar to the target values. The initial HYPE model overestimates both target values with a N-leaching of 55.8 kg N/ha/yr and a groundwater reduction percentage of 63%. The HYPE model was therefore recalibrated resulting in a N-leaching of 31.3 kg N/ha/yr and a groundwater reduction percentage of 52% for the recalibrated HYPE model.

In figure 9B is seen the simulated N-load at the downstream monitoring station in Norsminde for the MIKE SHE model and the HYPE model. The MIKE SHE model is seen to simulate a N-load rather close to the observed. The initial HYPE model is seen to greatly overestimate the N-load, but after the recalibration the HYPE simulated N-load is greatly improved.

For Odense the target values for calibrating the N-reduction in groundwater and the N-leaching also originates from the alternative data sources and are seen in table 3. The N-leaching in the MIKE SHE/NLES model is seen to be similar to the target value, but for groundwater reduction the MIKE SHE/NLES model is very different from the target value, because the model assumes that all reduction in the catchment occurs in groundwater. The initial HYPE model has a N-leaching of 59.7 kg N/ha/yr and a groundwater reduction percentage of 68% and thereby overestimates both target values. The HYPE model for Odense was therefore recalibrated and this resulted in a N-leaching of 41.8 kg N/ha/yr and a groundwater reduction of 45%. The simulated N-load at the most downstream monitoring station is 17.1 kg N/ha/yr in the recalibrated HYPE model. This is close to the observed N-load at the station of 16.7 kg N/ha/yr.

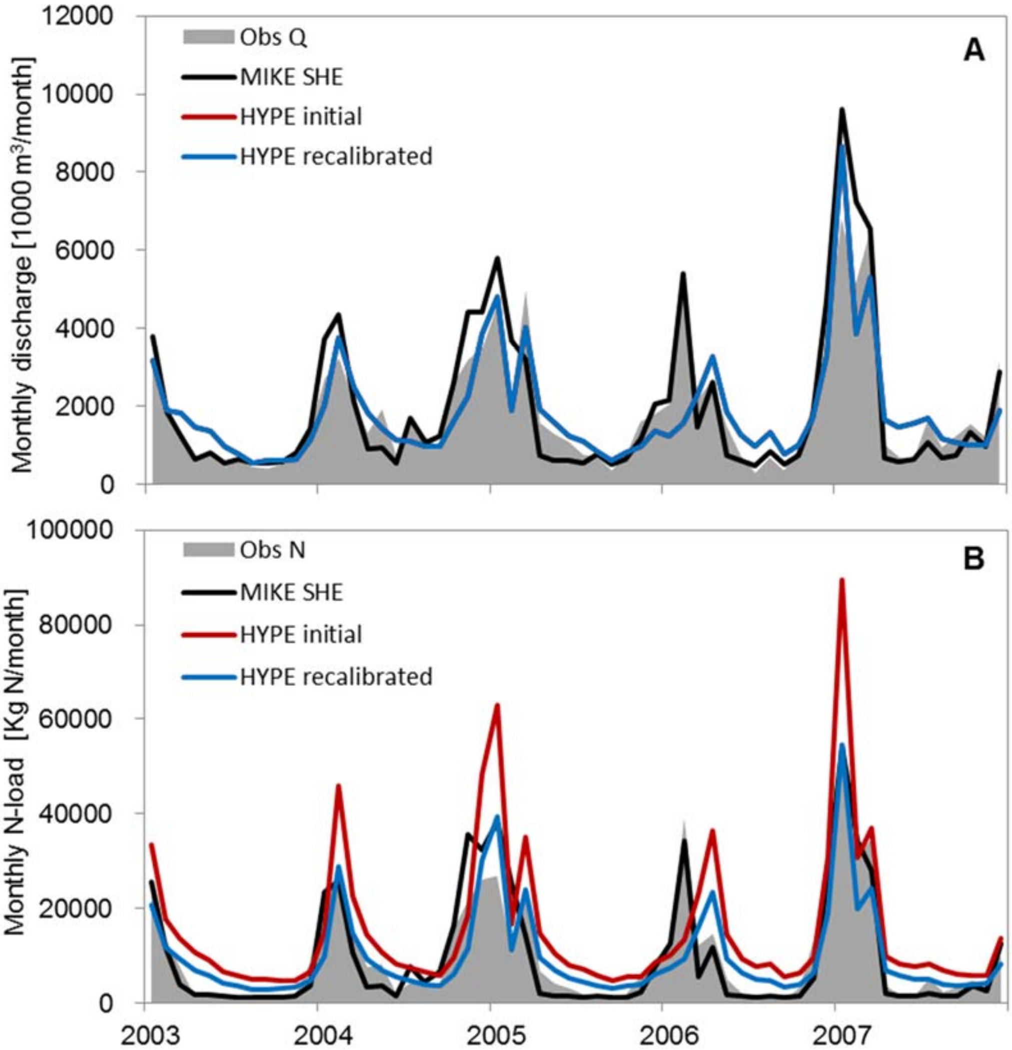

Figure 9 Monthly discharge (A) and N-load (B) at the downstream monitoring station in Norsminde from the MIKE SHE model and the HYPE model before and after upscaling. On the graph is also seen the observed discharge and observed monthly N-load (estimated by interpolating between observations)

Table 2 Mass balance for Norsminde catchment to the most downstream monitoring station for the period 2000-2010. The numbers marked with green are the target values for the re-calibration of HYPE.

Norsminde Unit: Kg N/ha/yr	**Target values**	**MIKE SHE/NLES**	**HYPE initial**	**HYPE recalib**	**HYPE impact**
N-fluxes					
N-leaching (eq. 1)	37.8	35.3	55.8	31.3	31.3
N-load to stream (eq.2)		16.5	20.7	15.1	11.5
Other N sources to stream		1.8	4.5	4.5	4.5
Total N-load to stream		18.3	25.2	19.6	16.0
N-load at monitoring st.		14.6	22.6	14.6	12.1
Groundwater/surface water reduction					
GW reduction		-18.8	-35.0	-16.3	-19.8
GW%[1]	55%	53%	63%	52%	63%
SW reduction		-3.7	-2.7	-5.0	-3.9
SW%[2]		20%	11%	26%	24%

1) The groundwater reduction percentage (GW%) is defined as the GW reduction divided by L_{M_leach}.
2) The surface water percentage (SW%) is defined as the SW reduction divided by the GW load to stream plus other N sources to stream.

Table 3 Mass balance for Odense catchment to the most downstream monitoring station for the period 2000-2010. The numbers marked with green are the target values for the re-calibration of HYPE.

Odense Unit: Kg N/ha/yr	**Target values**	**MIKE SHE/NLES**	**HYPE initial**	**HYPE recalib**	**HYPE impact**
N-fluxes					
N-leaching (eq. 1)	37.5	35.1	59.7	41.8	41.8
N-load to stream (eq.2)		12.9	19.4	22.8	17.1
Other N sources to stream		2.9	3.0	3.0	3.0
Total N-load to stream		15.8	22.4	25.8	20.1
N-load at monitoring st.		15.8	20.8	17.1	13.5
Groundwater/surface water reduction					
GW reduction		-22.2	-40.3	-19.0	-24.6
GW%[1]	48%	63%	68%	45%	59%
SW reduction		0	-1.6	-8.7	-6.6
SW%[2]		0%	7%	34%	33%

1) The groundwater reduction percentage (GW%) is defined as the GW reduction divided by L_{M_leach}.
2) The surface water percentage (SW%) is defined as the SW reduction divided by the GW load to stream plus other N sources to stream.

3.3.3. Step 3 - Simulating impacts of spatially targeted regulation with E-HYPE

The increase in groundwater reduction (ΔGW%) due to a spatial targeted regulation is first estimated for Norsminde and Odense using equation 4 and the arable land fraction in E-HYPE (table 4). The estimated ΔGW% for Norsminde is 22% and for Odense 21%. Then using equation 9 the required change in decay rate denitrlu3 to increase the groundwater reduction is estimated to be an increase of 137% in Norsminde and 167% in Odense. For Odense the area-weighted average of the catchment variables for the 2 subbasins (Odense 1 and 2) covering the downstream monitoring station are used for estimation of ΔGW% and ΔDenitrlu3. The estimated ΔDenitrlu3 values are applied to the decay rate in model layer 3 for the two catchments to run the impact scenario. In Odense the estimated ΔDenitrlu3 of 167% is applied to both subbasins.

The resulting mass balance for the impact scenario for Norsminde is seen in table 2 and for Odense in table 3. For Norsminde the groundwater reduction is increased from 53% to 63% which is an increase of 22% and for Odense the groundwater reduction is increased from 45% to 59% which is an increase of 30% (table 4).

The HYPE simulated impact of a spatially targeted regulation is plotted against the local-scale model simulated impact in figure 10. The E-HYPE simulated impact is seen to be larger than the local-scale model simulated impact for both Norsminde and Odense. The main reason for this is that the arable land fraction, used to estimate the increase in groundwater reduction, in the E-HYPE model is larger than in the local-scale models. This is a question of different input data between the local-scale models and E-HYPE. For Norsminde the E-HYPE simulated impact plots nicely on the curve describing the relation between the arable land and the impact, whereas for Odense the E-HYPE simulated impact plots above the curve. The equation (eq. 9) to estimate ΔDenitrlu3 works well for Norsminde but less well for Odense, which was also seen when developing the equation (figure 8).

The decrease in N-load for Norsminde is seen to be overestimated with E-HYPE compared to the local-scale model due to the overestimation of the increase in groundwater reduction (figure 11). In Odense however, the N-load decrease simulated with E-HYPE is smaller than the local-scale model result, even though the increase in groundwater reduction was overestimated with the upscaling methodology.

Table 4 Catchment variables used to estimate ΔGW and ΔDenitrlu3, the estimated ΔGW and ΔDenitrlu3 (using equation 4 and 9) and the HYPE simulated ΔGW after changing decay rate according to the estimated ΔDenitrlu3. For Odense the area-weighted average of the catchment variables for the 2 subbasins (Odense 1 and 2) covered by the downstream monitoring station are used and the estimated ΔDenitrlu3 is applied to both subbasins.

	Arable land [%]	Soil moisture in layer 3 [mm]	Avg. N-Leaching to layer 3 [kgN/ha]	Estimated ΔGW [%]	Estimated ΔDenitrlu3 [%]	HYPE simulated ΔGW [%]
Norsminde	80%	355	28.7	22%	137%	22%
Odense	81%	699	32.0	23%	167%	30%

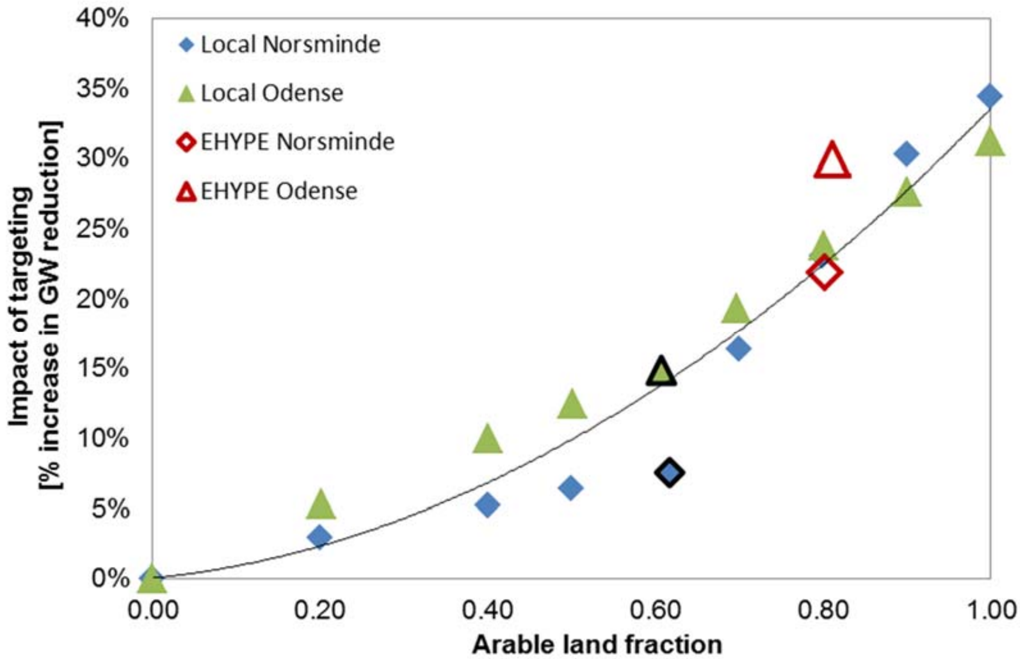

Figure 10 E-HYPE simulated impact of a spatial targeted regulation compared to impact simulated with the local-scale models for Odense and Norsminde

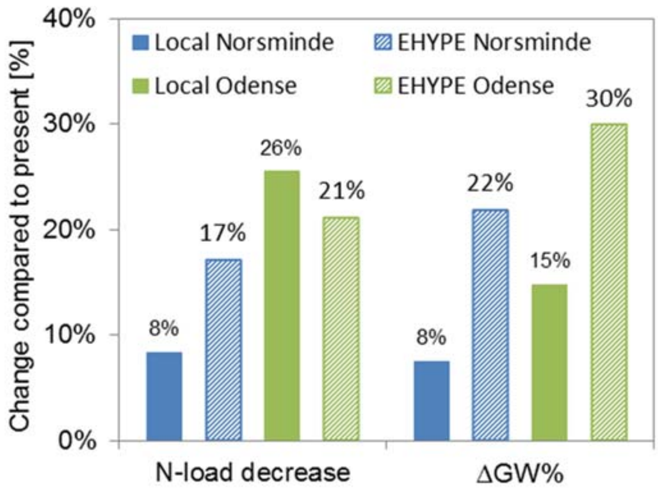

Figure 11 Simulated decrease in N-load and increase in groundwater reduction due to spatially targeted regulation for the local-scale models and E-HYPE

4. Discussion and conclusion

In this paper we have presented a methodology to upscale knowledge on the impact of a spatially targeted regulation from two local-scale physically based MIKE SHE catchment models to the large-scale and more conceptual E-HYPE model with the aim of using this model to simulate changes for the Baltic Sea basin. We use a dynamic upscaling approach in a manner similar to Bronstert et al. (2007), but we have extended this approach from only considering flow to also including N-transport and reduction. We have used the local-scale models to develop a relationship between the impact of spatially targeted regulation in a catchment and the fraction of arable land and subsequently derived a relationship between the change in the HYPE decay parameter denitrlu3 needed to simulate this impact and two catchment characteristics available in HYPE. We are only aware of one similar example in literature (Bronstert et al., 2007) of using a local-scale model to infer how parameters should be modified in the large-scale model to simulate something the large-scale model can otherwise not simulate.

We have tested the upscaling approach for Norsminde and Odense catchments and we overestimate the increase in groundwater reduction for both catchments. The main reason for this is that the fraction of arable land, which is used to estimate the impact, in E-HYPE is higher than in the local-scale models. Arable land fractions in the local-scale models are based on data from the Danish General Agricultural Register. Currently the fraction of arable land in E-HYPE is derived from the CORINE database for the EU (smallest mapping unit 25 ha) and GLOBCOVER 2000 for areas outside the EU at a resolution of 1 km for the year 2000. Assuming that the CORINE data give a reasonably correct estimate of the arable land for the entire Baltic Sea basin, these local differences will generate errors at catchment scale (Norsminde and Odense), but are likely to cancel out on the Baltic Sea basin scale. Taking into account this difference in arable land fractions, the increase in GW% was estimated very well for Norsminde but overestimated for Odense. Hence, we suggest that the error is acceptable, but that uncertainties of at least this magnitude should be taken into account when analysing the upscaled scenario results.

There are some critical assumptions made in the upscaling methodology. In the methodology it is implicitly assumed that the local-scale models are more correct than the large-scale model. Supported by the study by Stisen et al. (2011) we argue that local-scale models that are able to incorporate a wider range of data in the model, and not least using more data types for calibration, will be more constrained and hence provide a more narrow prediction uncertainty interval for small scale applications than large-scale models using less data and focusing on water and nutrient balances for rather large catchments. Nevertheless, the results from the local-scale models are associated with considerable uncertainties related to input data, model structure and parameters (Refsgaard et al., 2007; Hansen et al., 2014). This is illustrated by the surface water reduction in the local-scale Odense model and the National N-model. The first assumes negligible reduction based on the study by Hansen et al. (2009), while the latter estimates a considerable reduction based on empirical models for reduction in streams, wetlands and lakes (Højberg et al., 2015).

The most critical assumption in the upscaling methodology is that we assume our two case studies, Odense and Norsminde, to be representative for the entire Baltic Sea Basin. Ideally, more catchments representing different climates, geologies, agricultural growing regions and farm structures should be included in developing the upscaling relationships. It would also have been a good idea to test the approach on an independent catchment. However, currently Norsminde and Odense catchment are the only study areas where local-scale models have been set up with N-reduction maps with sufficient spatial details to support spatially targeted regulation decision making.

Some issues in relation to the construction of the impact scenario run in the local-scale models may in practice constrain the impact from a spatial targeted regulation. When constructing the scenarios, it is not considered whether the redistribution made is possible in reality. The estimated impacts of a spatially targeted regulation in Odense and Norsminde are therefore maximum potential impacts (Hansen et al., Submitted).

Altogether, the upscaling methodology provides an opportunity to simulate how spatially targeted regulation may affect nutrient loads to the Baltic Sea. Given all the recognised uncertainties, we conclude that E-HYPE using this upscaling methodology is likely to be able to simulate the correct trends and order of magnitudes at the Baltic Sea basin scale and as such provides a sound base for large scale policy analysis. However, we do not expect it to be sufficiently accurate to be useful for detailed designing of local-scale measures.

The general principles in the presented upscaling approach can be used for other model codes and for upscaling of other processes than N-reduction in groundwater. The practical implementation of the upscaling approach is model and process dependent and all applications will therefore be unique. The upscaling relationships presented in this study are therefore not transferable to other processes and model codes. In order to make it possible to use the upscaling approach it is required that there is some degree of similarity between models in order to design a learning scheme. The upscaling relationships must also be based on some proxy data readily available on large scale.

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Appendix B

Review and assessment of nitrate reduction in groundwater in the Baltic Sea Basin

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Abstract

Riverine load of nitrogen to the Baltic Sea has been reduced in recent years, but further amendments are required to meet the goal of the EU Water Framework Directive. The largest contributor from anthropogenic activities is agriculture and reduction in the load from farming praxis is inevitable. Regulation of nitrogen has typically relied on uniform regulation, i.e. imposing the same restrictions on farming all over. During transport from the field to the sea nitrogen undergoes natural reduction, but with large spatial variations, due to variation in the hydrogeological and hydro-geochemical conditions. Mapping this variation would allow more optimal regulation strategies, where most restrictions are imposed in areas with low natural reduction. Most assessments on nitrogen reduction take a catchment scale approach in which the total removal for the catchment is estimated, not differentiating between surface water and groundwater. Discriminating between the two domains is nevertheless important in order to identify the correct type of mitigation measure to implement. In the present study, a map for nitrate reduction in groundwater is developed by groundwater experts from five countries in the Baltic Sea Basin based on a review of data and previous studies. The study shows significant variation in groundwater reduction between the countries and within most of the countries, indicating that different mitigation measures and strategies may be optimal for the different countries.

Keywords: Baltic Sea; nitrate reduction; groundwater

1. Introduction

Nutrient load, primarily by waterborne riverine transport, has changed the environmental conditions for the Baltic Sea from oligotrophic to eutrophic conditions in most parts (Larsson et al., 1985). To revert the conditions, the Baltic Sea Action Plan (BSAP) was adopted in 2007 (HELCOM, 2007) and revised in 2013 (HELCOM, 2013). In the most recent pollution load compilation (PLC5.5) it is estimated that nitrogen loads has been reduced by 9% from the reference period (1997 – 2003) to 2013, but it is also found that a further amendment of 14% is required (HELCOM, 2015). The estimated required amendment varies significantly with a reduction requirement of 26% in the Baltic Proper as being the sub-basins with the highest reduction requirement. Furthermore, specific estuaries and coastal waters may require even higher specific abatements in order to protect coastal and transitional water ecosystems and comply with the good status objectives of the Water Framework Directive (e.g. Hinsby et al., 2012).

A significant part of the nutrient reduction to the Baltic Sea can be attributed major developments in the wastewater treatment, due to technological developments and more people being connected to municipal wastewater treatment plants. In combination with the improvement of fish farms, direct nitrogen input from point sources to the Baltic Sea has been reduced by 43% between 1994 and 2010 (HELCOM, 2015). Although some further reduction in nutrient load to the Baltic Sea may be realised by improved wastewater treatment, the fifth Baltic Sea pollution load compilation (PLC-5) (HELCOM, 2011) estimates that the diffuse sources with a share of approximately 45% constitutes the largest anthropogenic contribution of riverine nitrogen of which 70 – 90% is estimated to originate from agriculture.

During transport from the root zone to the discharge into the sea nitrogen may be removed in either the groundwater or the surface water system. Removal occurs by different natural biogeochemical processes or sedimentation, often referred to as retention or reduction and expressed as a percentage removal. The magnitude of the retention/reduction depends on the actual hydro-biogeochemical conditions and may vary significantly. Applying the statistical model MESAW to the 117 drainage basins in the Baltic Sea, Stålnacke et al. (2015) estimated a total surface water N retention of approximately 40%. As noted by the authors, this estimate is substantially higher than the estimate by Mörth et al. (2007), reporting a mean in-stream N retention of 15% in the Baltic Sea rivers. Combining a statistical N-leaching model with a fully distributed groundwater/surface water hydrological model and statistical surface water retention models, Højberg et al. (2015) developed a national nitrogen model for Denmark. They estimated mean retentions of 63% and 25% for groundwater and surface water, respectively, but with large spatial variations.

Natural removal of nitrogen is, however, generally not sufficient to reduce the diffuse N loads to the required levels. Implementing various N mitigation measures has successfully reduced the diffuse loads from several of the HELCOM countries. These measures have typically been implemented in response to a general and uniform regulation, i.e. a regulation imposing the same restriction in all areas without considering the variation in the natural reduction of nitrogen. Reaching a further abatement in nitrogen load calls for new and innovative measures and regulation strategies, where measures are targeted towards areas, where the natural retention is low and the measures thus most cost-effective (Jacobsen and Hansen, 2016). A wide range of different mitigation measures may be employed to combat the N load from diffuse sources. These may be located on the agricultural fields, e.g. catch crops, at the edge of the fields, such as constructed wetland (MST, 2015) or in the surface water system upstream the outlet to the Baltic Sea. Efficiency of the mitigation measures varies according to the type of measure and where in the hydrological regime they are located. Studies devoted the estimation of optimal location of measures have primarily studied on-field measures, based on the estimation of nitrogen leaching from the root zone and possible protection areas (Hirt et al., 2012; Andersen et al., 2016; Hiscock et al., 2007; Kunkel et al., 2008; Rode et al., 2009). Most studies only consider either the groundwater or surface water system, but fail to include the entire hydrological system and are thereby not able to evaluate whether the optimal location of measures are on-field or in-stream.

Studies at the Baltic Sea scale (Stålnacke et al., 2015) and for Finnish drainage basins discharging to the Baltic Sea (Lepistö et al. 2006; Huttunen et al. 2016) have found a generic relationship between N retention and the areal fraction of lakes in the different drainage basins, indicating that retention in lakes is dominating, when the percentage of the area covered by lakes is high. On the other hand, studies in other Baltic Sea basins with a small percentage of lakes indicate that retention is dominated by subsurface processes (Hansen et al., 2009 Hesse et al., 2013; Windolf et al., 2011). Although reduction in groundwater is an important process, it is most commonly only included in a lumped approach in nutrient modelling studies. In a review of the current state of distributed catchment nutrient water quality modelling Wellen et al. (2015) find that most modelling concepts are based on empirical descriptions of groundwater at sub-catchment level, and of the 275 scientific studies included in the review they report only one study that includes directly simulations of groundwater flow. Some recent studies emphasizes the need to include the groundwater nitrate transport more explicitly in the models (Hesser et al., 2010; Rode et al., 2010), but such studies have primarily been restricted to detailed small scale studies, with few exceptions such as the Danish national nitrogen model (Højberg et al., 2015).

Knowledge on how much and where nitrogen is degraded in the groundwater is a prerequisite for designing optimal cost-effective mitigation measures utilising the natural nitrogen reduction in the subsurface. Spatially variable nitrate reduction in groundwater has, however, only been considered by few studies, either by detailed geochemical modelling (Wriedt and Rode, 2006; Hesser et al., 2010), or by the estimation of travel times and the association of different first-order decay rates for different geological formations (Kunkel et al., 2008; Wendland et al., 2004; Tetzlaff et al., 2013). Assuming the subsurface to be divided into an upper oxic part with no nitrate transformation and a lower reduced part with instantaneous removal of nitrate, Hansen et al. (2014b) studied the impact of a spatially heterogeneous redox interface on the reduction of nitrate in the groundwater. No previous study has focused on the spatial heterogeneity at the Baltic Sea scale. The objectives of the present study are thus to:

- Provide a review of studies addressing nitrate reduction in groundwater in the Baltic Sea Basin
- Develop a map of nitrate reduction in groundwater in the Baltic Sea Basin with best possible spatial resolution, based on a compilation of exiting knowledge

2. Materials and Methodology

2.1. Approach

Nitrate reduction in groundwater has been assessed to various degrees by the countries in the Baltic Sea Basin. While laboratory and plot to small catchment scale studies have been undertaken in most of the countries, few have assessed it at regional or national scale and evaluated the importance for the total nitrogen load to the Baltic Sea. In Denmark, Sweden and Germany regional to national scale models have been developed, which have been used to quantify the N-reduction in groundwater and its spatial variation. For the other countries there is no reporting in the international literature on N-reduction at national scale, and information was thus insufficient for the construction of a groundwater N-reduction map for the entire Baltic Sea Basin.

As part of the BONUS project Soils2Sea (www.soils2sea.eu) a workshop with national experts was therefore arranged with aim of 1) providing an updated overview of national studies on nitrate reduction in groundwater at various scales, and 2) develop an approach to quantify N-reduction in groundwater based on existing studies in the countries and available data at national scale. Representatives from Finland, Lithuania, Poland, Sweden and Denmark participated in the workshop.

2.2. Definitions

The major source of nitrogen to the surface water system originates from leaching under agricultural fields, and is predominantly present in the form of nitrate. Reduction of nitrogen in groundwater is thus similarly related to nitrate, which is the primary focus of the present study and the term nitrogen (N) refers to nitrate unless specified otherwise. Differences in the amount of nitrate leaving the root zone and found as riverine load may be caused by two processes: 1) retention, which is the temporal storage of nitrogen, typically in surface water sediments, and 2) reduction that is the permanent removal of nitrate. Despite the differences in the two processes the term retention is commonly used to cover both, especially in surface water research. While the term reduction will primarily be used to indicate removal of nitrogen in the present study, the terms will be used loosely.

Nitrate reduction is in the present study defined as the difference between the nitrogen lost from the root zone and found in the surface water system. Using the notation in Figure 1, reduction in the groundwater ($Nred_{GW}$) is given by:

$$Nred_{GW} = \left(1 - \frac{DRN_{GW \rightarrow RIV + GW \rightarrow RIV}}{RZ \rightarrow GW}\right) X\ 100\% \qquad \text{(eq.1)}$$

Where RZ→GW is N-leaching from the root zone to the groundwater zone, DRN_{GW}→RIV is transport from the groundwater to the surface water system by drainage and GW→RIV is transport from groundwater to the surface water system.

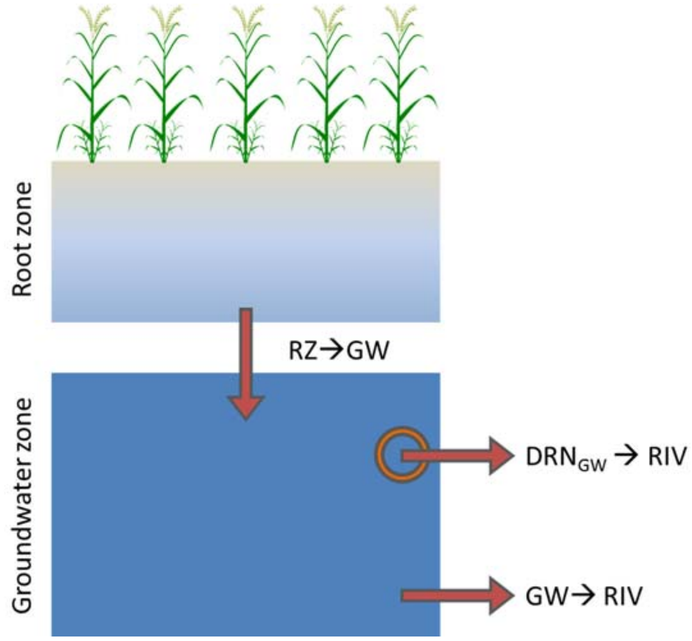

Figure 1 Concept figure for nitrate transport in the root zone and the groundwater zone

2.3. Site description

The total Baltic Sea drainage basin comprises 1,720,270 km², which is just above four times larger than the area of the Baltic Sea itself. The Basin spans large climatic variations with annual mean temperatures and precipitation ranging from 0°C and 400 mm in north to 10°C and 1000 mm in south. Agriculture takes up 60-70% of the land in the southern countries Denmark, Germany and Poland. Forestry becomes more important towards the north, and agriculture only accounts for approximately 10% in Finland and Sweden, while Latvia, Estonia and Lithuania are in the middle range with 30-50% agricultural land (HELCOM, 2004). The most intensive agriculture in terms of nutrient application is found in Denmark, Germany and southern part of Sweden, and the least intensive countries are Latvia, Estonia and Lithuania (HELCOM, 2015). More than 84 million people live in the Baltic Sea Basin and since the early 2000's there has been a steady increase in the percentage of the population connected to secondary and tertiary wastewater treatment plants.

3. State-of-the-art in the Baltic Sea Basin

3.1. Nitrate reduction processes in groundwater

3.1.1. Redox conditions

Nitrate can be transformed naturally by reduction where nitrate acts as the electron acceptor. The reduction process have several intermediate stages (NO_2^-, NO and N_2O), but N_2 is the predominant reaction product. Reduction of oxygen has the highest energy yield and is thermodynamically preferred and nitrate reduction therefore only occurs under anaerobic conditions. Furthermore, an electron donor must be present for nitrate reduction to occur. In Quaternary glacial sediments the most important electron donors in the reduction of nitrate are organic carbon, pyrite (FeS_2) and structural Fe^{+2} in some minerals (Pedersen et al., 1991; Postma et al., 1991; Korom, 1992; Rodvang and Simpkins, 2001; Appelo and Postma, 2005). Dissimilatory nitrate reduction to ammonium (DNRA) has recently been shown to occur under certain conditions (Behrendt et al. 2013; Necpalova et al. 2012).

Nitrate reduction by organic carbon is referred to as denitrification and is a well-documented process in the saturated zone (Bradley et al., 1992; Starr and Gillham, 1993; Klimas, 1996). Oxidation of organic carbon is catalysed by microorganisms (Korom, 1992) and the reactivity of the organic matter is often controlling the reaction rate (Appelo and Postma, 2005). Nitrate reduction by pyrite has also been reported in several studies (Kolle et al., 1983; Pedersen et al., 1991; Postma et al., 1991; Robertson et al., 1996; Jorgensen et al., 2009) and is also mediated by microorganisms (Appelo and Postma, 2005; Jorgensen et al., 2009). Nitrate reduction by organic carbon is thermodynamically favoured over pyrite (Korom, 1992). However, the sequence of these two reactions is also determined by reaction kinetics. Studies have shown that the reaction with pyrite can be more important than organic carbon in cases where the reactivity of the organic matter is low e.g. if the organic matter is old or has a high molecular weight (Kolle et al., 1983; Postma et al., 1991; Wriedt and Rode, 2006). Finally, the reaction with structural Fe^{+2} in minerals (clay minerals and some silicate minerals) has been reported as both a microbial (Ernstsen et al., 1998b; Weber et al., 2001) and chemical process (Postma, 1990; Ernstsen, 1996; Ernstsen et al., 1998a).

The total reduction potential available in a sediment for reducing nitrate (or oxygen) can be expressed by the term redox capacity, which is the total amount of reduced compounds (organic carbon, pyrite, Fe^{+2}) in the sediment and can be estimated as a lumped measure not discriminating between the individual reduced species. The content of reduced compounds is lower in sandy materials than clayey materials (Ernstsen et al., 2001; Ernstsen, 2013; Postma et al., 1991). In Figure 2 is shown measured amounts of reduced compounds at different depths in a borehole located in the clayey till Lillebæk catchment in Denmark. The transition from low to high amounts of reduced compounds at approximately 7 m depth corresponds to the redox interface.

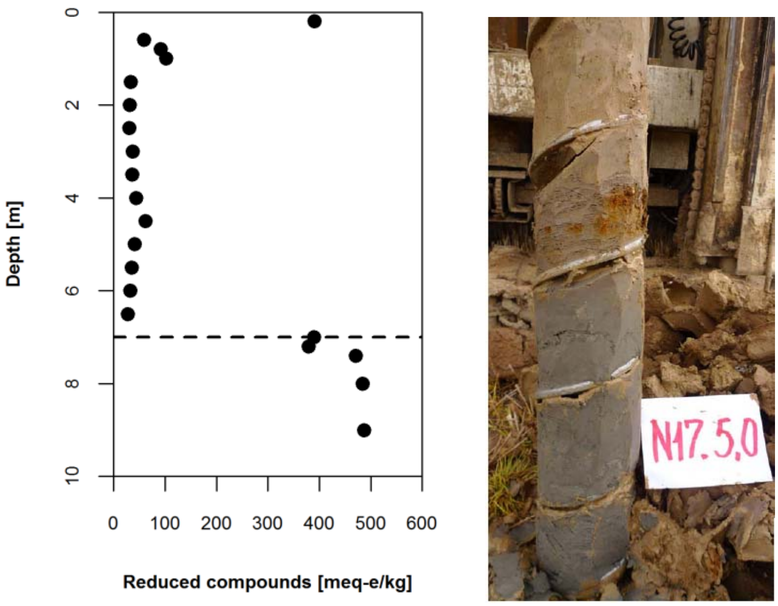

Figure 2 Amount of reduced compounds (meq e–/kg) at different depths in a till sediment (left), figure from Hansen et al. (2014a), and (right) colour change in till sediment from brown to grey indicating the location of the redox interface, picture from Ernstsen (2013)

The redox condition of the sediment and location of the redox interface can be estimated directly in the field from the sediment colours (Ernstsen and Morup, 1992; Ernstsen, 1996; Hansen et al., 2008; Pedersen et al., 1991; Robertson et al., 1996). Oxidized sediments have red, yellow and brown colours, whereas reduced sediment have grey and black colours. The change from oxidized to reduced conditions at the redox interface can therefore be observed as a change in the sediment colour, Figure 2. The redox potential can also be measured by the voltage difference between an inert and a standard hydrogen electrode. In aerated soils the applicability of redox potential has been criticized (Bartlett and James 1995), but under reduced conditions redox potential is found to give results that are comparable to values obtained in the laboratory (e.g. Connell and Patrick 1968, Pan et al. 2014) and in fields (Patrick et al. 1996, Mansfeldt 2004), especially when iron and/or sulphur are present in abundance.

3.1.2. Spatial and temporal variations

In areas where the stratigraphy is more or less uniform a well-defined redox- interface can be found. A well-defined upper oxic zone above an anoxic zone was e.g. found by Postma et al. (1991) in the sandy Rabis Creek, Denmark, and in Finnish acid sulphate soils by Palko (1994). In areas where the stratigraphy is more complex multiple redox interfaces can exist (Robertson et al., 1996, Ernstsen, 2013), and anaerobic micro-environments can be found above the redox interface (Fujikawa and Hendry, 1991; Pedersen et al., 1991; Ernstsen et al., 1998b).

The spatial variability of depths to the redox interface can locally be large. Several studies have reported that the location of the redox interface in tills can vary several meters over short horizontal distances (Fujikawa and Hendry, 1991; Ernstsen, 1996; Hansen et al., 2008). Spatial heterogeneity of the redox interface have been adressed by Hansen et al. (2008) and Hansen et al. (2014a) by means of variogram analysis. Using borehole data with a spacing of 200 m Hansen et al. (2008) found no correlation in observed redox depths in a clayey till area on the island of Funen. Using a finer spacing between cores Hansen et al. (2014a) found a correlation length of 289 m for a clayey till area in on the east coast of Jutland.

The redox capacity in a sediment is being depleted when nitrate and also oxygen is reduced. Due to the continuous supply of oxygen and nitrate with recharging water the redox interface is therefore not static but moving downwards with time (Bohlke et al. 2002, Postma et al. 1991, Robertson et al. 1996, Wriedt and Rode 2006). The depth to the redox interface is thus dependent not only on the redox capacity, but also the age of the sediment, the downward water flux and land use, i.e. whether nitrogen has been applied on the surface or not (Ernstsen et al., 2001; Virtanen et al. 2016). Today both oxygen and nitrate act as oxidants, but nitrate input has only been high for the last 60-70 years. For pre-anthropogenic conditions, with only input of oxygen, Robertson et al. (1996) reported a vertical migration rate of the redox interface of 0.04 cm/yr in till sediments of Wisconsian age. In sandy aquifer materials Bohlke et al. (2002) reported a vertical rate of 0.26 cm/yr and Postma et al. (1991) reported a vertical rate of 0.34 cm/yr. The present high input of nitrate has accelerated the migration rate of the redox interface. For input of both oxygen and nitrate Robertson et al. (1996) reported a vertical rate of 0.1 cm/yr, Bohlke et al. (2002) a vertical rate of 2.2 cm/yr and Postma et al. (1991) 1.8 cm/yr. Assuming a low pyrite concentration Wriedt and Rode (2006) simulated a significantly higher vertical rate of 10 cm/yr for input of both oxygen and nitrate in a till of Saalian age. The variations in migration rates are caused by differences in amount of redox capacity of the sediments, concentration of oxygen and nitrate and the vertical flux. Mass balance calculations indicate that the reserve of reduced compounds in young tills (Weichselian and Wisconsian age) under natural groundwater flow conditions, i.e. not forced groundwater flow due to abstraction or similar, is large enough to reduce nitrate for at least 100 years (Rodvang and Simpkins, 2001; Hansen et al., 2014b).

3.2. Studies in the Baltic Sea

3.2.1. Denmark

The part of Denmark draining to the Baltic Sea is dominated by young till sediments from the Weichsel glaciation. The clayey till sediments have a high content of reduced compounds dividing the subsurface into an upper predominantly oxidised and lower reduced part. Due to the relatively short exposure time of the sediments (compared to sediments of Saalian age or older) the redox interface are found at shallow depths within 1- 5 m from the ground surface in most of the area. The shallow redox interface together with a groundwater dominated hydrology leads to a large N-reduction in groundwater in Denmark.

Local scale studies

Several local studies exist from Denmark of which the best known is the Rabis Creek study by Postma et al. (1991). The studies have found high nitrate concentrations in the oxidized zone and very low concentration or concentrations below the detection limit in the reduced zone below the redox interface (Ernstsen and Morup, 1992; Ernstsen, 1996; Ernstsen et al., 1998a; Hansen et al., 2008; Pedersen et al., 1991; Postma et al., 1991). In the studies by Ernstsen (1996) and Ernstsen and Morup (1992) age dating of the groundwater by tritium showed that the lack of nitrate below the redox interface was not because the water had infiltrated before the intensification of agriculture and thus was nitrate free from start. Not many of the Danish studies have quantitatively assessed the denitrification rate. Jorgensen et al. (2009) conducted a lab experiment with sandy aquifer sediment from an agricultural site at Fladerne Creek and found a denitrification rate of 2-3 μmol NO_3^- kg^{-1} day^{-1}. Column experiment in the lab on clayey till columns from an agricultural site in Grundfør Jorgensen et al. (2004) found denitrification half-lifes ($T_{1/2}$) of 0.11 days (1.7-2.2 m depth) and 0.8 days (2.4-2.9 m depth).

Catchment scale studies

On catchment scale the amount of nitrate reduction in groundwater has been estimated using distributed hydrological models for Karup catchment (Styczen and Storm, 1993a, b; Refsgaard et al., 1999; Thorsen et al., 2001), Odense catchment (Refsgaard et al., 1999; Hansen et al., 2009; Hoang et al., 2014), Horsens Fjord catchment (Hinsby et al., 2012) and Norsminde Fjord catchment (Hansen et al., 2014a, b).

The most detailed catchment scale studies on nitrate reduction in groundwater in Denmark are carried out by Hansen et al. (2014a, b) for Norsminde Fjord catchment. In these studies maps of groundwater N-reduction was produced on 100 m scale using particle tracking and assuming instantaneous N-reduction below the redox interface. A spatially distributed redox interface was estimated based on the recharge flux and the redox capacity of the sediments (Hansen et al., 2014a), and the uncertainty on the nitrate reduction map due to geological uncertainty was estimated using stochastically generated geological models (Hansen et al., 2014b). Within a 37 km^2 study area groundwater reduction varied from 0 to 100 % and the standard deviation between the geological models was up to 40 %. The studies show that N-reduction in groundwater may be both highly variable and uncertainty at local scale.

National scale studies

National estimates of total N-reduction have been addressed in national projects, with the latest being the study by Højberg et al. (2015). They develop a national nitrogen model consisting of a statistical N-leaching model (NLES), a coupled surface water groundwater hydrological model setup in MIKE SHE on 500 m grid (Højberg et al., 2013) and statistical models describing N-retention in surface waters. Groundwater reduction is simulated by particle tracking and assuming instantaneous and complete reduction below the redox interface. The model is calibrated to stream data on N-transport for a 21 year long period, and is used to estimate surface water and groundwater reduction of nitrogen with a spatial resolution of approximately 1500 hectares, as shown in Figure 3 for groundwater reduction. The redox interface used in the model is developed from approximately 13.000 borehole data and manual interpretation based on soil types, landscape morphology and auxillary geological data (Figure S1).

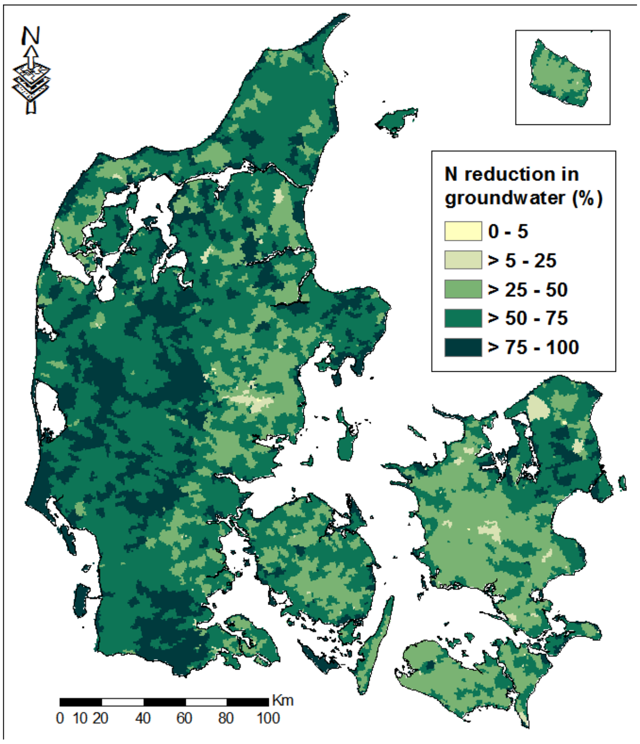

Figure 3 Groundwater N-reduction map for Denmark, modified from Højberg et al. (2015).

3.2.2. Sweden

Sweden is dominated by bedrock and relatively thin till soils from the Weichselian glaciation, with a relatively shallow groundwater table. Discharge in the till soil landscape is dominated by groundwater, with surface runoff generally only occurring during times of saturation, for example during major snow melt events. Agriculture is mainly concentrated to a few areas in Sweden with post glacial soils. Around half of the arable land is tile drained. Major groundwater aquifers utilised for water abstraction are found in the eskers in the central and southern part of the country.

Local scale studies

Few studies on nitrate reduction in groundwater have been carried out in Sweden. Maxe (2015) made an analysis of nitrate concentrations from sampling of Swedish wells and found correlations with land use, soil characteristics, well type and chemical characteristics of the groundwater. Concentrations of nitrate in the wells were negatively correlated with distance from agricultural fields. Soil clay content on agricultural soils also influenced the nitrate concentration, with a higher likelihood to find high nitrate concentrations in soils with low clay content. The study also looked at correlation with the redox status of the groundwater determined by the concentrations of iron, mangan and sulphate in the samples from the wells. Elevated nitrate concentrations are mainly found in water with high redox potential. Of the samples with low redox potential most are found in wells in the south-west of Skåne, the southern-most county of Sweden. However, this study does not allow for detailed mapping of nitrate reduction in Sweden. Arheimer and Brandt (1998) found in a modelling study that much of the nitrogen retention in southern Sweden takes place in lakes and main rivers of the different sub basins, but that locally retention in groundwater and small streams/ditches, contributes significantly in many areas.

National scale studies

While N transport at national scale has been estimated by modelling studies, no studies have assessed the groundwater reduction and its impact on the total N-load to the Baltic Sea. In the present study, the groundwater reduction map for Sweden was developed on the basis of two model systems; the S-HYPE and

SMED-HYPE. S-HYPE (Strömqvist et al. 2012) is the national application of the HYPE model for Sweden. HYPE (Lindström et al. 2010) is a process based semi distributed model with integrated calculations of hydrology and nutrients in soils, groundwater and surface waters. In a typical HYPE application the landscape is divided into sub basins, each with different distributions of hydrological response units (HRUs) comprising unique soil and land use combinations. Each HRU is simulated as a soil profile with up to three layers of which the top two layers constitute the root zone. Simulated runoff and leaching of nutrients from the different soil layers (and tile drains) are routed through the hydrological network of local rivers, main rivers and lakes.

The SMED-HYPE modelling system was developed for periodic reporting of nutrient loads from Sweden to the Baltic Sea for the Baltic Marine Environment Protection Commission (HELCOM). The SMED-HYPE system works in a similar way to the S-HYPE but instead of using the HYPE model´s own root zone leaching it uses root zone nitrogen leaching values from the SOIL-N model (Bergström et al. 1991) for agricultural areas and standard leaching values for other land uses. Additionally, N-transport and reduction in the groundwater system and small streams/ditches, i.e. transport and reduction between the root zone and major streams, are described by a lumped process in SMED-HYPE, while the processes are described separately in S-HYPE, Figure S2.

Due to the different model structures, groundwater reduction was calculated in different ways. For S-HYPE the retention was calculated as the quota between the reduction of inorganic nitrogen in the ground water zone and the sum of the root zone leaching and losses through direct runoff to the stream network from the upper soil, including tile drain flow. For SMED-HYPE, which does not separate between retention in groundwater and in local streams and ditches, retention was calculated by comparing the gross load from root zone leaching and the load to the main river in each sub basin. The two modelling systems are assumed to be equally good and the final retention map is hence calculated as the mean of the retention estimated by the two models, Figure 4.

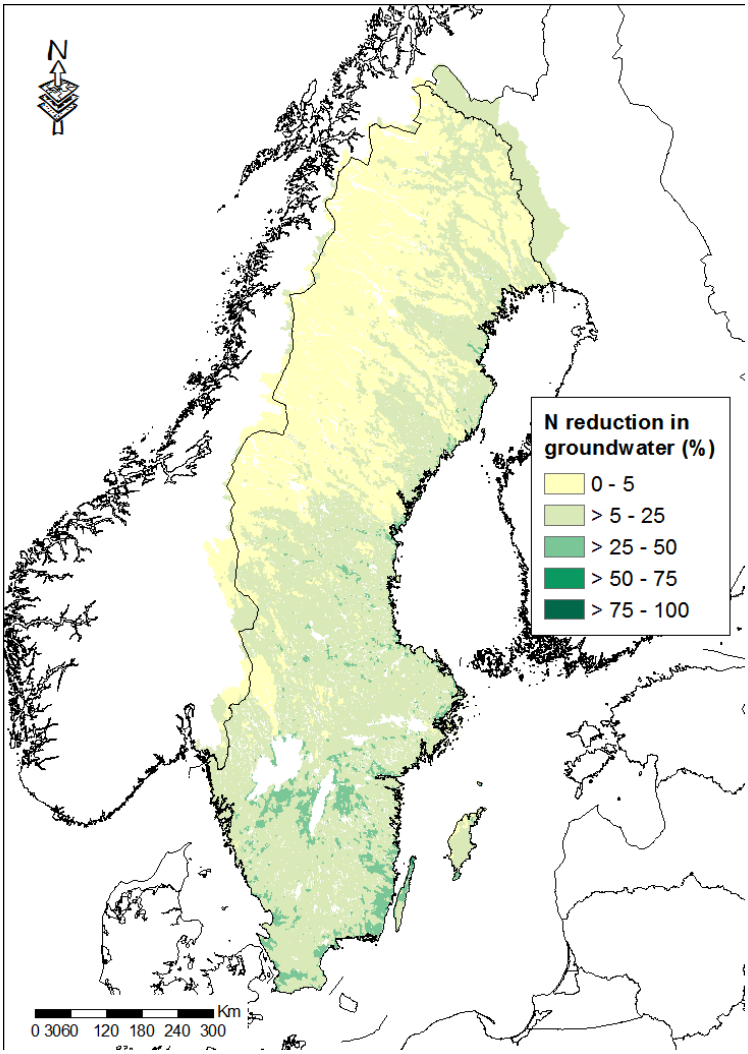

Figure 4. Groundwater N-reduction map for Sweden.

3.2.3. Finland

The soils in Finland consist mainly of till and sediments from the Weichselian glaciation as well as post-glacial sedimentary soils and peat (Figure S3). The depth of soils is variable and bedrock can be uncovered or located at 100 m depth, but the mean thickness is only 8.5 meters. The land area in Finland increases continuously due to isostatic land uplift, adding 50,000 km^2 to the land area since the Litorina stage. Recent studies (Beucher et al., 2015) have estimated that 5-12% of the Litorina soils in Finland may contain acidic sulphate (AS) soils and up to 1,300 km^2 of the AS soils are cultivated. Even though AS soils contain a substantial amount of sulphides (up to 2% w/w) the nitrate leaching is high from these areas.

Water saturation of the entire soil profile is common during snow melting in spring or autumn rainfalls but during summer and winter the groundwater drops deeper. Drainage is a prerequisite for cultivation in Finland. Some fields still have open ditch drainage, but 75–88% of the fields are subsurface drained in the southern and south-western parts of Finland, where 50% of the cultivated fields consist of clay soils. In cultivated subsurface drained fields the groundwater may drop up to 2 meter below the drains in summer (Äijö et al. 2014; Österholm et al. 2015) exposing subsoils for atmospheric oxygen and converting redox conditions from reduced to oxidized. The mean depth of redox interface in cultivated fields determined by redox potential measurements was found to be 1.5 meter (Puustinen et al, 1994), Figure S4. In AS soils the redox interface was closer to soil surface in clay soils than in coarser soils (Virtanen et al. 2016) which contain less sulfides than finer-textured AS soils.

Local scale studies

Forest and peat land are the dominant land use types in Finland and therefore the largest volume of water discharges from these areas to the Baltic Sea, generally with very low nitrate concentrations. In forests, nitrate loads originate mainly from organic nitrogen and atmospheric deposition, but forestry management practices increase nitrate leaching to surface waters (Piirainen et al. 2007) and also nitrate concentrations in groundwater (Kubin, 1998).

From agricultural fields, the leaching of nitrogen (mainly in the form of nitrate) have been studied in long-term experiments (e.g. Seuna and Kauppi, 1981; Turtola and Paajanen, 1995; Äijö et al. 2014). Results have been analysed by balance calculation (e.g. Salo and Turtola, 2006; Äijö et al., 2014) as well as by modelling (e.g. Karvonen et al., 1999; Knisel and Turtola, 2000; Rankinen et. al 2007; Salo et al. 2015). Only few modelling studies consider nitrification or denitrification during water flow from soil surface to drain pipes or transport in groundwater separately because the N-load to rivers from groundwater and denitrification in the subsurface have been considered low (Seuna and Kauppi, 1981). However, in an AS soil field, the lack of nitrate below drainage depth was attributed to denitrification (Simek et al. 2011).

Catchment scale studies

In forest catchments, hydrological processes and the major processes controlling the N losses including denitrification have been modelled (e.g. FEMMA model, Laurén et al., 2004), but the denitrification was found to be minuscule. In agricultural areas, catchment scale nitrogen leaching have been estimated using data from a network of small hydrological basins and national monitoring data from river basins (e.g. Rekolainen, 1993; Tattari et al. 2015), by nitrogen balance of fields (e.g. Salo and Turtola, 2006) and by modelling (Puustinen et al. 2010; Granlund et al., 2007; Rankinen et al. 2016). However, denitrification in groundwater has not been quantitatively assessed at catchment scale. In a state-of-the-art study Randall et al. (2014) noticed that the lowest measured nitrogen concentrations were below the lowest modelled concentrations, suggesting that some reduction processes may be missing in the model.

National scale studies

From mass balance studies, including the majority of catchments in Finland, Myllys (1992) estimated that approximately half of the N surplus from agricultural fields is lost to the surface water system, and the rest is removed by denitrification. Denitrification in groundwater was, however, not separated from denitrification occurring in root and vadose zone in their study. In well-structured heavy soils Paasonen-Kivekäs et al. (1999) found that nitrate concentration in surface water was nearly equal to that in the drain water, while the nitrate concentration in groundwater was only 2% to 16% of the surface water concentration. This implies that drainage is the major transport pathway for nitrate from agricultural fields to surface water at the study site. The low nitrate concentrations in the groundwater indicate that nitrate reduction may occur below the drains but due to the very low hydraulic conductivity of the clayey subsoils, the vertical flux below the drainage system is very limited. Defining groundwater reduction as the difference between the root zone leaching and loads to the surface water, the total groundwater reduction for such well-drained soils is low.

From the past studies, reduction of N in groundwater appears to be very limited in Finland. This may be related to the fact that the highest N losses occur during autumns and springs in occasions which are reported to take only a few days. The rapid outflow and low temperature limits microbial activity and thus the denitrification or other N reducing processes. In summer, when temperature is higher, elevated groundwater might result in nitrate reduction by denitrification and decrease nitrogen losses from soil to sea.

Retention for all catchments in Finland has previously been estimated by Lepistö et al. (2006) and Huttunen et al. (2016) as a total catchment scale retention, i.e. with no differentiation in surface water and groundwater. At a more detailed scale Rankinen et al. (2016) have estimated the retention using the MESAW model for 20 catchments. None of the previous studies have, however, assessed the N-reduction in groundwater. In the present study, results from Rankinen et al. (2016) was therefore analysed and the total catchment retention estimated for the 20 catchments was divided into surface water retention and groundwater reduction, respectively. For each catchment a relation between the estimated groundwater reduction and the areal fraction of eskers in the catchment, was established. Like in Sweden, the dominant groundwater aquifers are associated to the Eskers, which have thus been used as a proxy for the size of the groundwater aquifers. The relation between the area of the eskers and the groundwater reduction was used to divide the total catchment scale retention by Lepistö et al. (2006) and Huttunen et al. (2016) into surface retention and groundwater reduction for the remaining part of the country not covered by the 20 catchments. The national N-reduction map for Finland is provided in Figure 5.

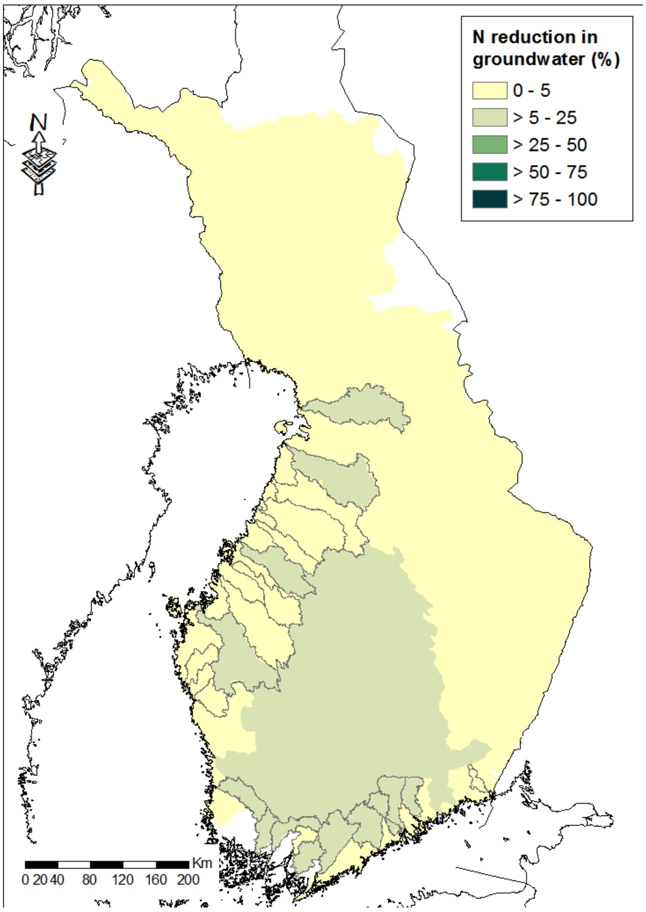

Figure 5 Groundwater N-reduction map for Finland. Catchments subject to detailed studies by Rankinen et al. (2016) and used to extrapolate groundwater reduction to the entire Finland is indicated by black outline.

3.2.4. Lithuania

Lithuanian is dominated by sandy and clayey Quaternary sedimentary cover of glacial origin, whose thickness varies from 10 to 100–200 m. Pre-Quartenary sandy, clayey and carbonate sediments are found below the Quarternary and the thickness of the active groundwater exchange zone is 200–300 m (Grigelis et al., 1994). Groundwater chemistry is controlled by the petrographical and mineralogical composition of aquifer and aquitard deposits, as well as the connection to surface water and atmosphere (i.e., their confinement degree). One or two aquifers can typically be found in a vertical section through the Quaternary deposits, but in some places up to five or six exists, where intermediate confined groundwater is the transitional type of groundwater between unconfined and deep confined groundwater. They are usually hydraulically interconnected and may have good hydraulic contact with both underlying artesian water and overlying unconfined groundwater. Deep, confined water can be found in deposits of all geological ages and is one of the main sources for large, public water supplies in Lithuania. In recharge areas (topographical highs) deep confined groundwater is recharged from intermediate confined and unconfined groundwater.

Local scale studies

The lateral and vertical distribution of nitrogen and the distribution of oxidised and reduced nitrogen species in the groundwater were studied by Klimas and Paukstys (1993). In aquifers with high oxygen concentrations, nitrogen is mostly found as nitrate, while ammonia concentrations are low. Anoxic conditions often occur in confined aquifers, where the redox potential may be controlled by iron and nitrogen predominantly is found as ammonia. It appears that the low nitrate and elevated ammonium contents typically found in deep confined groundwater are the result of the chemical reduction of nitrate to ammonium in aquitards below unconfined aquifers (Klimas and Paukstys 1993).

Depth to the redox interface has been studied based on descriptions of change in till sediment colour in more than 1000 wells, where distinction was made between; highlands where the regional recharge is high, river valleys which are predominantly discharge zones; and transit zone between the highlands and river valleys (Klimas, 1996). The results is shown in Table 1, where wells have been divided into three categories depending on the colour changes, where "End of brown/red zone" indicate the transition from oxidized to reduced condition, "Start of grey zone" indicate a more reducing environment; and "start of blackish/greenish zone" the most reducing environment. A single or several of the colours may occur within a well. The data confirms the expectations of a deeper redox interface in groundwater recharge areas, where the flux of oxygenated water and nitrate is large, while the interface is found closer to land surface in regional groundwater discharge zones.

Table 1. Depth below surface to reducing environment determined from changes in sediment colours

Hydrogeological zone	Mean depth to change in sediment colour (in m below surface)		
	End brown/ red zone	Start gray zone	Start blackish/greenish zone
Recharge	12	36	51
Transit	18	19	34
Discharge	11	12	16

An extended study of nitrogen in groundwater (Status, 2011) revealed, that only a small part of nitrogen, applied as fertilisers on land surface, infiltrates to groundwater (1 – 20%). Highest concentration of nitrates in groundwater is found in river basins, where agricultural activities are most developed (Nevėžis and Šešupė), but the total nitrate loads to surface water bodies are highest in river basins, where recharge to groundwater is highest (Neris, Merkys, Šventoji). Typically, such river basins are located in highlands, dominated by sandy deposits. The nitrate load to rivers in lowlands covered with clayey deposits is far less, which may be attributed to higher reduction potential in clayey deposits.

National scale study

A national assessment of N-reduction in groundwater has not been carried out previously for Lithuania. The map developed in the present study is compiled by combining a national map of the Quaternary geology with lithology and genesis of the uppermost deposits, Figure S5, (Guobytė R., 1998) and a map of the shallow groundwater recharge, Figure S6, (Putys P., 2013). Based on previous studies and groundwater monitoring data it is assumed that sandy deposits with high infiltration rate have a relatively low nitrate reduction potential, clayey deposits with medium and low infiltration rate have medium to high reduction, and peatland is associated with very high nitrate reduction potential. Nitrate reduction has not been the primary focus in previous projects and the present nitrate reduction map for groundwater (Figure 6), based on existing data and knowledge and a simple index approach, is thus the first attempt.

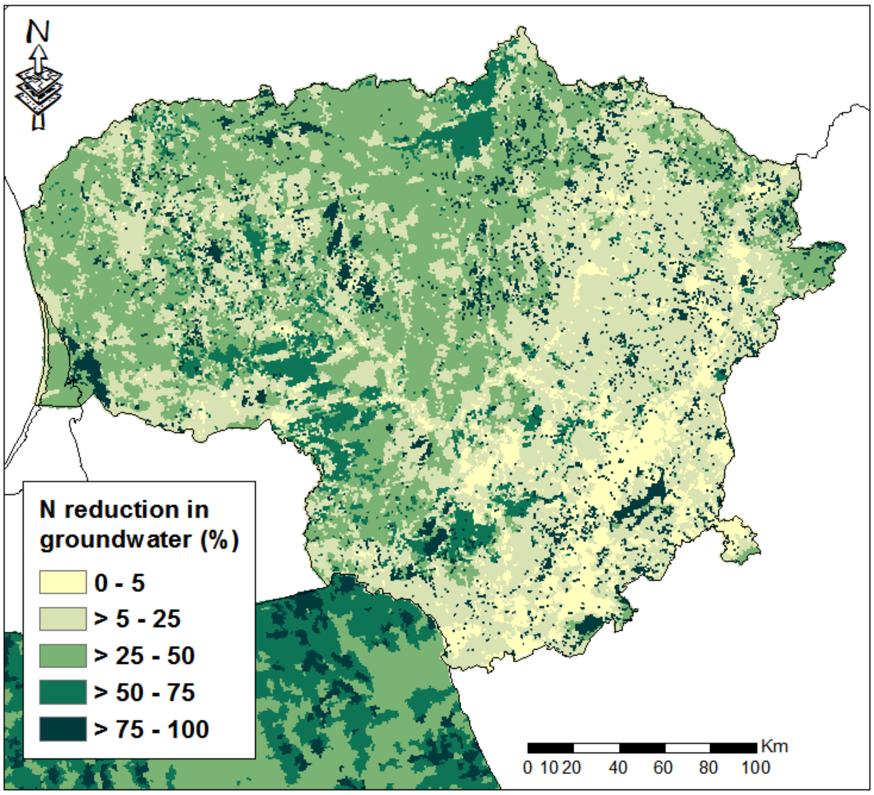

Figure 6 Groundwater N-reduction map for Lithuania.

3.2.5. Poland

In the Polish national monitoring programme nitrate was found to exceed the quality limit of 50 mg NO3/L in only 5.1% of the monitoring points for the period 2008-2011 (Rojek et al., 2013). The national monitoring programme is, however, affected by a low density and uneven distribution of monitoring points for shallow groundwater and by the inadequacy of sampling protocols (Alterra, 2007). Consequently, results of the national monitoring network tend to underestimate nitrate levels and many local studies reveal nitrate pollution, especially in shallow groundwater in agricultural areas (e.g. Płochniewski and Macioszczyk, 1983; Błaszyk and Górski, 1989; Żurek, 1991; Mikołajków, 1995; Kaźmierczak-Wijura, 1996; Malina et al., 2007; Ćwiertniewska et al., 2008; Michalczyk et al., 2016).

Local to catchment scale studies

Published studies on nitrate reduction in groundwater do not provide a scientific basis for relating nitrate reduction potential directly to geological and hydrogeological conditions in different parts of Poland. Nevertheless, groundwater nitrate reduction is generally controlled by hydrogeological conditions, particularly the redox potential, and higher nitrate concentrations are associated with lighter, more permeable soils, while the lowest concentrations occur under clayey soils. Hydrological conditions in Poland show a distinct bimodal distribution dividing Poland into two hydrogeological provinces, Figure S7. The Lowland Hydrogeological Province (LHP) is associated with Quaternary porous aquifers developed mostly in glacial deposits with diverse nitrate reduction potentials. The Mountain-Upland Province (MUHP) prevailing in the south of Poland is associated with fissured, fissured-porous or fissured-karstic aquifers, usually covered with thin Quaternary deposits, inter alia, loess with low denitrification potential (Kleczkowski, 1991; Kleczkowski et al, 1990; Kleczkowski and Witczak, 1990).

Quaternary aquifers of the LHP, particularly in the valley, ice-marginal valley and intermorenic aquifers, show low redox potentials favouring denitrification (Żurek, 2002), which is often confirmed by high concentrations of iron and manganese (Witczak et al., 2013). In the polluted aquifers nitrate concentrations distinctly decrease with depth (Dragon et al, 2016). Górski (1989) found a direct relation between the thickness of glacial tills and nitrate concentrations, with 20 m thick till covers providing protection of groundwater from nitrate pollution.

The hydrogeological conditions in the MUHP do not support denitrification in groundwater (Żurek et al., 2010a; Żurek and Mochalski, 2010) as exemplified by observations of high nitrate concentrations in the fissured-karstic (Kryza J. and Kryza H., 2001; Dąbrowska et al., 2005; Malina et al., 2007; Żurek et al., 2010a, 2010b; Śledzik, 2014) and fissured - porous (Żurek, 2007) aquifers.

National scale studies

There have been no previous attempts to develop a national map for N-reduction in groundwater for Poland, and in the present study the map was developed by combining existing data at national scale in three steps:

Step 1 - Identification of major hydrogeological units.

Delineation of the two hydrogeological provinces: Lowland Hydrogeological Province (LHP) (represented by green colour in Figure 7) and Mountain-Upland Province (MUHP) prevailing in the south of Poland (red colour in Figure 7).

Figure 7 Hydrogeological Provinces in Poland: Lowland Hydrogeological Province (LHP) (green colour) and Mountain-Upland Hydrogeological Province (MUHP) (red colour) (after Kleczkowski, 1990)

Step 2 – identification of dominating lithology

Nitrate reduction in groundwater is assumed to be proportional to the residence time and the redox potential of the geological formation (lithological type), where the redox potential in till generally is larger in clay compared to sand (Ernstsen, 2013). The classification of lithology from the Groundwater Vulnerability Map of Poland (GVMP) (Witczak et al. 2007; 2011) was applied (Table 2, Figure S8). This classification generally reflects the flow condition in saturated zone and hence the nitrate reduction capacity, with one exception. The semi- and low permeable porous sediments (class 7, Table 2) comprise generally clayey sediments, but also loesses not containing clay particles, which form the protective cover over aquifers in the MUHP. Due to the low hydraulic conductivity in loess, the travel time through the unsaturated zone is high, but the nitrate reduction capacity is low due to oxidizing conditions. The contribution of the semi- and low permeable sediments (class 7) in the unsaturated zone profile that constitute the aquifer cover (C_{lp}) (cf.

Figure S8 and explanations in Table 2) was taken into consideration as a separate layer included in the GVMP (Witczak et al. 2007; 2011; Wachniew et al. 2016).

Table 2 Classification of typical lithologies of unsaturated and saturated zone (under the root zone) in Poland (from Witczak et al., 2007, 2011) with suggested nitrate reduction.

Class number	Groundwater environment	Typical lithologies	Nitrate reduction [%]
1	Permeable fissured-karst	limestones, dolomites	<10; 20[1]; 25[2]
2	Permeable fissured	granites, metamorphic rocks	<10; 20[3]; 30[4]
3	Permeable fissured-porous	sandstones, flysh rocks	<10; 15[5]
4	Permeable porous-fissured	chalk marls, opokas, chalk	15; 20[6]; 25[7]
5	Permeable porous	gravels and sands	60[8]; 50[9]
6	Permeable porous	silty sands, loamy sands, etc.	70
7	Semi- and low permeable porous (K $<10^{-6}$ m/s)	loess, glacial till, silt, loam, etc.	80[10]

Explanations: [1,2] - for fissured-karst rocks covered with sediments of class: [1] no.5; [2] no.6 and 7 (only loess); [3,4] - for fissured rocks covered with sediments of class: [3] no.5; [4] no.6; [5] - for flysch rocks with dominated shales; [6,7] - for porous-fissured rocks covered with sediments of class: [6] no.6; [7] no.7 (only loess); [8] – contribution of semi- and low permeable materials in the cover (C_{lp}) equal or higher 0.4 (Figure S8 – right panel); [9] – contribution of semi- and low permeable materials in the cover (C_{lp}) equal or lower 0.3 (Figure S8 – right panel); [10]– does not apply to loess (see text).

Step 3 – influence of artificial drainage

Assessment of the influence of drainage on nitrate reduction is not straightforward. The density of drainage network is not directly related to the soil conditions but also to the individual farmer's economy and therefore differs significantly between regions ranging from 1% in Lubuskie to 32% in Wielkopolskie (Figure 8). The installation of the drainage networks occurred mostly before 1990. Since then the density of the functional drains have not increased, but rather decreased due to lack of maintenance. Loadings of nutrients in drainage networks were estimated in several studies (Lipiński, 2002; Fic et al., 2003; Durkowski and Lipiński, 2010).

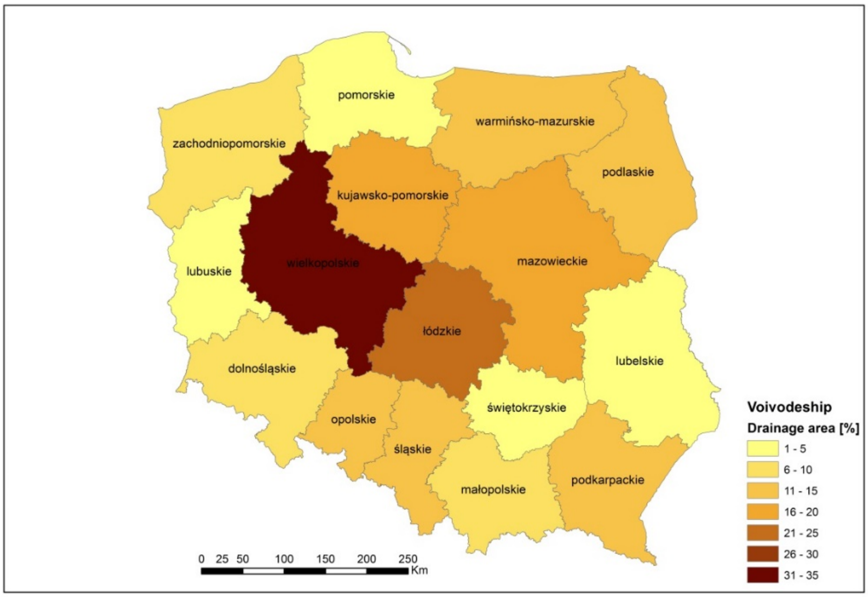

Figure 8. Fractions of the drained areas in the agricultural land in all voivodships (provinces) based on data from the Ministry of Agriculture and the Central Statistical Office (data from 2010).

Only in two voivodships (provinces), Wielkopolskie and Łódzkie, more than 20% of the agricultural land is drained. In the Wielkopolskie Voivodship 32% of the agricultural land is drained. A similar percent of the area is covered by fine grained soils (clays, silty clays, clay loams) as identified in one of the information layers of the GVMP (Witczak i in. 2007; 2011). This correspondence indicates that in this voivodship all fine grained soils in the area have been drained. No relationship between the area of the drained land and soil types was found for Łódzkie Voivodship. The influence of drainage was considered only for Wielkopolskie and for the other voivodships drainage was assumed to have no significant influence on nitrate routing. Due to its similarity in geology and drainage between Wielkopolskie and the drained clayey soils in Denmark, a nitrate reduction of 40% was adopted from the Danish study. The final map for estimated N-reduction in Poland is shown in Figure 9

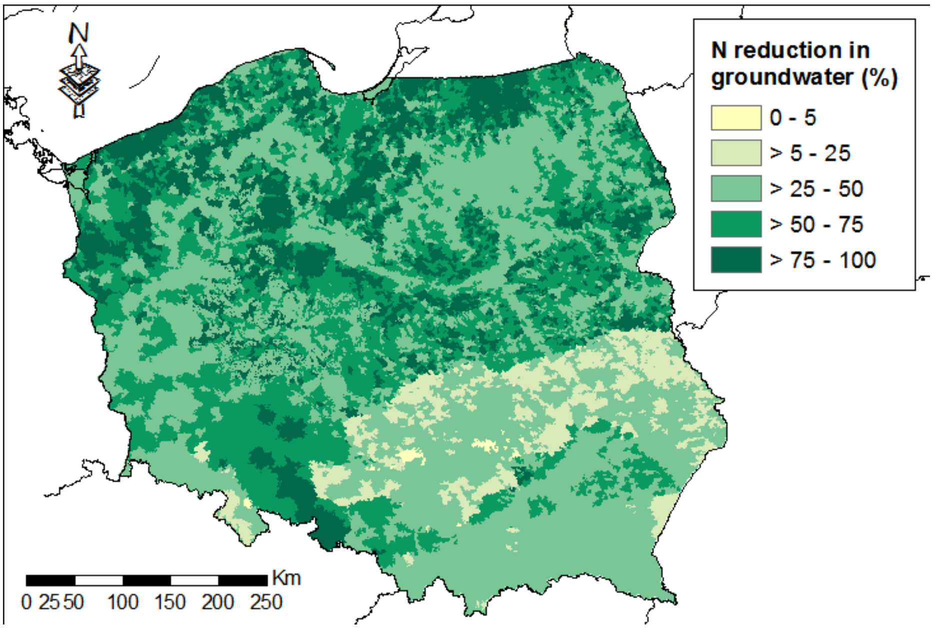

Figure 9. Groundwater N-reduction map for Poland in percentage.

3.2.6. Germany

The two northernmost federal states in Germany; Schleswig-Holstein and Mecklenburg-Vorpommern discharge to the Baltic Sea. This part of Germany was covered by ice during the last glaciation (Weichsel). The recent geological history and depositions is thus comparable to the part of Denmark discharging to the Baltic Sea. The importance of geology on local scale N-reduction in the groundwater is therefore expected to be similar to the observations in Denmark.

Catchment and national scale

Nitrogen transport and reduction has been assessed for Mecklenburg-Vorpommern by Wendland et al. (2015). They utilised a model complex described by Wendland et al. (2004) consisting of a nutrient balance mode, a water balance model (GROWA), a reactive nitrate transport model in soil (DENUZ) and a reactive nitrate transport model in groundwater (WEKU). In the model complex the nitrate reduction is assumed to occur by first-order decay, where rate constants are estimated from groundwater analyses. High and low decay rates are associated to reduced and oxidised groundwater, respectively, while intermediate rates are used for aquifers that cannot be unambiguously categorized as aerobe or anaerobe. Actual reduction in the groundwater is calculated from the decay rates and resident times. Assuming two-dimensional groundwater flow parallel to the groundwater table, residence times are estimated in a GIS approach from groundwater gradient and expected hydraulic conductivity and porosity. Spatial variation in N-reduction in groundwater is thus determined by a combination of the geochemical conditions (assigned from monitoring data) and estimated residence times.

Similar to the Danish conditions, drainage was found to be an important pathway transporting approximately 35 % of the N-leaching from the fields to the surface water system. For Mecklenburg-Vorpommern Wendland et al. (2015) estimated that 54% of the nitrogen leaving the root zone is reduced before it reaches the surface water system. With groundwater reduction being proportional to the residence time, which is determined by the proximity to surface waters, the estimated N-reduction in groundwater is expected to vary at a fine scale. However, since this was not the purposes of the study this variation is not reported and a single N-reduction of 54% is assigned for N-reduction in groundwater for the Mecklenburg-Vorpommern as well as Schleswig-Holstein.

3.2.7. Baltic Sea Basin scale

The first model study including the entire Baltic Sea Basin in one uniform model setup was made by Mörth et al. (2007) using a the lumped hydrological model CSIM. In the model a fixed percentage of nitrogen in manure was assumed retained in the soils, thus no spatial variation was considered, nor was it considered whether retention occurred in the root zone or below.

Wulff et al. (2014) recognise the importance of considering nitrogen retention in the design of cost-effective mitigation measures, as retention between the source and the outlet will impact the effectiveness of the measures. They define the catchment-scale retention as the difference between anthropogenic nutrient inputs and observed riverine export, which conceptually can be sub-divided into retention in soils, groundwater and surface water. Building on the results of the BONUS project RECOCA, Wulff et al. (2014) present a multi-model approach to describe nutrient transport and retention between the source and the sea, which can be used to evaluate various management options. Retention in the surface water system is calculated by the statistical model MESAW as reported by Stålnacke et al. (2015). The model simultaneously estimates catchment N-loads, using N-export coefficients for different land uses classes, and surface water retention. Model performance is evaluated for 88 measured rivers in the Baltic Sea Basin and the total surface water retention for rivers draining to the Baltic Sea is estimated to be approximately 40%, with the largest retention occurring in catchments with a high areal fraction of lakes. Using the soil-vegetation-atmosphere model DAISY and a data resolution of 10 x 10km Andersen et al. (2016) develop a regression model for N-leaching for dominant combinations of climate, soils and agricultural management. The regression model is used to estimate N-leaching from the root-zone for the entire Baltic Sea Basin at a resolution of 10 x 10 km, i.e. much finer resolution than the catchment scale assessed by Stålnacke et al. (2015), from which N-loss hot-spots areas are identified.

Andersen et al. (2016) construct a mass balance for riverine N-load at the catchment scale. Utilising average riverine N load for the period 1994-2006 from the HELCOM pollution compilation PLC-5, together with the estimated N-leaching from their regression model and surface water retention calculated by Stålnacke et al. (2015), the mass balance is solved for groundwater reduction. Hence the groundwater retention is not estimated independently, but is the residual given the N-leaching, surface water retention and long-term riverine N-load is known.

4. Results and discussion

The national maps for N-reduction in groundwater are compiled to a single map in Figure 10, which for comparison also includes the map by Andersen et al. (2016). For Finland the estimates in the present study is similarly based on MESAW at the same scale. In the remaining countries included in this study the reduction is estimated at a finer scale. For Denmark, Sweden and Germany (Mecklenburg-Vorpommern) the estimates are based on the use of models that are conceptually very different but all includes a spatially description to resolve heterogeneity in land use, soil properties and, except for Sweden, subsurface geology at sub-catchment scale. The estimates for Germany were not accessible at the fine resolution and hence only the aggregated value is included in the final map. The estimates for Poland and Lithuania are based on overlay of relevant geological and hydrological maps, and the final resolution in these countries reflects the resolution of the maps used.

Comparing the two maps in Figure 10 reveals obvious differences with respect to both the magnitude of groundwater reduction and the pattern of high/low values. In all assessments based on a modelling approach, observed riverine N-loads have been utilised to constrain and evaluate model performance. Hence the estimated input and retention/reduction in the catchment equals the observations, within a certain model performance. However, N-input (dominated by root zone leaching), retention and riverine N-loads are interlinked. If leaching is estimated too high it must be compensated for by a too high retention. Differences in magnitudes, and to some extent also the pattern, between the two maps may thus partly be ascribed the different approaches used to estimate the N-leaching in the different studies. Retention in surface water and groundwater is similarly interlinked, as the total catchment scale retention is the combination of the two. An overestimation of the surface water retention has to be counteracted by a groundwater reduction that is too low. This was observed by Højberg et al. (2015), who noticed that the model used to describe retention in lakes might overestimate the retention, where multiple lakes are connected, which would result in an underestimation of the groundwater reduction. A similar situation may occur in Andersen et al (2016), where groundwater reduction is computed as the residual in the catchment scale mass balance for riverine N-load, given that the other variables are known.

In Poland and Lithuania the groundwater retention is estimated based on geological and hydrogeological data combined with monitoring data and knowledge from previous national studies. Assumptions made with respect to variations in reduction potentials for different geological formations were based on studies described in section 3.1 and the studies carried out in the individual countries. The estimates for Poland and Lithuania thus draws on knowledge obtained from other local and catchment scale studies, and using national data a best estimate of the N-reduction in groundwater and its spatial distribution has been developed. Since no model study was involved, it has, however, not been possible to test the magnitude of the estimated N-reduction by combining estimated leaching, reduction and observed riverine loads.

The two groundwater reduction maps developed by Andersen et al. (2016) and in the present study represent two independent estimates. It is not possible to measure the N reduction in the groundwater directly, and it is thus not possible to validate either of the maps directly and provide evidence on which of the maps is most correct. In the present study the maps for Denmark and Germany have been developed by considering the subsurface hydrology in determining the transport and reduction of nitrate in the groundwater. For Denmark the difference between the two maps is distinct in terms of both magnitude and pattern. In the work by Højberg et al. (2015), a consistent approach was used for the entire country utilising a three dimensional hydrogeological model to describe groundwater flow (Højberg et al., 2013) and data from approximately 340 discharge stations and is thus better constrained to data than the approach by Andersen et al. (2016) including only few catchments and only surface water data for Denmark. For Finland the two studies utilises the same model (MESAW) to estimate surface retention at identical catchment scale. However, the estimated groundwater reduction in the coastal catchments is very different, with the estimate of the present study being much lower, which appears to be in better agreement with the local and catchment studies in the country.

The details in the maps from Poland, Sweden and Lithuania could not be resolve by Andersen et al. (2016) and it is difficult to evaluate how different the estimates are with respect to the total reduction at the catchment scale. The present estimate does, however, tend to estimate lower N reduction in groundwater for several of the Swedish catchments, and the bimodal distribution in Poland is obviously only identified in the present study due to the finer resolution.

Both studies indicate that the spatial variation in nitrate reduction in groundwater can be significant in most countries. This signifies that in many areas there is a potential for utilising the natural N reduction in future regulations, by focusing the mitigation measures to areas, where the reduction is low. Utilising more detailed data and drawing on local and regional studies we consider the map developed in the present study, in general, to be an improvement compared to the estimate by Andersen et al. (2015).

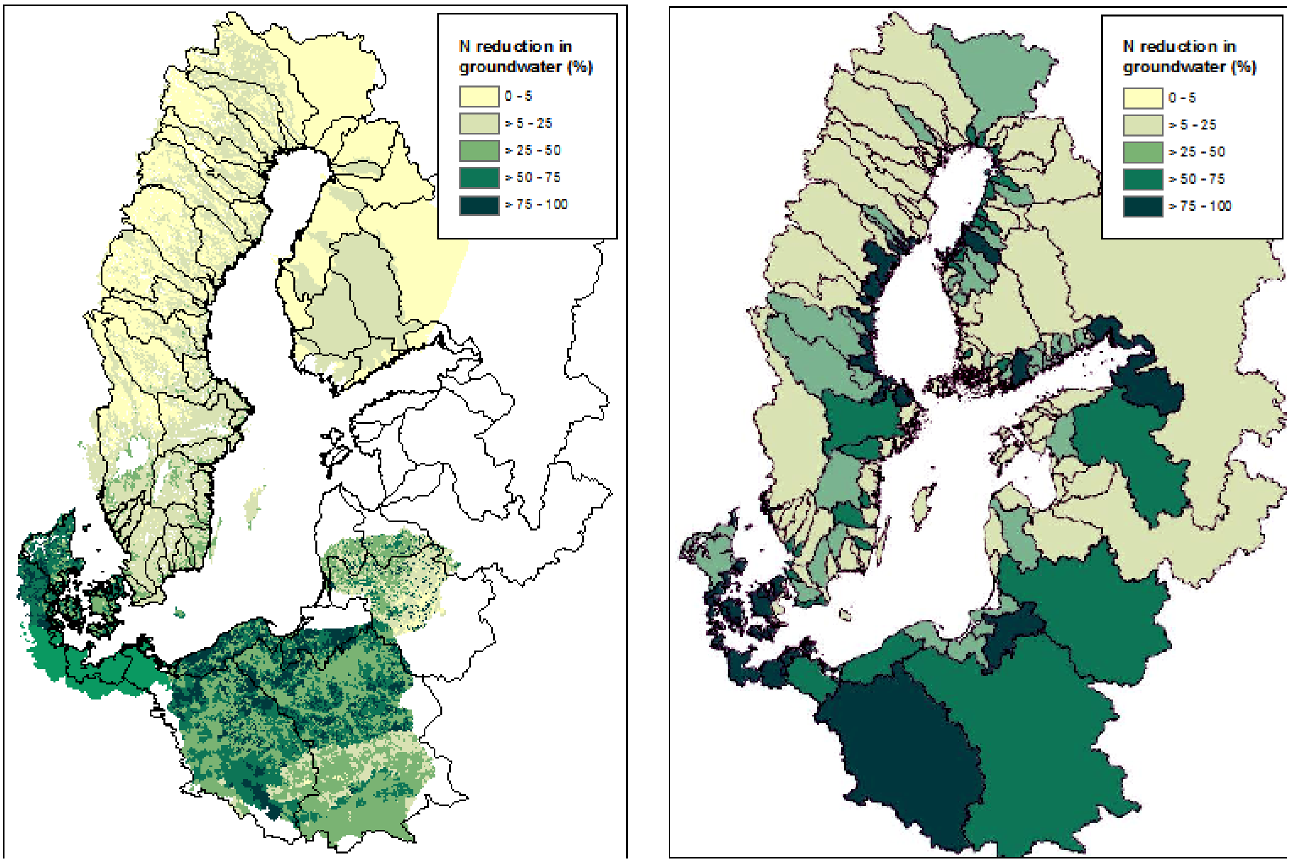

Figure 10.Nitrate reduction in groundwater as estimated a) in the present study and 2) by Andersen et al. (2016). Note that the interval 0-5 was not included in the assessment by Andersen et al. (2016)

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Supplementary Material

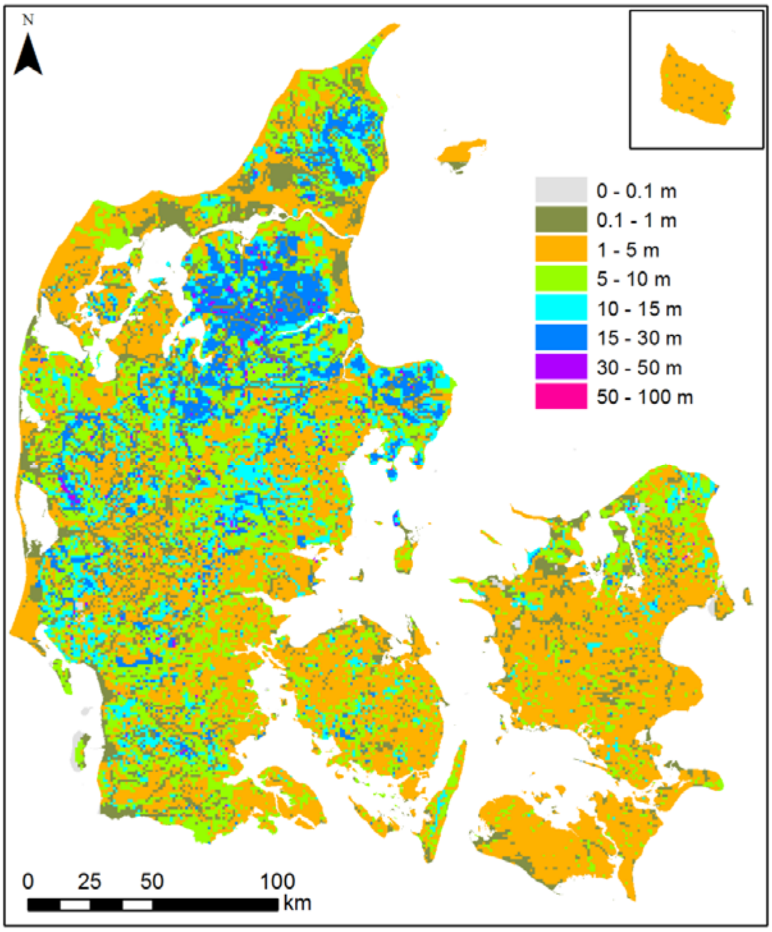
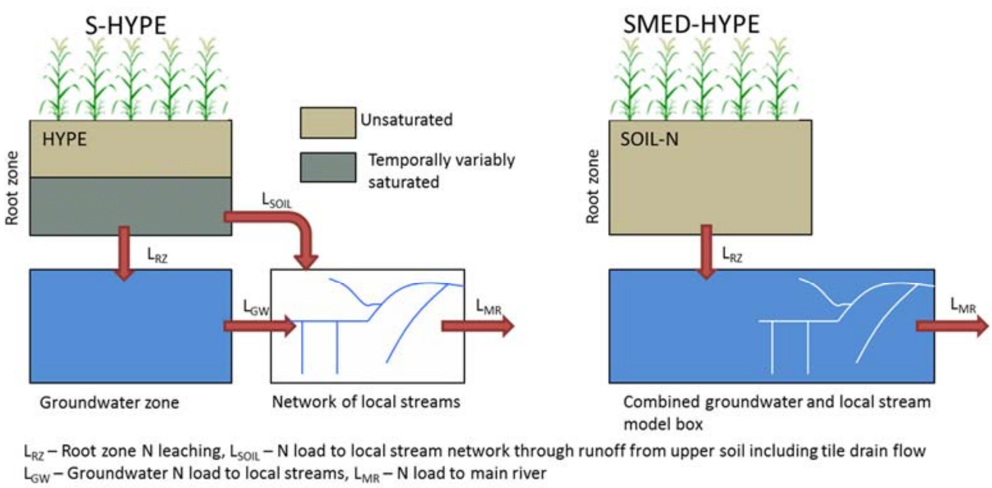


Figure S1. Depth to redox interface in Denmark. Modified from Ernstsen et al. (2006).



$$Ret_{S-HYPE} = \frac{L_{RZ} - L_{GW}}{L_{RZ} + L_{SOIL}} \qquad Ret_{SMED-HYPE} = \frac{L_{RZ} - L_{MR}}{L_{RZ}}$$

Figure S2. Conceptual figure of the nutrient flows and retention calculations in S-HYPE and in SMED-HYPE.

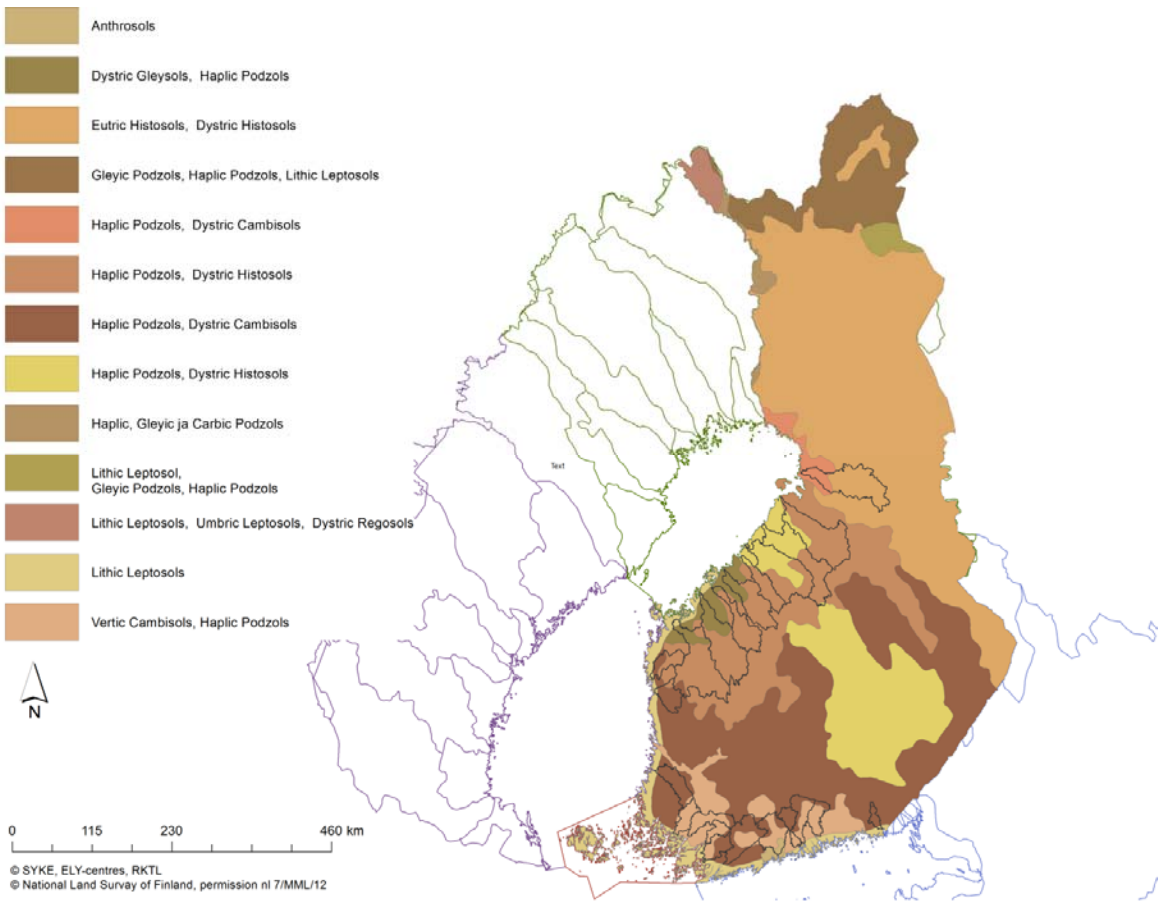


Figure S3. Main soil types in Finland. Outlines indicate the location of the 20 catchments used to develop a method to extrapolate the N-reduction in groundwater to the entire Finland

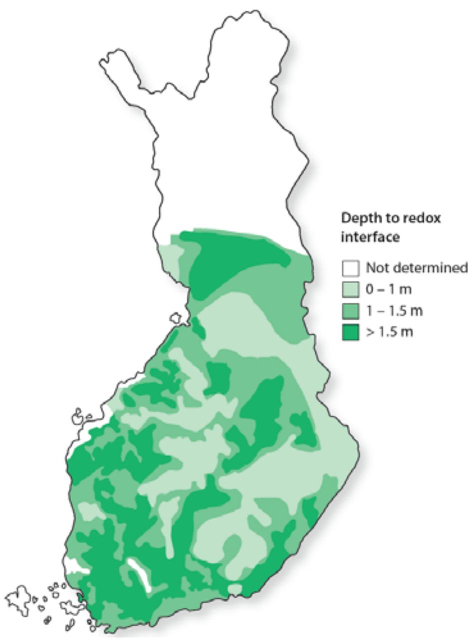


Figure S4. Depth of redox interface in cultivated fields in Finland (Puustinen et al. 1994)

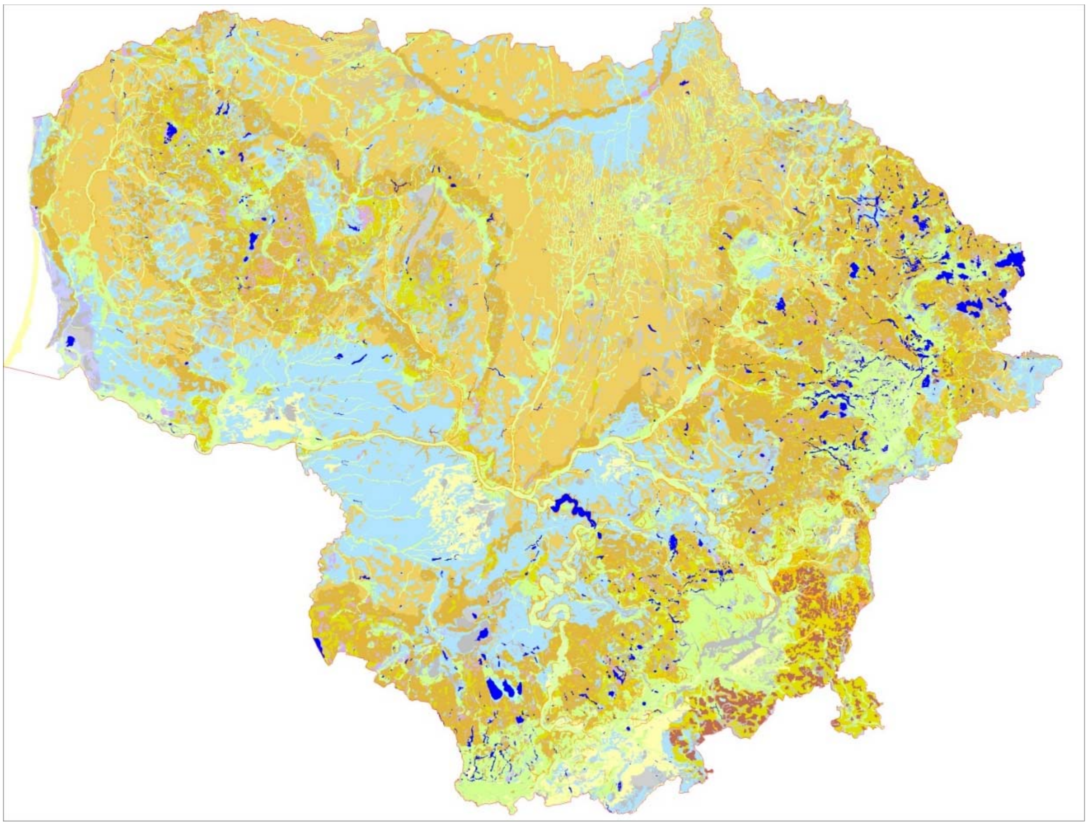
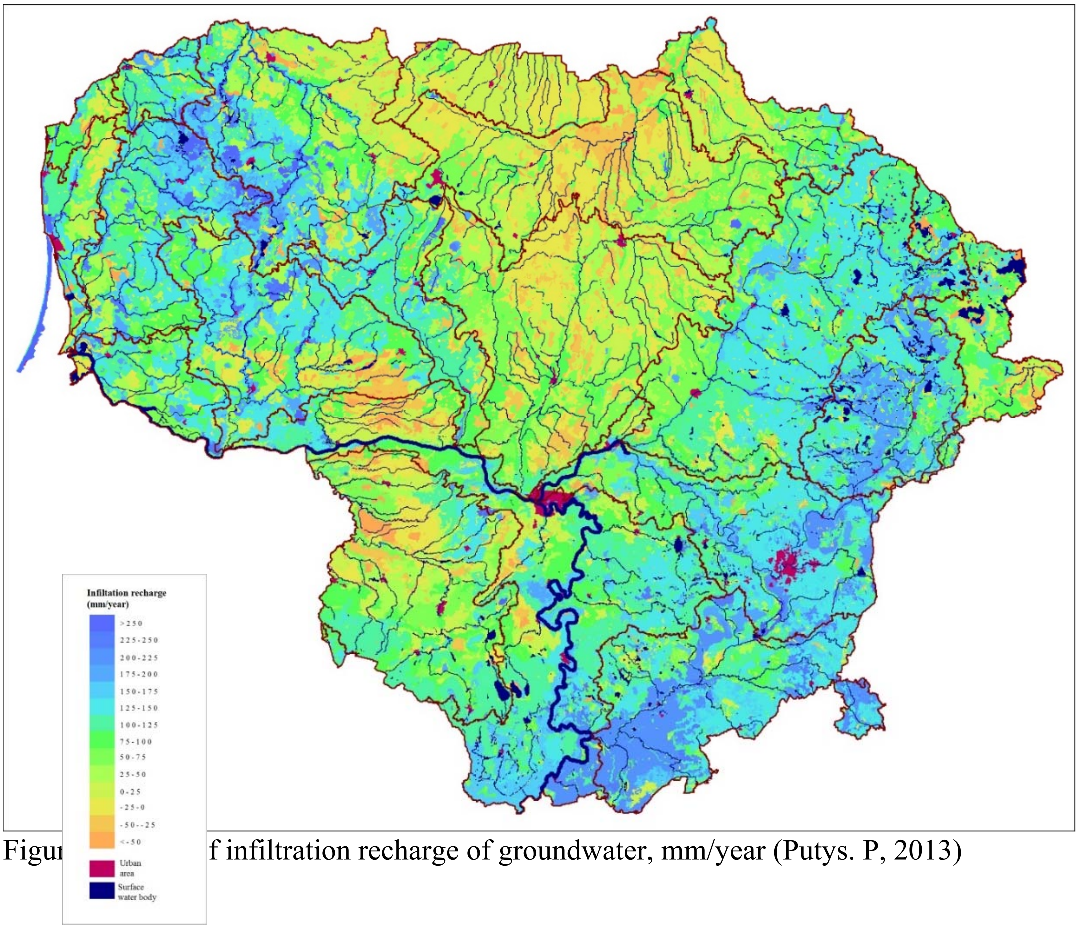


Figure S5. Geological map of Lithuania (Guobytė, 1998)



Figur f infiltration recharge of groundwater, mm/year (Putys. P, 2013)

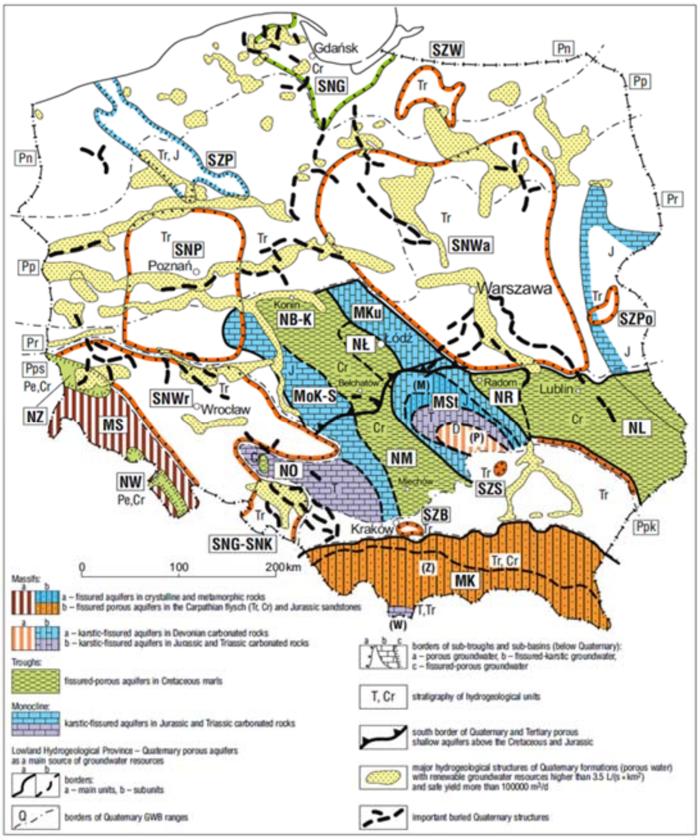


Figure S7 Map of hydrogeological units and structures of Poland (Kleczkowski, 1990)

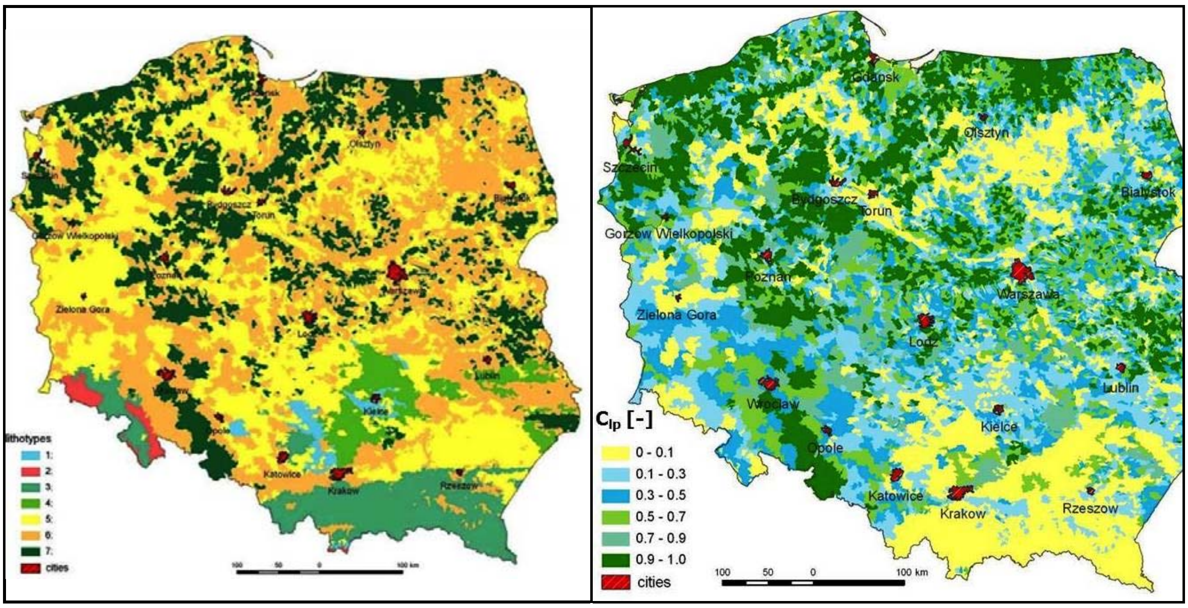


Figure S8. (left panel) Map of dominating lithologies in Poland with class number as in Table 2 and (right panel) contribution of semi- and low permeable sediments (class 7 of lithologies) in the aquifer cover (C_{lp}) (from Witczak, 2011)

5. References

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Appendix C

Summary of upscaling approach for surface water

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Technical Note

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KTH/SMHI summary of upscaling approach for surface water

1. Designing and quantifying effects of remediation actions in streams

There is a set of various remediation actions that can be considered for stream restoration projects:

I) Riffle and pool structures
II) Overflow dams
III) Meanders
IV) New substrate
V) New vegetation

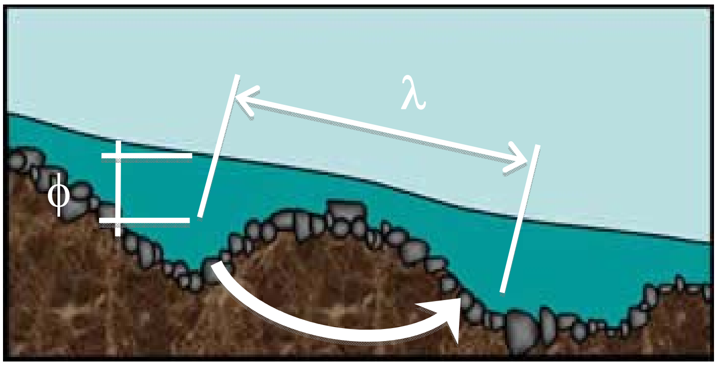

Fig 1. Schematic of how a remediation action, here a riffle-and-pool sequence, is characterized by a wavelength and a hydraulic head loss. The exact head loss over the feature has to be estimated from detailed analyses. The white arrow indicates hyporheic flows through the subsurface that offer retention and decay of nutrients.

Most of the above features can be described in terms of a spatial extension, i.e. a wavelength, λ (m) and a hydraulic head loss ϕ (m). Such linkage of remediation actions and geometric parameters is key for design estimates and can be done in various ways not described in detail here. One approach is to use a series of head loss functions to represent each single remediation feature or a natural stream-bed topography. Details of this approach is described by Morén et al. (2016). However, a simple approach is to use a single harmonic function as a schematic representation of different stream features. Hence, the stream features are represented by hydraulic head loss amplitude ϕ_m (m) and its wavelength λ (m). However, the allocation of head loss over a remediation feature is limited by the overall available head drop along the stream, which can be expressed by the inequality

$$\frac{2}{\pi}\frac{\phi_m}{\lambda} < S_b \tag{1}$$

where S_b = slope of stream By following this simple approach for a specific feature i the average exchange velocity W (m/s) with storage zones over the wavelength can be expressed as (Boano et al., 2014).

$$W_i = \gamma_i K_i \frac{\phi_{m,i}}{\lambda_i} \tag{2}$$

where γ = geometric factor that is feature specific and K = hydraulic conductivity (m/s) . It can be shown that the mean residence time is given by $T_i = 2\varepsilon / W_i$ under assumption of an impermeable surface located at a depth ε (m). The hydraulic head loss can be either of dynamic or static nature, but regardless of this nature there is a total available head loss for a reach ϕ_{Total} that can be distributed either on in-stream friction losses or used as subsurface flow friction losses, hence, driving the hyporheic exchange. Furthermore, by linking the remediation action to exchange velocity W and residence time T of the transient storage zone it is possible to express the total travel time τ for solutes on a stream reach on the form (Morén et al., 2016)

$$\tau_{N,P} = \frac{x}{u}\left(1 + WT\frac{P}{A}\frac{1+K_{sz}}{1+K_{mc}}R_{sz}\right) \tag{3}$$

in which x = travel distance in stream (m), u = stream mean velocity (m/s), P = wetted perimeter (m), A = cross sectional area of stream channel (m^2), K_{sz} = sorption partition coefficient for bed [-], K_{mc} = sorption partition coefficient for main channel [-], R_{sz} = reaction rate factor for the storage zone. From Eqn. (3) one can see that if there is no exchange with a transient storage zone the travel time for solutes in a stream is given by x/u. Thus, the second term within parenthesis of Eqn. (3) acts as a form of retardation factor reflecting the hydrodynamic exchange with the storage zone and biochemical reactions, such as sorption and decay. For solutes subjected to decay a relevant measure is the percentage mass decay, D (Morén et al., 2016):

$$D = 1 - \exp\left[-\frac{x}{u}\left(r_{mc} + \frac{W}{1+K_{mc}}\frac{P}{A}(R_{sz}-1)\right)\right] \tag{4}$$

2. Quantifying multiple restoration sites in a stream network

To quantify the effects of remediation actions over a sub-watershed it is essential to consider the source distribution of the solutes that is subject to the analysis, such as phosphorus and nitrogen, as well as the distribution of the remediation action and general stream character. Riml and Wörman (2011) showed how the effluence export of solute mass can be expressed as an integration over a load weighted PDF of the transport distances in the watershed *G(X) = Γ(X)W(X)*, where *Γ(X)* is a dimensionless distribution of load over the network and *W(X)* is the width function defined as the probability density function (PDF) of the transport distances (m^{-1}). This approach leads to a formulation of the average travel time $\langle \tau_{N,P} \rangle$ and the average percentage mass decay $\langle D \rangle$ over the stream network. The parameterization of the HYPE model requires that these two entities are identical in both model concepts (Riml and Wörman, 2010).

3. Model functions in HYPE that account for retention and decay

HYPE is a semi-distributed model with spatially delineated sub-basins and hydrological response units (HRU) within each sub-basin (Arheimer et al., 2011 a and b). Fluxes are calculated in discrete time steps. Along with water fluxes, the model represents the inorganic (IN) and organic (ON) forms of nitrogen and particulate and soluble phosphorus (PP, SP). Streams in the conceptual framework of the HYPE model are divided into local streams and main streams. Local streams receive all drainage from the sub-basin's land surfaces. Main streams receive all water flows from local streams and upstream sub-basins, and the model routes the combined flow to the next downstream sub-basin. Due to the non-delineated HRU concept streams are not further divided into stream segments, and drainage fluxes fill the local stream storage instantaneously during each time step under the assumption of complete mixing.

In-stream travel time of solutes in HYPE can be derived from water course volume and discharge flux. The water volume at each time step is defined by incoming and existing volumes. Water volumes in streams are routed to sub-basin outlets with a combined translation and attenuation routine. Translation follows a plug flow movement of incoming fluxes to future days, while attenuation is achieved by routing through a linear storage. A parameter *damp* (-) is used to weigh between translation and attenuation. With damp > 0, attenuation is activated, and translation time is reduced the overall routing extended by the attenuation routine which receives input from the translation output.

Translation time (d) is calculated as:

$$translation = (1 - damp)\frac{rivvel}{rivlen * 86400}, (d) \tag{5}$$

An average river velocity *rivvel* (m/s) is given as a constant model parameter, whereas stream lengths *rivlen* (m) is treated individually for local and main streams: *rivlen* of local streams are implemented as square root of the sub-basin area in HYPE, while *rivlen* of main streams are provided as sub-basin-specific estimates. Fractional *translation* times are used as weights to distribute over two target days. The total flow time will be the sum of the translation times for the local and main streams.

Attenuation (d) is calculated as *damp*-weighted complement to translation routing:

$$attenuation = damp \frac{rivvel}{rivlen * 86400}, (d) \quad (6)$$

and the resulting delay is used to construct a linear storage recession coefficient *rc* for flow attenuation of accumulated translation output fluxes by:

$$rc = 1 - attenuation + attenuation\, e^{(1/attenuation)}$$

This attenuation recession provides an approximation of the overall routing delay time (*translation + attenuation*) for the median of fluxes through the routing routine.

In order to provide additional mixing volume for solute turnover processes, a dead mixing volume V_D in the attenuation box or translation-only fluxes can be introduced with an additional parameter *dead*:

$$V_D = dead * uparea * rivlen \ (m^3) \quad (7)$$

Dead mixing volume is implemented as function of upstream area *uparea* and stream length, *dead* (-) is a linear scaling parameter for this relationship. A fixed width-depth relationship of 10:1 is assumed in all calculations to derive storage depth.

Mean travel time τ (s) in rivers is then described by a combination of translation and attenuation time through river volume V_R including dead mixing volume V_D:

$$\tau = (rivlen / rivvel) * (V_R + V_D)/V_R \quad (8)$$

This expression can be used in combination with Eqn. (3), requiring the same mean travel time, to facilitate a parameter translation between the two models. In this purpose we can also use that the mean travel time τ = rivlen / rivvel = 86400 (attenuation + translation) (s). In BaltHYPE, to be used in Soils2Sea for Baltic Sea basin estimates, translation plays only a minor role, and nearly all stream routing is performed by the attenuation routine. Constructed stream remediation measures as listed above would most likely affect local streams in the HYPE concept. Full stream travel time within a sub-basin is described by the sum of local and main stream travel times.

The parameter translation will be based also by requiring the same solute mass decay between the two models, which is important for non-reversible reactions such as denitrification. Nitrogen and phosphorus are transported along with water fluxes, and denitrification, primary production and mineralization are implemented as turnover processes.

Sedimentation and re-suspension from bottom sediments are modelled for particulate phosphorus (PP). All nutrient turnover processes use flow velocity and water course width and depth estimates, which are derived from parameterised empirical relationships based on current flow and 365 day rolling mean flow (see http://www.smhi.net/hype/wiki/doku.php?id=start:hype_model_description:hype_np_riv_lake#common_things_in_lakes_and_rivers). River width estimates are limited by the routing routine width estimate (lower) and a parameter *maxwidth* (upper). The resulting estimates are time-dynamic, and higher flows give increased velocity, width, and depth.

Water temperatures T_w are estimated as a low-pass filter based on air temperatures T_{air} with a weighted updating function:

$$T_{w,i} = \frac{20T_{w,i-1} + T_{air,i}}{21} \tag{9}$$

Denitrification affects inorganic nitrogen solute concentration in the stream water (c_{IN}, mg/l), and is a function of estimated bottom area *barea* *(m²)*, as well as and water temperature through the dimensionless factor *ftmp (-)* that is a function of T_w . A calibration parameter *denit* (kg/(day m²) allows for adjusting the rate of denitrification in units (kg/day).

$$denitri = \min\begin{cases} maxdenitr * pool_{IN} \\ denit * barea * fconc * ftmp \end{cases} \tag{10}$$

w<0

$$fconc = \frac{c_{IN}}{c_{IN} + halfsatIN} \tag{11}$$

where the dimensionless factor *fconc* (-) accounts for a non-linear reaction rate and the half saturation parameter *halfsatINwater* is a constant set to 1.5 mg/L. Hence, fconc is limited to 1 for large c_{IN} and tends to c_{IN} /halfsatIN as c_{IN} decreases.

In order to express the percentage mass decay D similar to Eqn. (4), we formulate the mass balance to the stream for the equilibrium case when the inflows to the stream is fully balanced by the outflow driven by advection and denitrification. The outgoing nitrogen fluxes in units (kg/day) are as denitri + c_{IN} Q, where Q is the discharge in the stream (m³/s). The discharge is generated by the sum of several sources from the upstream part of HYPE: $Q = Q_{S1} + Q_{S2} + Q_{S3} + Q_{MD} + Q_S$ (m³/s), where subscripts S1, S2 and S3 denotes various soil layers (inflow through groundwater), subscript MD denotes drainage pipe inflow and subscript S denotes surface water runoff. In each water source there is a specific nitrogen concentration (cIN_{S1}, cIN_{s3}, etc. in units (mg/L). This means that the nitrogen flux balance to the stream under assumption of equilibrium (i.e. no storage in the stream) and the lower case of Eqn. (10) can be written as

$$Q\, cIN_{\mathrm{inflow}} = \frac{denitri\, 10^6}{86400} + Q\, c_{IN} \qquad (12)$$

where η denotes the fraction of the individual flow of the total flow and cIN_{inflow} is the flow weighted average of the inflow concentrations. The percentage mass decay can , thus, be expressed from the ratio of the nitrogen inflow (left-hand side of Eqn. (12)) and the denitrification flux

$$D = \frac{denitri\, 10^6}{86400\, Q\, cIN_{\mathrm{inflow}}} \qquad (13)$$

For the case of small nitrogen concentration in the stream (fconc = c_{IN} /halfsatIN) Eqn. (13) becomes

$$D = \frac{1}{0.0864} \frac{denit}{Q\, cIN_{\mathrm{inflow}}} \frac{cIN}{halfsatIN}\, barea\ ftmp \qquad (14)$$

This equation can be directly compared with Eqn. (4) as a basis for parameter translation between the two model approaches.

Water temperatures limit denitrification along two threshold values. Denitrification is also influenced by IN saturation, and works less efficiently at lower IN concentrations c_{IN}. This saturation dependency is formulated with a concentration factor *fconc* in the denitrification model, and *fconc* uses a half-saturation constant *halfsatIN* (1.5 mg/l) to limit denitrification at lower IN concentrations. Overall denitrification is limited by a fraction *maxdenitr*, which is currently hard-coded to 50% of the available in-stream IN pool.

Primary production and mineralization acts as source for ON and as a sink for IN and SP. They are functions of water temperature (for details see http://www.smhi.net/hype/wiki/doku.php?id=start:hype_model_description:hype_np_riv_lake#primary_production_and_mineralization).

Sedimentation and re-suspension affect PP in streams, and deposited PP is stored in a sediment pool until it is re-suspended for downstream transport. Sedimentation of re-suspension conditions are derived from the deviation of current flow conditions from a 1-year moving average, and there is a calibration parameter *sedexp* available to tune the relationship.

4. Parametrization of HYPE to quantify solute retention and decay in streams

The effect of remediation actions can be estimated by use of Eqn. (3) and (8), hence, requesting equal mean travel time in both model concepts. This will make it possible to use knowledge of remediation actions in streams on the mean travel time through Eqn. (3) and translate that to model parameters in HYPE. If we take rivlen = x and rivvel = U, one obtains

$$1+\frac{V_D}{V_R}=1+WT\frac{P}{A}\frac{1+K_{sz}}{1+K_{mc}}R_{sz} \tag{15}$$

The approach suggested for such a parameterization is to describe how a previously calibrated parameter values of V_D/V_R change for a changes in parameters on the right-hand side of equation (15) due to a specific remediation action. More specifically it is suggested to evaluate the relative change in the parameter group due to remediation actions by normalizing the equation with the value of the parameter group prevailing before remediation action:

$$\frac{\Delta\left(WT\frac{P}{A}\frac{1+K_{sz}}{1+K_{mc}}R_{sz}\right)}{\left.\left(WT\frac{P}{A}\frac{1+K_{sz}}{1+K_{mc}}R_{sz}\right)\right|_0}=\frac{\Delta\left(\frac{V_D}{V_R}\right)}{\left.\left(\frac{V_D}{V_R}\right)\right|_0} \tag{16}$$

where $(V_D V_R)|_0$ is the originally calibrated value and the left-hand side expresses the percentage change of the stream transport parameters predicted theoretically in the design phase of remediation actions. Similarly, if we use Eqns. (4) and (14) for small nitrogen concentration in the stream (linear case) we can equate

$$\frac{\Delta\exp\left[-\frac{x}{u}\left(r_{mc}+\frac{W}{1+K_{mc}}\frac{P}{A}(R_{sz}-1)\right)\right]}{\left.\exp\left[-\frac{x}{u}\left(r_{mc}+\frac{W}{1+K_{mc}}\frac{P}{A}(R_{sz}-1)\right)\right]\right|_0}=\frac{\Delta\frac{denit}{Q}\frac{barea\ ftmp}{halfsatIN}}{\left.\frac{denit}{Q}\frac{barea\ ftmp}{halfsatIN}\right|_0} \tag{17}$$

Consequently, the relative change of the left-hand sides of Eqns. (16) and (17) are evaluated for specific remediation actions affecting the parameters W and T. This is done as described in section 1. Subsequently, the relative change of the calibrated HYPE parameters can be enforced by use of Eqns. (16) and (17), hence selecting a change of any appropriate single or multiple parameters of their right-hand sides.

5. Baltic basin-scale application

Given the coarse scale of the Baltic Basin model application in combinations with the available data sources for the model set-up, it seems most relevant to focus on changes in local river stream lengths and changes in velocities to alter reaction times. In order to parameterize scenarios of in-stream remediation measures, this requires a compilation of changes of these attributes at the sub-basin scale.

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Appendix D

Lagtimes in unsaturated and saturated zones for Poland, aggregated to HYPE subcatchments

Anna J. Żurek, Kazimierz Różański, Stanisław Witczak
AGH

Technical Note

Lagtimes in unsaturated and saturated zones for Poland, aggregated to HYPE sub-catchments

Anna J. Żurek, Kazimierz Różański, Stanisław Witczak

1. Introduction

Transport of conservative contaminants through groundwater systems (e.g. nitrate under oxidized conditions) is significantly delayed when compared to movement of those contaminants through surface water compartments. As repeatedly demonstrated by environmental tracers (for review see Wachniew et al., 2016), characteristic time scales of groundwater movement may easily reach tens or hundreds of years. This results in large lagtimes of contaminant transport in the subsurface when compared to surface water systems. These lagtimes are particularly important when response of river basins to measures aimed at recovery of good groundwater status is considered. Long lagtimes may confound our ability to make the improved management practices successful in the predefined timeframe and may discourage needed restoration efforts (Sophocleous, 2012). Incorporating lagtime principles into water quality regulations may result in more realistic expectations when such policies are designed and implemented.

Time scales of nutrient losses from soils to inland water and to sea were reviewed by Grimvall et al. (2000). For instance, Stålnacke et al. (2003) found very little evidence for the impact of major changes in agricultural practices resulting, among others, in reduced application of fertilizers, on the riverine concentrations of nitrogen in Latvia rivers. This is not surprising in the light of the above-indicated lagtime concept. For rivers in which groundwater-controlled baseflow constitute a significant portion of the total river discharge, the arrival times of contaminants can be orders of magnitude larger when compared to time scales of contaminant transport associated with surface runoff. For the two largest rivers in Poland discharging to the Baltic Sea (Vistula and Odra) the contribution of groundwater-controlled baseflow to the total discharge is in the order of 50% (Kleczkowski, 2001).

One of important milestones of Soils2Sea project is upscaling of the methodologies developed for assessing transport of nitrates in the studied catchments (Kocinka, Norsminde) to the scale of entire Baltic Sea basin. It is envisaged that this upscaling will be done in the project with the aid of HYPE model. The HYPE model is based on balance of water and nutrients (N, P) and does not incorporate physically-based modelling of flow and transport processes in the subsurface. Thus, there is a need for proper accounting for the above-discussed lagtime in the upscaling scheme based on HYPE. Kocinka catchment will serve as a test ground for probing different scenarios of such accounting.

2. Lagtime concept

The lagtime of contaminant transport in the subsurface with respect to transport through surface and near-surface (drainage) runoff can be separated into two components: (i) the delay associated with travel time of water (and contaminants) through the unsaturated zone, and (ii) the delay linked to time scales of groundwater flow, from the recharge area down to the discharge zone (river). Thus, the travel time of water through unsaturated and saturated zones can be considered a quantitative measure of the lagtime. Figure 1 illustrates this concept.

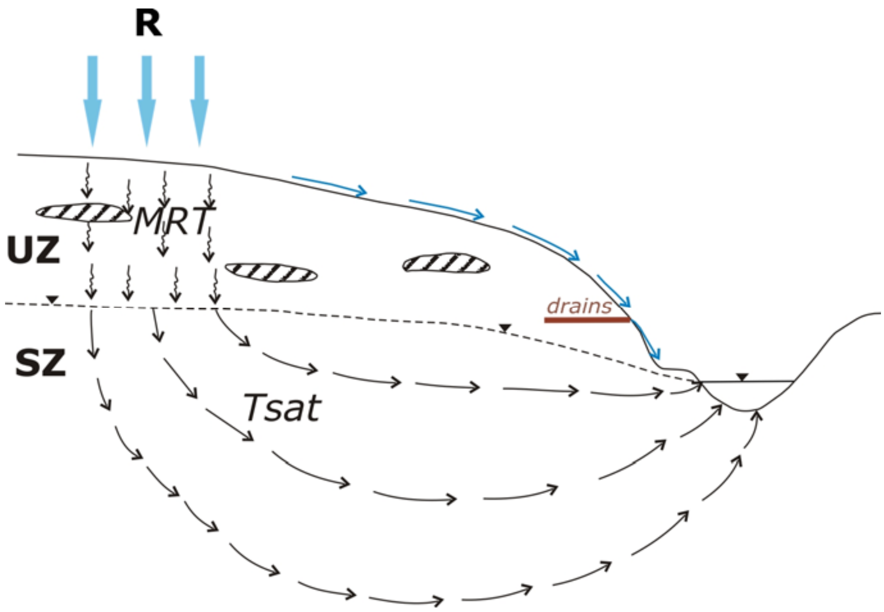

Fig. 1 Concept of lagtime in unsaturated (UZ) and saturated zone (SZ)

2.1. Lagtime in the unsaturated zone

Lagtime in the unsaturated zone on the territory of Poland was assessed on the basis of the existing Groundwater Vulnerability Map of Poland (GVMP) (Witczak et al., 2007; 2011). For preparation of the 1:500,000 GVMP, a GIS-based operational approach relying on evaluation of time scales of water movement was adopted (Wachniew et al., 2016). The GVMP illustrates intrinsic vulnerability of shallow groundwater systems in Poland to conservative pollutants. The adopted approach relies on MRT (Mean Residence Time) of water in the strata separating the saturated aquifer from the land surface, as an integrated vulnerability index. In the framework of GVMP, the MRT is calculated as turnover time of the infiltrating water in the vadose zone. The piston-flow type of water movement through the unsaturated zone is considered. In this approximation, the MRT is the sum of partial MRTs calculated as the ratio of the water present in the given layer of the unsaturated zone to the mean annual recharge:

$$MRT = \sum_{i=1}^{n} \frac{z_i \cdot \theta_i}{R}$$

where R [L/T] is the direct groundwater recharge rate, z_i [L] is the thickness of the given layer in the unsaturated zone, and θ_i [L^3/L^3] is the mean volumetric water content in this layer.

Appropriate calculations were performed for each individual pixel on the map corresponding to one hectare (100m x 100m). Five information layers were used to calculate the MRT: (i) volumetric water content of the soil profile down to 1.5 m depth, (ii) groundwater recharge, (iii) depth to the water table, (iv) volumetric water content of dominating lithotypes of rocks present in the vadose zone below the root zone, (v) contribution of low-permeability rocks in the vadose zone profile (Witczak et al., 2007; 2011; Wachniew et al. 2016). The resulting map of MRT (lagtime in unsaturated zone) is shown in Fig. 2.

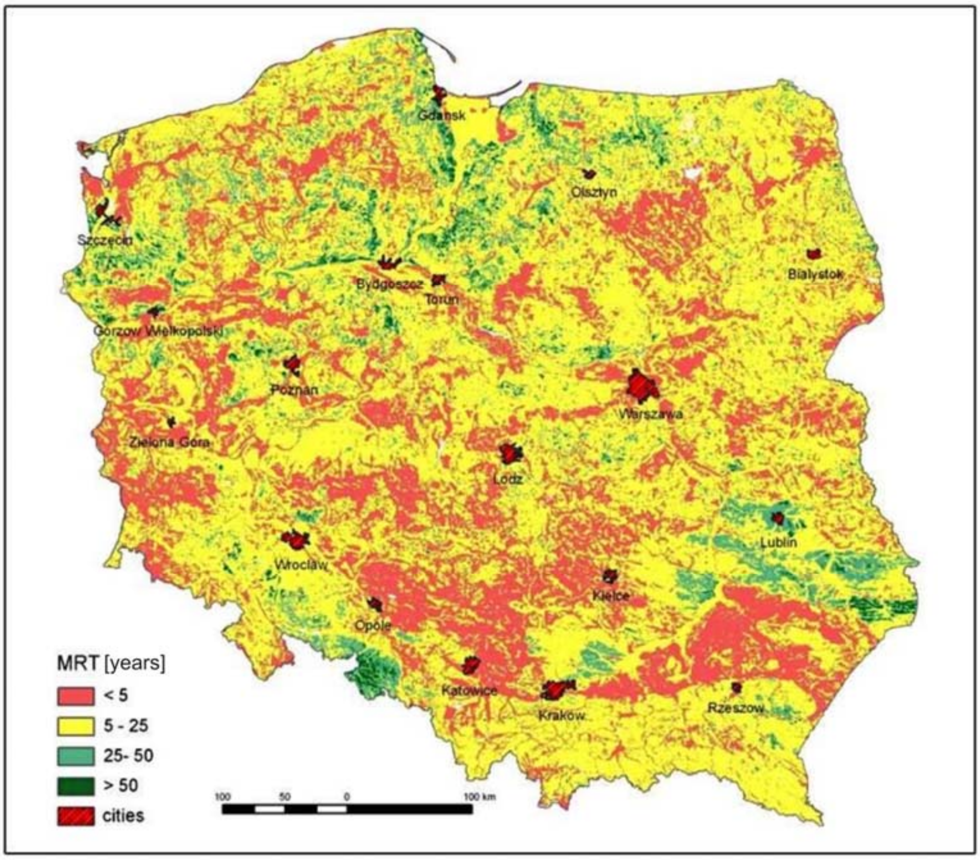

Fig. 2 Mean Residence Time (MRT) of water in the unsaturated zone, calculated for the territory of Poland (resolution 100m x 100m), reflecting lagtime of contaminant transport in this zone (acc. Witczak et al. 2011)

For upscaling the lagtime values in the unsaturated zone (resolution of 100mx100m) the aggregation procedure for each of 1044 HYPE catchments in Poland represented in GIS was performed. The median value of lagtime distribution within each Hype catchment, based on MRT values calculated for each pixel, was considered as representative for this catchment. Example of this upscaling procedure adopted to each HYPE catchments in Poland is presented in Tab. 1 for Kocinka catchment (HYPE ID 9001322). The aggregated MRT values in the unsaturated zone, calculated for all HYPE catchments in Poland are presented on the map in Fig. 3.

Table 1. Aggregated MRT values for Kocinka catchment (HYPE catchment ID 9001322)

HYPE catchment ID	Number of pixels	Catchment area [km^2]	Min MRT [years]	Max MRT [years]	Range [years]	Mean of MRT [years]	STD* [years]	Median [years]
9001322	26046	260.46	1	40	39	7.8	5.7	6

* - standard deviation of a normal distribution

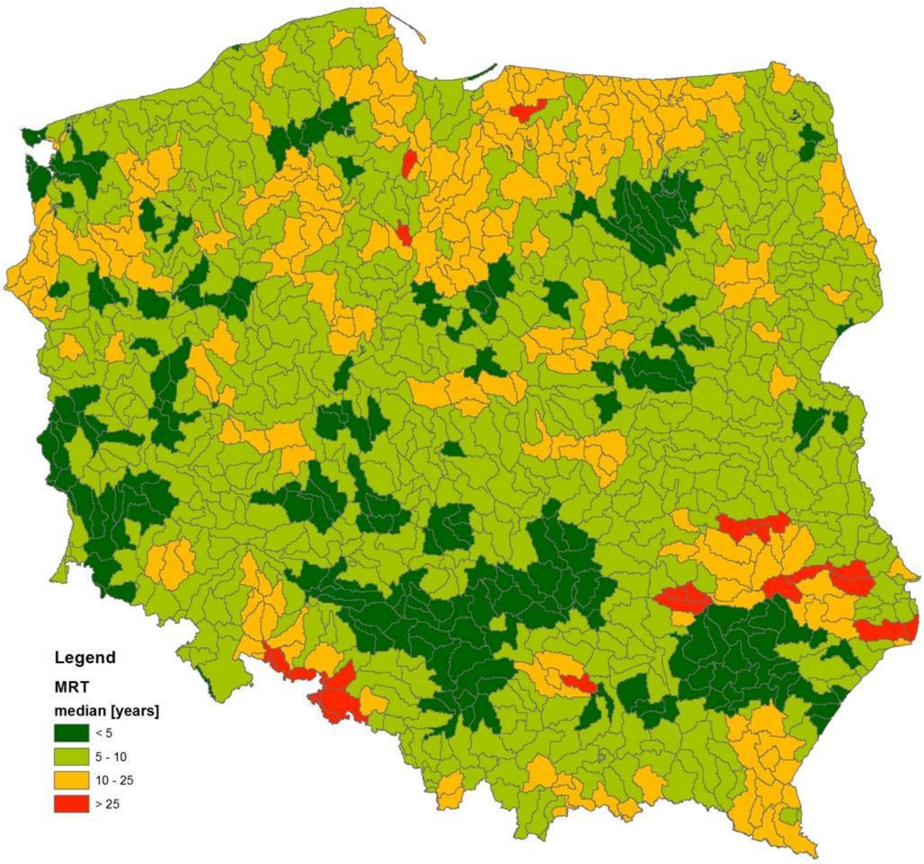

Fig 3 MRT classes (in years) in the unsaturated zone for the territory of Poland, aggregated to HYPE catchments.

2.2. Lagtime in the saturated zone

The lagtime in the saturated zone can be approximated by travel time of water labeled as T_{sat} in Fig. 1. This parameter was derived for the entire territory of Poland using the following operational approach.

T_{sat} was assumed to be equal the travel time of water in the saturated zone, flowing along the local hydraulic gradient, from hillslope to the closest river.

$$T_{sat} = \frac{L}{U}$$

where:

L - distance to the closest river within catchment area higher than 60km^2 [L]

U - groundwater velocity (Darcy velocity) at a representative observation well for which values of k, J and n_a are available [L/T]:

$$U = \frac{k\,J}{n_a}$$

where k stands for hydraulic conductivity [L/T], J signifies gradient and n_a is porosity. The catchment area > 60km^2 is similar to catchment size considered in continental-scale hydrology and water quality model for Europe (Abbaspour et al., 2015).

Representative directions and T_{sat} values for groundwater flow in the saturated zone, between the recharge areas and the drainage areas (rivers, lakes) are visualized on the GVMP by a system of arrows (Fig. 4). The individual numbers next to the arrows reflect horizontal travel time of water in years through saturated zone over the distance corresponding to the length of the arrows (3 km) (Witczak et al., 2007; 2011; Wachniew et al. 2016).

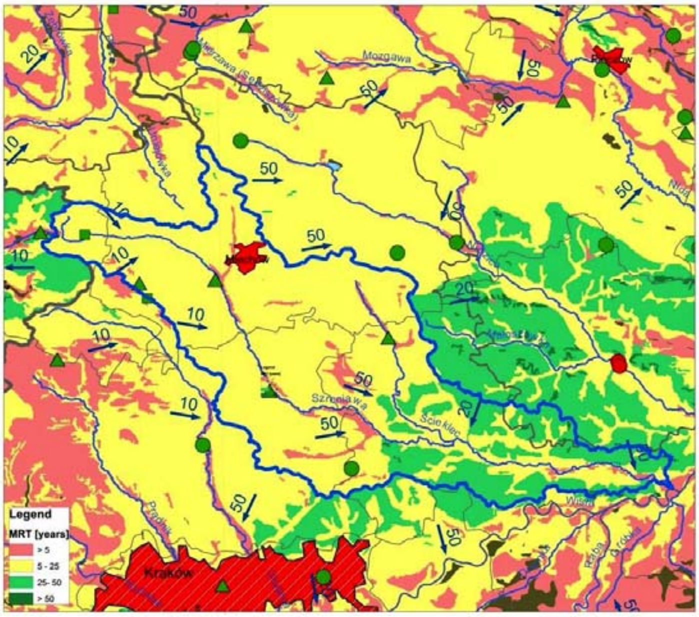

Fig. 4 Excerpt from groundwater vulnerability map of Poland (Witczak et al., 2011; modified) with the Szreniawa river catchment marked with bright blue line. Colors represent MRT through the unsaturated zone. Arrows with individual labels reflect horizontal travel time of water (in years) through the saturated zone over the distance corresponding to the length of the arrows (3 km). The map provides estimates of lagtimes associated with transport of conservative contaminants through the saturated zone and their appearance in surface water of a given catchment (Wachniew et al., 2016)

As in the case of MRT, the T_{sat} values were first calculated for each pixel on the map. Then, median values were derived for each HYPE catchment. Example of this upscaling procedure adopted to each HYPE catchments in Poland is presented in Tab. 2 for Kocinka catchment

(HYPE ID 9001322). The aggregated T_{sat} values in the unsaturated zone, calculated for all HYPE catchments in Poland are presented on the map in Fig. 5.

Table 2 Aggregated T_{sat} values for Kocinka catchment (HYPE catchment ID 9001322)

HYPE catchment ID	Number of pixels	Catchment area [km2]	Min Tsat [years]	Max Tsat [years]	Range [years]	Mean of Tsat [years]	STD* [years]	Median [years]
9001322	26046	260.46	0	67	67	7.6	8.5	6

* - standard deviation of a normal distribution

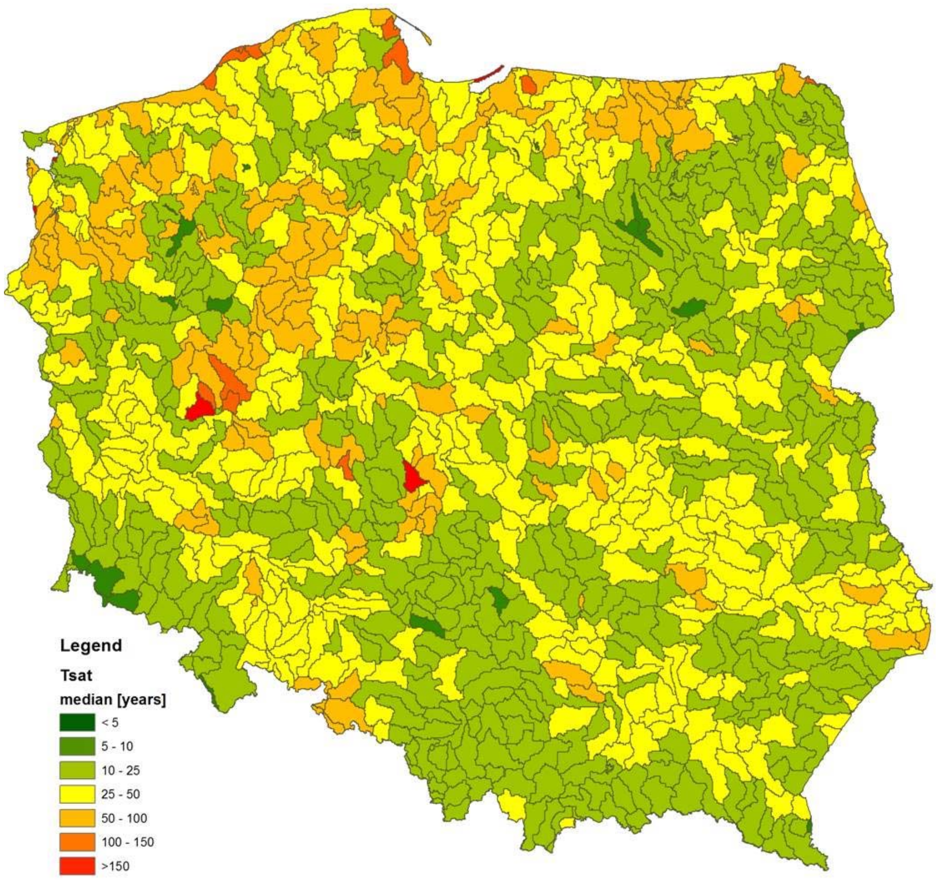

Fig. 5 Classes of T_{sat} (in years) in the saturated zone for entire territory of Poland, aggregated to HYPE catchments.

2.3. Response of river systems to pollutant load on the catchment area

To quantify the lagtime of river systems with respect to changes in pollutant load (e.g. nitrate) on the catchment area, one should sum up the travel time of water through the unsaturated zone (*MRT*) and the travel time associated with movement of water in the saturated zone (T_{sat}). The map in Fig. 6 shows the total travel time (MRT + Tsat), calculated for each pixel.

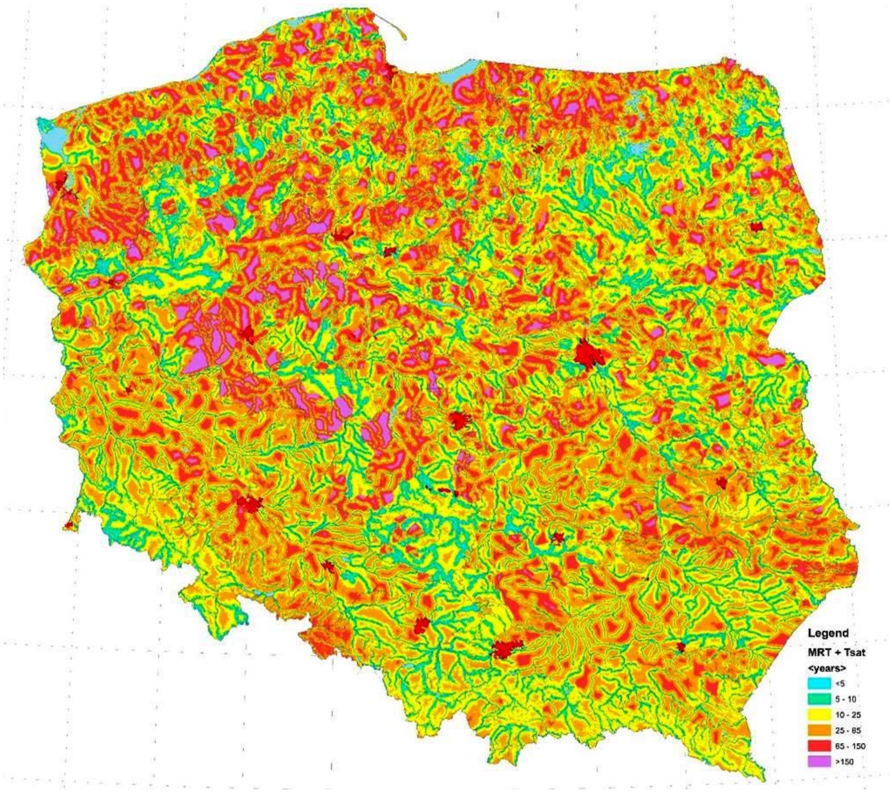

Fig. 6 Lagtime of river systems to changes of pollutant load on the catchment (MRT+T_{sat}) calculated for entire territory of Poland (resolution 100m x 100m).

Preliminary assessments of total lagtime (*MRT + T_{sat}*) based on GVMP methodology suggest that for the territory of Poland the mean values of total lagtime of conservative contaminant is in the order of 25 years, with the range of 10 to 60 years corresponding to one standard deviation (cf. figure below).

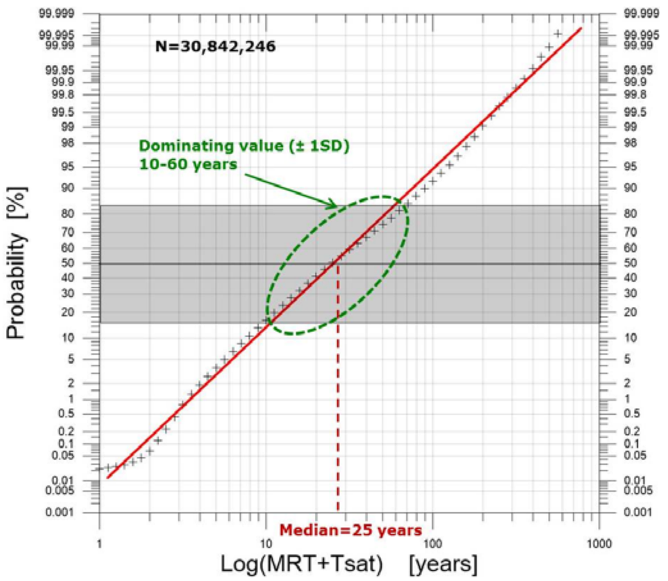

Fig. 7 Log-normal probability distribution of the total lagtime of conservative contaminant arriving in river systems of Poland. The lagtime consists of two components: (i) mean residence time of water in the unsaturated zone (*MRT*), and (ii) transit time of water through the saturated zone (*T_{sat}*).

As in the case of *MRT* and T_{sat} values, the total travel time of water through the subsurface was first calculated for each pixel on the map. Then, the median values were derived for each HYPE catchment. An example of this upscaling procedure adopted to each HYPE catchments in Poland is presented in Tab. 3 for Kocinka catchment (HYPE ID 9001322). The aggregated $MRT + T_{sat}$ values calculated for all HYPE catchments in Poland are presented on the map in Fig. 8.

Table 3 Aggregated $MRT + T_{sat}$ values for Kocinka catchment (HYPE catchment ID 9001322)

HYPE catchment ID	Number of pixels	Catchment area [km2]	Min MRT+Tsat [years]	Max MRT+Tsat [years]	Range [years]	Mean of MRT+Tsat [years]	STD* [years]	Median [years]
9001322	26046	260.46	1	98	97	15.9	11.4	13

* - standard deviation of a normal distribution

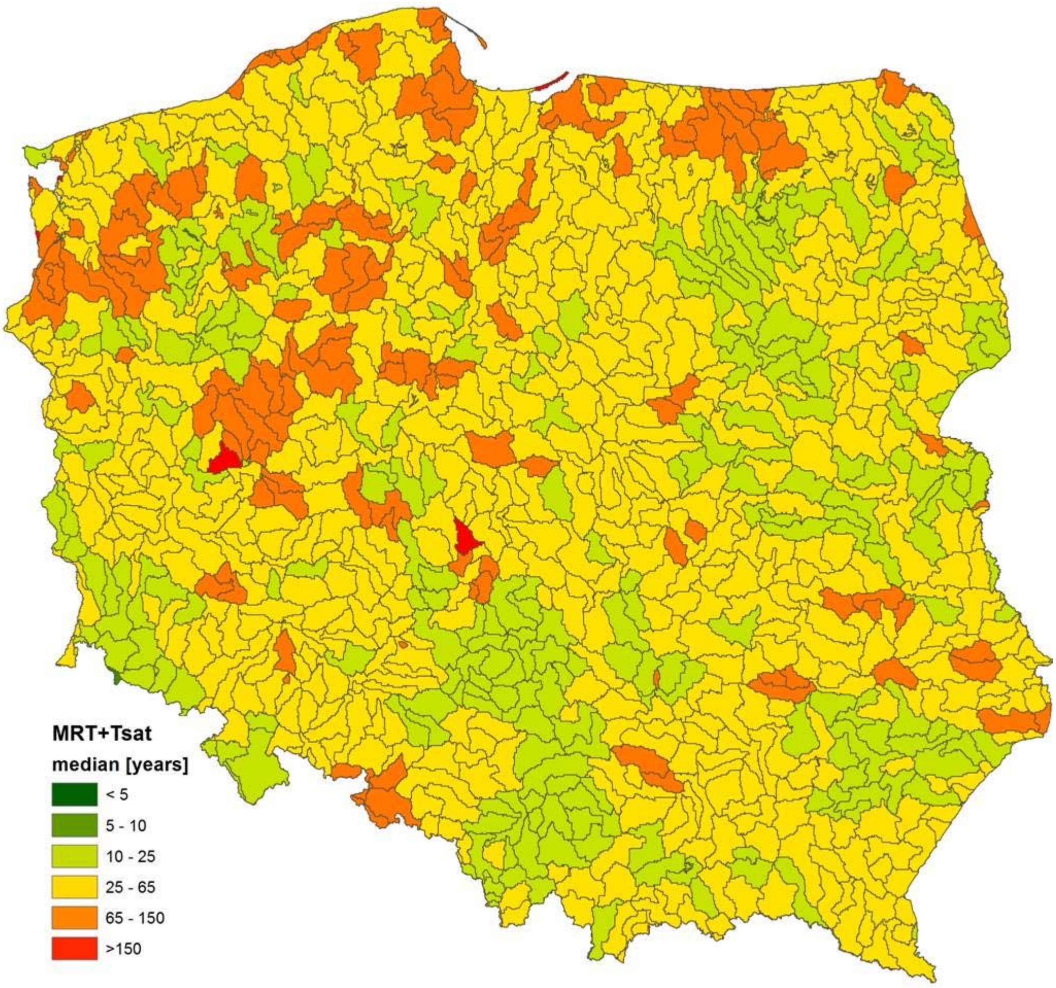

Fig 8. Classes of $MRT + T_{sat}$ values (in years) for Poland entire territory of Poland and aggregated to HYPE catchments.

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